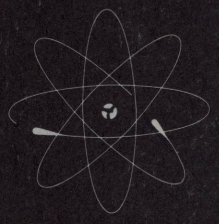
HASL-58

health and safety laboratory

SYMPOSIUM ON OCCUPATIONAL HEALTH EXPERIENCE AND PRACTICES IN THE URANIUM INDUSTRY

Held in New York City, October 15 – 17, 1958
Sponsored by U.S. Atomic Energy Commission Division of Biology and Medicine
and the Health and Safety Laboratory



UNITED STATES ATOMIC ENERGY COMMISSION
NEW YORK OPERATIONS OFFICE

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(Health and Safety)

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PRINTED IN USA
PRICE $4.00

Available from the
Office of Technical Services,
Department of Commerce
Washington 25, D.C.

September 1959 1136 copies

Foreword

S.A. Lough
Health and Safety Laboratory, US AEC, New York, New York

About two years ago the Oak Ridge Operations Office set up a committee to examine working conditions in uranium processing plants under contract to Oak Ridge. Mr. W.B. Harris was a member of this committee as a representative of the Health and Safety Laboratory. Partly as an outgrowth of the report of this committee, which revealed wide differences in health protection practices, the General Manager of the Atomic Energy Commission asked the Health and Safety Laboratory to make a plant survey in connection with this problem. Mr. Harris visited a number of plants operating under AEC contract and conferred with many staff members to get their points of view and general philosophy regarding methods of hazard control. He found major differences in attitudes toward what should be done and how to do it.

Therefore, it seemed reasonable to suppose that those involved would welcome an opportunity to discuss their viewpoints with others who might or might not agree with them. This symposium is the outgrowth of this supposition, and its purpose is to bring together the diverse opinions in the interest of a better general understanding of the problem. The discussions will probably result in the conviction that the problem is complex and can have no simple solution; however, it is hoped that they may serve as a basis for bringing about some uniformity in practice.

In the search for solutions, it must be remembered that those who deal with health and safety have two fundamental responsibilities: not only to control the hazards in the working environment, but also to maintain good production levels.

It is gratifying that many who are familar with controlling the hazards in this industry are present. It is certain that views will be vigorously expressed and that much information will be contributed for consideration. If the exchange of ideas results in each participant taking away more than he brought, the meeting will have been a success.

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Session IV, October 16, Afternoon

Session V, October 17, Morning

Session VI, October 17, Afternoon

Introduction

W.B. Harris
Health and Safety Laboratory, US AEC, New York, New York

A brief description of uranium plants, past and present, may help in explaining the importance of this symposium. Large-scale production of uranium in the United States was begun in about 1942. Between 1942 and 1948, many companies were involved in various aspects, because each segment of production was assigned to a chemical concern with qualifications for performing that particular part of the process. The companies originally involved were the DuPont Company, Linde Air Products Division of Union Carbide, the Electromet Division of Union Carbide, Harshaw Chemical Company, and Mallinckrodt Chemical Works. The total number of employees in the five companies who handled uranium averaged around 300 to 500.

At that time the extraction plants were handling very high grade ores, containing between 40 and 60% uranium, as compared with the ores being used today, which contain about 0.25 to 0.3%. Because the ore was so rich, the hazard of primary interest to the producers was direct radiation due to the high concentration of radium in the ore and to the nature of operations with solutions containing high grade ore. No maximum permissible concentration had been set for uranium. The level generally thought desirable was of the order of 500 $\mu g/m^3$, and maintaining it was not normally considered to be a problem.

This was the state of the art in 1948 when the Health and Safety Laboratory became interested in the problem because of its connection with the New York Operations Office, which had the primary responsibility for uranium production. At that time the DuPont plant had already been closed down. The Linde refinery, which had made UO_3, was also shut down, and all refining operations were concentrated at the Mallinckrodt Chemical Works. The Vitro Chemical Company was operating a scrap reprocessing plant; Harshaw was making UF_4 and UF_6; the Electromet plant was making metal; and the Linde plant was making UF_4. The total employment was about 400.

The Health and Safety Laboratory conducted an industry-wide survey including a thorough investigation of the exposures in each plant. In general, relatively little difference was found between plants. The average exposure was about 5000 $d/m/m^3$, many being higher and many lower. Further information about these estimates of exposure will be found in the paper by A.J. Breslin in Session I.

This average concentration of 5000 $d/m/m^3$ seems to me a conservative estimate of the levels that had existed during the period from 1942 to 1948. The best investigation possible in 1948 failed to reveal any record of a single case of occupational illness during the previous 7-year period.

Since 1948 enormous strides have been made throughout the industry, both in production levels and in the reduction of exposures to uranium. The present level of employment in the production areas is about 5000. In general, each employee is exposed to a concentration of uranium at or below the average permissible levels now in use. It must be emphasized that this improvement has involved a great deal in terms of manpower, production cost, and dollars of plant investment.

So much for history. Now it will be useful to look at the data. Despite the very high past exposures, to date there is still no evidence that anyone in this industry has been made ill as a result of exposure to uranium, i.e., there has been no overt injury that can be traced. In the paper by J.A. Quigley there will be mention of a few cases in which symptoms were found, but these were of short duration and cannot be considered as real injury.

In 1948, when the exposures seemed tremendous and the lack of illness startling, there were people who warned against the premature drawing of any conclusion, on the basis that it takes at least 15 years for the symptoms of radiologic injury

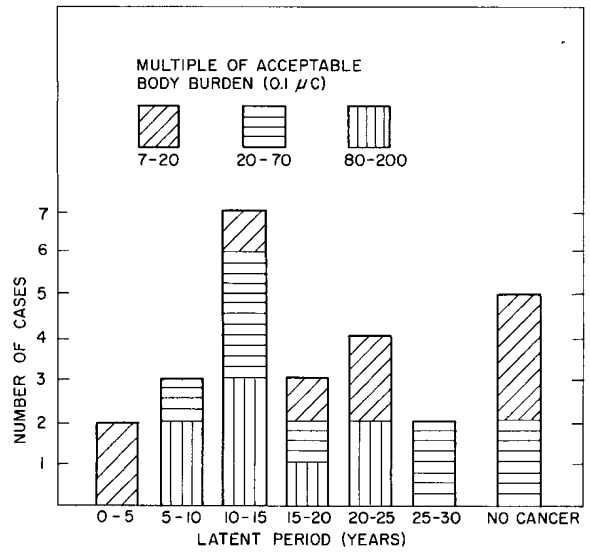

Figure 1. Number of cases of bone cancer diagnosed versus latent period, with an indication of radium exposure in each case. Of 26 persons examined, 21 had cancer. (From AUB, EVANS, HEMPLEMAN AND MARTLAND, Late effects of internally deposited radioactive materials in man, *Medicine* **31**, No. 3, Sept. 1952.)

to develop. While this is probably not completely untrue, it is certainly worth examination. Figure 1 has been plotted from published data on 26 persons exposed to radium of whom 21 developed bone cancer. The distribution of cases looks reasonably normal. The exposures are indicated in terms of multiples of the permissible body burden, which is 0.1 microcurie in the body, and divided into three groups: 7 to 20 times the permissible body burden, 20 to 70, and 80 to 200. The earliest cases, which developed within the first 5-year period, were in persons in the group having the lowest body burdens, and there does not appear to be any significant correlation between body burden and length of time before the incidence of cancer in the cases plotted in Figure 1. The last column represents 5 persons who had not been found to have cancer at the time of investigation, which was approximately 30 years after the initial exposure. The distribution of cases in Figure 1 with respect to exposure and latent period points up an important consideration: when a group of people is exposed to a concentration of radioactive material which is not only potentially but actually carcinogenic, some persons in the group will develop cancer early, and it is not necessary to wait 15 years to be sure that no cancer will develop.

The number of persons exposed to radium in Figure 1 is smaller than the number of persons exposed to uranium in our surveys, and the dosage in terms of calculated roentgens to the lung due to uranium inhalation is probably higher than the dosage due to radium deposits. Conditions in uranium plants have been under study for 15 years. Would it not be reasonable to expect some cancer cases among uranium workers? I merely want to raise this question wthout drawing any conclusions.

The next problem is that of chemical toxicity. In Table 1 are listed percentages of persons showing symptoms when exposed to certain chemicals (lead and mercury) supposedly comparable in toxicity to uranium. It can be seen that in the case of uranium, in contrast to the other two materials, no symptoms were seen. This will be further discussed in the paper by M. Lippmann in Session III.

To give some idea of attitudes toward hazard control, some questions and answers will now be presented. The questions appeared at the end of the report of the plant survey mentioned in the Foreword, and the answers were received from persons responsible for health protection in various plants who have a great deal of experience in the field, who will not be further identified. Some of

Table 1

Incidence of Symptoms

Material	Air concentration range in multiples of MAC	No. of employees studied	% Showing symptoms
Pb[a]	0.4 to 3.2	29	41
Pb[b]	3.0	1	100
Hg[c]	2.0 to 7.5	96	37*
Hg[d]**	0.0 to 0.7	≈500	5
	0.8 to 1.5		9
	1.6 to 2.3		14
	2.4+		23
U (sol)[e]	40 (av)	20	0
U (insol)	100 (av)	14	0

*Includes damage to central nervous system.

**These data were taken at a time when the maximum permissible concentration was twice as high as it is now.

[a]GIEL ET AL., *A.M.A. Arch. Ind. Health* **13**, 321 (1956).

[b]PAGNOTTO ET AL., *Am. Ind. Hyg. Assoc. J.* **19**, 73 (1958).

[c]BENNING, *Ind. Med. and Surg.* **27**, 354 (1958).

[d]C. J. SPIEGL, UR-469, Jan. 10, 1957.

[e]LIPPMANN, this symposium.

the answers have been paraphrased to some extent. It is interesting to see the wide divergence in philosophies despite the fact that the source of the maximum permissible concentration is the same in all cases, namely, the *NBS Handbook*. Again, no conclusions will be drawn, but the attitudes should be of interest.

QUESTION 1: Are we willing to pay the price it costs to keep human exposure at the lowest practicable level in all cases, or does the permissible level as defined by the NCRP contain an adequate factor of safety to insure protection in all cases?

ANSWER 1A: Industry must be willing to pay the cost of reducing the uranium risk to the same level as that acceptable for other types of industrial hazards. From a practical standpoint, it is unreasonable to expect industry to spend more than this. We do not know for sure at this time, but the operating information from this plant would certainly indicate that there is no damage from exposure at the permissible level over a period of 15 years. From medical information available to me, there has been no injury to a single employee in our plant, even though many of these employees have received exposures to air-borne uranium over a period of 15 years, which is equal to, or greater than, a life dose as defined by the NCRP.

ANSWER 1B: The policy should be to spend the requisite money to keep human exposure at the lowest practicable level in those cases where production is the prevailing operation.

ANSWER 1C: I do not believe the NCRP permissible levels contain adequate safety factors to insure protection in all cases. I think we do not have enough human experience to try to base permissible levels on the damage we have observed so far.

ANSWER 1D: The lowest practicable level is a relative condition, which, if interpreted as approaching zero, renders the price of processing radioactive materials prohibitive.

QUESTION 2: What is the significance of the daily rate of uranium excretion in the urine? Is the quarterly or semiannual sample adequate to define body burden, or do the week-to-week fluctuations in excretion rate provide an important index?

ANSWER 2A: Within any 24-hr period, which includes an 8-hr work period, the variation in concentration at each urination is so great as to make the information from a single sample of little value. A quarterly or semiannual sample, taken after an adequate period away from work, gives an indication of the body burden at the time the sample is taken. However, a single quarterly sample has minimal value, and it is not until a history of several exposure periods has been accumulated that any worth-while conclusion can be drawn. I personally feel that the quarterly sample is the one of greatest value in determining body burden, but daily and/or weekly samples may be used for special study purposes.

ANSWER 2B: On the basis of routine air-sample determinations for uranium, we can say that we do not have an air-borne uranium problem. We do routine urinalysis for detection of kidney disease on a periodic basis. At least for our operation, we would regard a quarterly sample adequate to define body burden.

ANSWER 2C: Quarterly or semiannual samples are not adequate to define body burden. I think the week-to-week fluctuations in excretion rate are an index to the environment.

ANSWER 2D: Quarterly or semiannual sampling is certainly adequate for the determination of uranium fixed in the body. A daily uranium excretion rate enables supervisors to detect specific exposures in day-to-day operations. Urine analysis used for this purpose, while more expensive, is a more direct indication of the uranium exposure than air-borne or surface contamination levels.

QUESTION 3: Should an individual be allowed to accept an overexposure for a brief period, or is it necessary to provide him with respiratory protection whenever a situation exists wherein temporary overexposures may be created?

ANSWER 3A: Our evidence indicates that there is no damaging effect as a result of overexposure for a brief period.

ANSWER 3B: In our operation, it is not planned that an individual will accept an overexposure, even for a brief period.

ANSWER 3C: We should accept the recommendations of the authorities and protect our personnel from temporary overexposures. (NOTE: This respondent did not specify what authorities he was referring to.)

ANSWER 3D: There is certainly a finite number of instances when a worker can accept a relatively high exposure for a brief period.

QUESTION 4: What is the maximum atmospheric concentration or quantity of uranium which an individual may breathe before it is considered that he has suffered an injury, however slight; or, is there no such amount?

ANSWER 4A: There is undoubtedly for each human being some concentration over some finite period of time which will produce some degree of injury, but we believe this concentration is orders of magnitude greater than the MAC.

ANSWER 4B: The policy should be to conduct an operation in such a fashion that the maximum allowable concentration, as defined by the NCRP, is not exceeded at any time.

ANSWER 4C: I am willing to accept the opinion of the NCRP.

ANSWER 4D: For the normal interpretation of the word injury, we would expect the concentration to be very high.

QUESTION 5: Inasmuch as uranium is a natural component of the earth's crust and is normally present in the human metabolism, is it reasonable to confine Commission-produced uranium in an absolute manner? It is generally believed, for example, that it is the policy of the Commission that absolutely no radioactive material in measurable quantities should be permitted to escape beyond the site boundary. This applies to materials carried by the feet along the ground, by the clothing into the home, and by the air discharged from plant processes.

ANSWER 5A: We do not know how to design a process for the refining of thousands of tons of uranium per year which would enable the absolute confinement of uranium. Our philosophy, therefore, has been that some minimal escape is unavoidable. We have certainly seen no indication of damage to people or property as a result of our operations.

ANSWER 5B: In practice it has been shown that one can approach absolute confinement with very little extension of the procedures necessary for partial confinement.

ANSWER 5C: Regardless of the source or kind of radioactivity, the Commission should attempt to prevent the escape of measurable quantities of activity beyond the site boundary.

ANSWER 5D: It is impossible to operate a uranium processing facility and prevent measurable quantities escaping beyond the site boundary.

QUESTION 6: Is there any justification, other than economic, for AEC contractors to launder clothing which has become contaminated with uranium? When one considers capital cost, is there an economic justification?

ANSWER 6A: We believe there is some justification for a plant-operated laundry, even though it may not be economical from the standpoint of capital cost or operating costs. We do not feel that health considerations alone provide sufficient justification.

ANSWER 6B: There are justifications other than the economic for AEC contractors to launder clothing which has become contaminated with uranium. One should consider the possibility of an employee getting powdered uranium on his coveralls and, perhaps, creating an undesirable situation at home.

ANSWER 6C: If laundry operators will take proper precautions to control exposure to their employees and patrons, I do not see any justification for AEC contractors to launder their own contaminated clothing.

ANSWER 6D: The extent of clothing contamination, as well as numbers of garments, should govern this situation.

QUESTION 7: Is it possible from air samples to provide an adequate definition of the exposure of operating personnel to air-borne uranium dust or fume? Can such a program of definition be accomplished as economically as a urine sampling program? Conversely, is it possible to define exposure adequately by means of a urine sampling program? If both are necessary, what is the optimum combination?

ANSWER 7A: Air samples can provide an adequate definition of the exposure of operating personnel. A program of air sampling in a uranium refinery is an absolute necessity because it is the key to sources of exposure and corrective action. No urine sampling program will pinpoint the individual sources of exposure. We feel that in areas where uranium is being processed routinely in quantity, there should be a complete dust study of all jobs by the weighted average method at least once each year, and that personnel working on jobs shown by air samples to produce exposures above ½ the MAC should be sampled by the urine method at least once every 3 months.

ANSWER 7B: We very definitely favor air sampling and have proceeded on such a policy.

ANSWER 7C: Exposure could be defined adequately with a daily urine sampling program.

ANSWER 7D: Air samples give a rapid indication of adverse working conditions. Urinalysis is a more direct measure of internal exposure, from ingestion or injection as well as inhalation. 10 CFR 20 is at fault in relying on air concentration as a criterion rather than urinalysis.

QUESTION 8: Has enough human exposure experience been accumulated so that one may use this in preference to the results on animals derived from laboratory exposures? Does this human experience add to or detract from our reliance on the permissible level as stated in the NCRP publications?

ANSWER 8A: I do not believe that human experience to date permits any revision of the MAC, but it does permit a more intelligent interpretation and application of the present limits.

ANSWER 8B: Human exposure experience as reported contributes very little which could either add to or detract from our reliance on permissible levels.

ANSWER 8C: I feel that our human experience data are inadequate.

ANSWER 8D: There is not enough human experience. (NOTE: It is to be hoped that this meeting will add to the information on human experience.)

QUESTION 9: Is there any value in the practice of transferring personnel who are believed to have received exposures to air-borne radioactive materials beyond a predetermined level?

ANSWER 9A: Personnel should be transferred who have received an exposure to air-borne radioactive material such that critical organs have received 50% of the permissible dose from that source. I do not believe that personnel should be transferred on the basis of short-term overexposures. This applies only to the radioactive nature of the materials.

ANSWER 9B: Any employee suspected of being injured from air-borne radioactive materials would be removed from any further uranium exposure, pending determination of his body burden and whether or not he had suffered any kidney damage.

ANSWER 9C: This would depend on the predetermined level.

ANSWER 9D: As long as there is an established MPL for internal contamination, there is no alternative but to remove the individual who exceeds it from further exposure. The true questions are, what constitutes a realistic MPL, and what is to be considered injury in the absence of clinical evidence of damage.

QUESTION 10: In view of the experience accumulated to date on exposure to UO_2F_2, it is still reasonable to assume that the toxicity of soluble uranium compounds should be based on potential kidney damage rather than on long-term radiation damage to the lung?

ANSWER 10A: We are at present of the opinion that any demonstrable damage from exposure to both soluble and insoluble uranium compounds will first appear as kidney damage. Generally speaking, it appears that all particles of uranium-containing material small enough to enter the lung are soluble to some degree. We would like to suggest that there be an extension of research into the radiosensitivity of lung tissue. There are some indications that the lung may be capable of accepting somewhat higher dose of radiation, particularly alpha radiation.

ANSWER 10B: We consider it reasonable to assume that the toxicity of soluble uranium compounds should be based on the chemical effects on the kidney rather than on the long-term radiation damage to the lung.

ANSWERS 10C AND 10D: None received.

It has often been said that, once controls are established, the cost of reducing the permissible level by an order of magnitude is negligible. This is simply not true. Only last week W. McAdams stated that the control of radiation hazards is quite simple if the rules are followed, but a health physicist is required for every 40 employees. How, then, can one industrial hygienist be sufficient in a plant in which 2000 employees handle mercury?

For practical reasons there would be a tremendous value in arriving at an accurate and reasonable definition of control procedures and establishing some degree of uniformity. As time goes on, more and more competition will enter the production picture, and the AEC will be able to apply the cost yardstick. The plant with the least control will have an economic advantage in the competitive situation. Eventually the AEC will step out of production and private industry will take over; again, the producer with the extra-clean plant will be penalized.

On the other hand, consider the problems of labor relations. In contract negotiations, the unions in this industry are tending to act more and more as a single organization. When they become completely unified, they will exert strong pressure for the extension throughout the industry of the highest degree of protection afforded in any present plant. This would create enormous problems. The alternative might be a demand for extra hazard pay, whether or not it is justified. Even at present, when a worker moves from one plant to another, where controls differ, he has a right to question the degree of protection being

provided him, and management must be in a position to give a sound answer.

In conclusion, what is it hoped that this symposium will accomplish?

The participants should consider the facts and relinquish the illusions. From the discussions there should evolve a unanimous agreement on minimum basic standards of health practice, which will be acceptable as sufficient by the AEC, and which should be published. It is not to be expected that significant changes in policy will result at all the various installations, but the level of control will be raised in some cases. Those who maintain standards higher than the required ones should recognize that the basis for this is other than health protection, whether it be pride, advantage in union negotiations, or an aid in attracting personnel.

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SESSION I

A Summary of Fifteen Years of Experience With Dust Problems in the Refining and Fabrication of Uranium

MONT G. MASON
Mallinckrodt Chemical Works, St. Charles, Missouri

This paper concerns chronic exposure to uranium dust in one part of the uranium industry, dealing only with natural uranium, not enriched uranium or uranium hexafluoride. A typical uranium plant consists of a series of chemical and metallurgical processes housed in industrial factory buildings. A new plant went into operation in 1957, covering some 40 acres of ground. The uranium feed material to this plant is a concentrate obtained by upgrading low-grade ores. These feed materials are usually packed into 30 or 55-gal steel drums at the mills and shipped by rail in full carload lots. The input capacity of a conventional plant is in the range of 5000 to 15,000 tons uranium content per year, so that up to 100,000 drums of feed are received per year which must be dumped, sampled, and processed. Figure 1 is a flow sheet for a typical plant.

The problem of controlling uranium dust involves the operation and maintenance of a commercial type of chemical plant with an annual through-put of several thousand tons of dry powders of a radioactive material. The feed material is a dry solid, usually finely divided, which dusts readily during drum dumping and all handling operations. Digestion and extraction are wet processes in which dusting is minimal, but the processing thereafter involves dry chemical reactions so that powdered uranium compounds present dust problems. If it were not for its radioactivity, uranium would fall into the same category as lead and other heavy metals because the

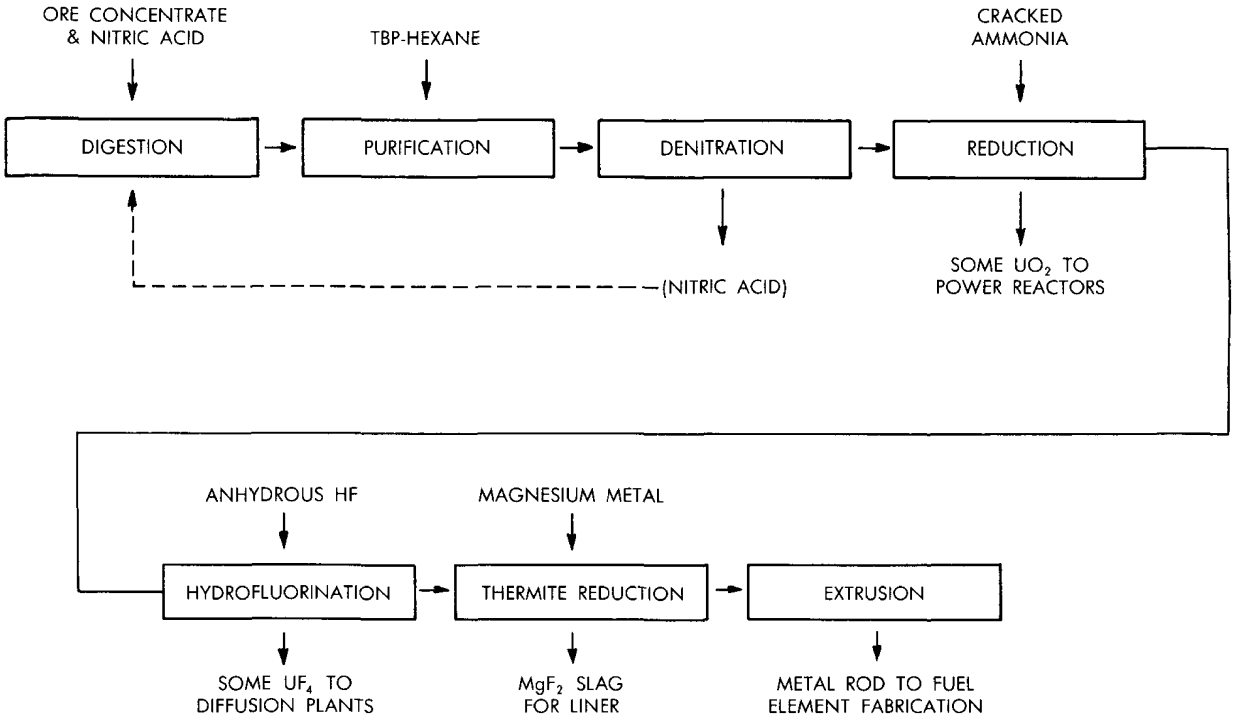

Figure 1. Flow sheet of Weldon Spring processing plant.

Table 1

Plant 6 Uranium Dust Concentration by Years and Process Steps in Multiples of 70 α d/m/m^3
(Production of UO_3 from Ore and Soluble Feed)

				UO_3 production			UO_2 production		
	Whseg.	Ore grinding	Feed digest	Milling	Pot rm.	Pkg.	Load	Unload	Pkg.
1946	3	190	6	180	111		76	45	161
1947	3	195	6	180	111		76	45	161
1948	3	195	6	180	111		76	45	161
1949				180	111				
	1	5	1	0	60	10	20	10	5
1950				*	11				
	0.4	5	1		5	10	10	5	5
1951	0.5	5	2		2	1	6	3	5
1952	0.5	3	5		3	2	6	3	5
1953	0.9	2	0.7		3	2	†	†	†
1954	0.3	2	0.6		2	2			
1955	0.3	*	0.8		2	2			
1956	0.3		0.4		3	4			
1957	0.3		0.8		3	1			

Prior to 1946 above operations done in Building 51; all work transferred to Weldon Spring in March 1957.

*Discontinued.

†Transferred to Plant 7 in October 1952.

chemical toxicity is of about the same order, and chemical toxicity is believed to be the controlling factor; but the radioactivity, although comparatively low, necessitates the added attention to dust control.

The early uranium production facilities in this plant began operating in 1942 and 1943, and, since they were then expected to operate for only 6 to 8 months, extensive dust control equipment was not provided, and in some operations the dust concentrations were considerably higher than present standards. At that time there were essentially no recognized standards for uranium dust exposure. In those early war years, the urgency and the secrecy entailed complications, but it was agreed between Mallinckrodt and officials of the Manhattan District that production would proceed on a priority basis with the understanding that extensive use of respirators would be required for dust protection in high dust areas. It was also agreed that all workers would be under close medical surveillance, and a rigid program of physical examinations was initiated which has been continued to the present time.

All workers have been carefully screened for abnormalities in the urine, blood, or chest, and any abnormal finding has automatically disqualified a man for work in the uranium operations. This has provided an unusually healthy group of employees, and any conclusions drawn from the clinical and exposure data should recognize it as a possible bias. The operating practices and medical control established in the early days have undoubtedly had a significant effect upon present industrial attitudes towards uranium health control.

No regular dust sampling program was in effect during 1943 through 1947, but sufficient samples were collected to show that air-borne uranium concentrations were high by present standards; concentrations of 50 to 100 times the present MAC were not uncommon, and some operations produced concentrations up to 1000 MAC for a few minutes. Note that these are air concentrations, not intake to the lungs.

Operation of this plant was not recognized as being permanent until 1946; therefore, a full-scale health program was not authorized until early 1947. The formal health program got under way early in 1948, as a joint effort between Mallinckrodt and the AEC. One of the first projects of the newly formed Health Department was a thorough

analysis of the dust data already accumulated and the immediate collection of additional data to enable an estimation of dust exposure already received by operation and maintenance personnel.

The history of dust concentration from 1943 through 1957 is presented in Tables 1 to 4. These tables and the graphs that follow show that high levels of exposure can be expected when there is minimal dust control, and that low levels can be achieved by well designed dust controls of the type normally specified for good industrial hygiene practice.

Table 1 presents uranium dust concentrations in a uranium refining plant that went into operation in 1945 and began receiving feed materials in 1946, partly pitchblende. This feed was ground, digested, extracted, and converted into UO_3 by denitrating uranyl nitrate hexahydrate in pots. The UO_3 dry powder was unloaded by hand-scooping from the pots into drums, and was next transferred by hand-scooping from drums into shallow trays, which were inserted into furnaces where the UO_3 was reduced to UO_2 by contacting with hydrogen at a high temperature. The trays of UO_2 were then unloaded by hand into drums. During initial operations dust control was minimal, and it can be seen that air concentrations were high during the period 1946 through 1948, when respirators were required for practically all plant operations. In 1949, under the new health program, immediate steps were taken to install good ventilation and dust control and to initiate process improvements in an effort to achieve sufficiently low levels to dispense with respirators. A major process revision eliminated all hand-scooping by installation of pneumatic unloading and conveying systems. The data show that the changes did result in a marked reduction in air concentrations, but not to the desired level in some parts of the operation.

Table 2

Plant 4 Uranium Dust Concentration by Years and Major Operations in Multiples of 70 α d/m/m^3 (Production of UF_4, KB-2, YM-5)

	UO_2 handling	UF_4 production	KB-2 production	YM-5 production
1943	30	34	17	36
1944	30	34	17	36
1945	30	34	17	36
1946	30	34	17	36
1947	30	34	17	36
1948	30	34	17	36
1949	6	4	4	11
1950	4	2	3	11
1951	4	3		
1952	4	3		

The first two processes were transferred to Plant 7 in 1953; the last two to Plant 6E in 1951.

Table 3

Plant 7 Uranium Dust Concentration by Years and Major Operations in Multiples of 70 α d/m/m^3 (Production of UO_2 and UF_4)

	Average	High	Source of high
1952	0.5	1.6	UO_2 dumping
1953	0.4	1.7	Furnace operation, TA-7 packaging
1954	0.5	7.0	Sampling and cleanup
1955	0.3	1.1	UO_2 dumping
1956	0.4	0.8	UO_2 dumping
1957	0.3	0.8	UO_2 dumping
1958	0.5	1.4	TA-7 packaging

Transferred to Weldon Spring about July 1958.

Table 2 presents information on a plant where the uranium dioxide was converted into uranium tetrafluoride and finally into highly purified massive uranium metal. (This plant was a former lumber sash and door works hastily converted in 1942.) As in the previous plant, there was extensive scooping and manual handling of the uranium materials, which was minimized by mechanization in 1948 and 1949. The data show that dust levels were reduced, but not to the desired level, and in 1949 it became apparent that this plant could not be brought under satisfactory control; therefore, Mallinckrodt and the AEC agreed on the necessity for constructing a new facility consisting of two plants. The new metal plant (Plant 6E) went into operation late in 1950, and the new green salt (UF_4) plant late in 1952.

Table 3 shows an average dust concentration in the new green salt plant (Plant 7) no greater than 25 $\mu g/m^3$, which was within the acceptable levels. This plant was designed with dust control as a primary objective. In addition to well designed ventilation, there were major changes in the processing methods, including new process technology and equipment, all of which contributed to the lowering of concentrations. Comparison of this

Table 4

Plant 6E Uranium Dust Concentrations by Year and Source in Multiples of 70 α d/m/m^3 (Production of KB-2, YM-5)

	KB-2			YM-5		
	Average	High	Source of high	Average	High	Source of high
1950	0.1	0.3	Charging	1.0	1.8	Cruc. asbly.
1951	0.3	0.8	Residue	1.1	1.8	Top furnace
1952	0.4	1.0	Residue	1.2	2.3	Burnout
1953	0.3	1.2	Charging	0.5	0.7	Top furnace
1954	0.5	3.0	Residue	0.9	1.7	Bot. furnace
1955	1.6	4.0	Residue	0.6	1.5	Bot. furnace
1956	0.4	1.6	Capping	0.5	0.6	Bot. furnace
1957	0.4	1.5	Burnout	0.7	1.2	Bot. furnace
1958	0.8	2.1	Breakout	1.2	2.1	Bot. furnace

Transferred to Weldon Spring in March 1958.

table with the first two columns of Table 2 shows that average dust concentrations were reduced by a factor of 60 compared to 1943 figures and by a factor of 8 compared to 1950 figures.

Table 4 shows dust concentration data for the new metal plant, which was also designed with adequate dust control as a primary objective and with manual handling almost entirely eliminated. Success is again evident from comparison of these data with those in the last two columns of Table 2. The reduction factor is about 20 compared to 1943 figures and 8 compared to 1950 figures.

Production rate information is not included in the above tables, but it is of interest that by 1956 all the plants were producing more than three times the original designed capacity. The increase in production rate was almost continuous, so that constant revision of dust controls to maintain acceptable levels was also necessary. By 1957 these plants were producing so far above designed capacity that it was virtually impossible to increase production further without completely losing control over health problems including dust exposures.

The data in the above tables indicate that considerable progress was made towards reducing dust exposure, but it is also apparent that at no time was the uranium so completely contained that it ceased to be a source of contamination to the plant air and to the plant in general. It is also evident that personnel did work in fairly high concentrations in the early days of operation, and that the exposures received depended partly on the effectiveness of the respirator program. Dust concentration data for each of the major dust producing operations have been plotted against time in Figures 2, 3, and 4.

Figure 4 presents information on the manual handling step, which is the production of orange oxide (UO_3). In addition to dust concentrations, this graph shows relative production rates, notations about actions taken to reduce concentrations, and some cost information. It is apparent that dust control is expensive and that this type of control alone is not adequate for manual handling operations. Operations requiring the worker to come into contact with the dry powders must be eliminated if adequate dust control is to be achieved; i.e., it is necessary to develop new production technology. This particular problem has been studied intensively, but to date no satisfactory production method including both adequate dust control and an acceptable product has been developed, and orange oxide production continues to present a troublesome dust problem.

PHILOSOPHY

Much of the present attitude towards the health aspects of uranium is the direct result of early operations. Because uranium is a radioactive material it falls within the broad scope of

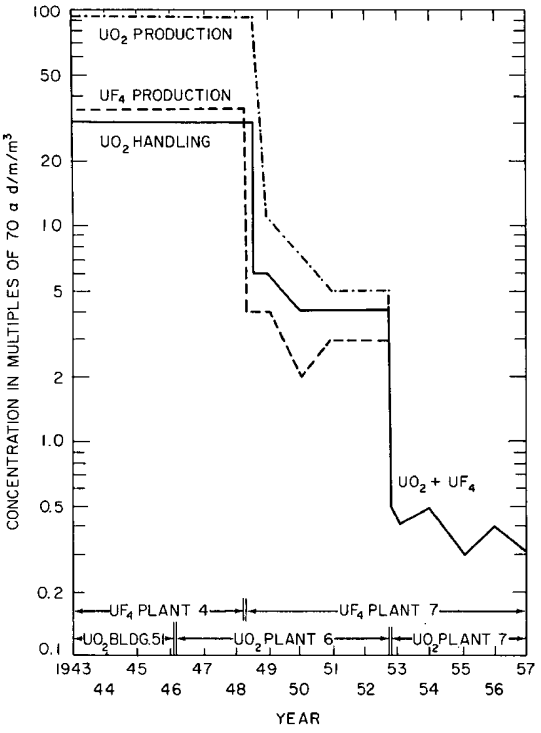

Figure 2. Dust concentration versus years; UO_2 and UF_4 through plant changes.

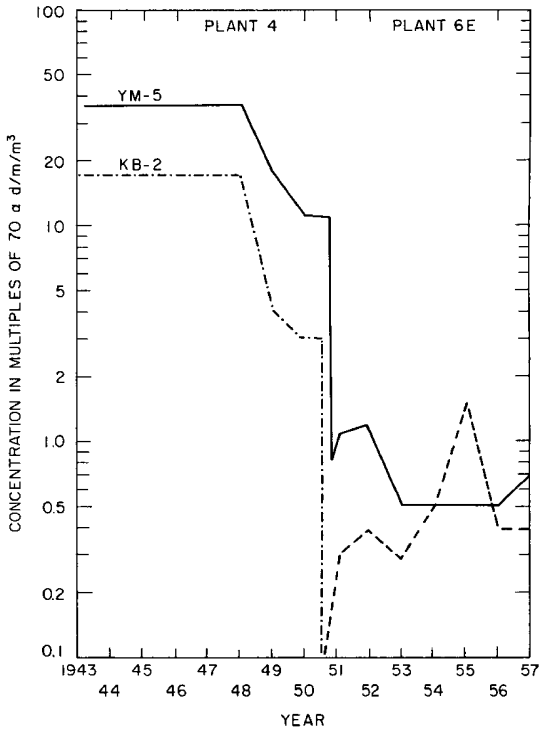

Figure 3. Dust concentration versus years; KB-2 and YM-5 from Plant 4 through Plant 6E.

the various handbooks and regulations aimed at controlling radiation and radioactive materials. Those working with the uranium health problem are frequently under fire from two directions. On the one hand, we are criticized if we do not apply the same philosophy of control as that applied, for example, to plutonium; on the other hand, we may be criticized from other quarters for spending too much money on our control programs because uranium is less radioactive.

The history of the Mallinckrodt uranium operations certainly contains all the elements of this problem. The Mallinckrodt management has been most insistent that a high level of health protection be achieved and maintained. Its ultimate goal for health protection is to limit exposures to no greater than 10% of the permissible levels. The AEC has consistently worked with Mallinckrodt towards this objective, but both the contractor and the AEC have been faced with the extreme cost of completely redesigning and rebuilding uranium production facilities to achieve it. To accomplish the degree of control exercised with other radioactive materials such as plutonium, it would be necessary to employ completely new production

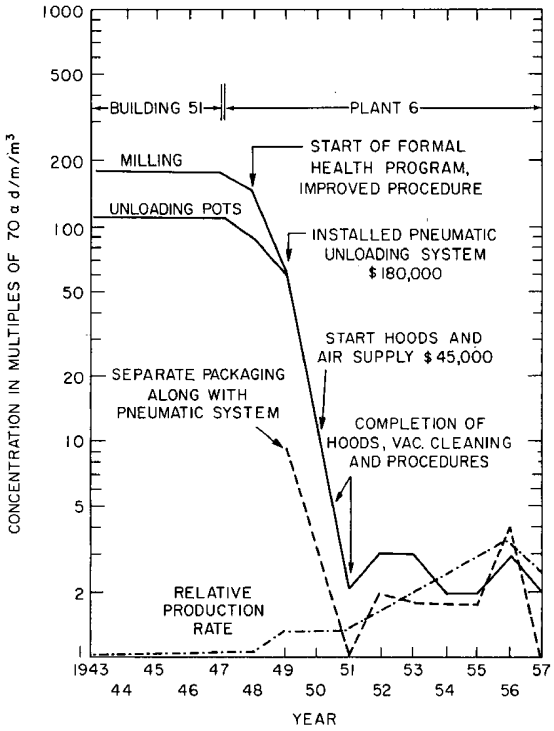

Figure 4. Dust concentration versus years; UO_3 operations.

technology and to design facilities along the lines of those used for plutonium; through the years, this has not been considered economically feasible. In our data some indication may be found of a framework for establishing the basic guides to a uranium health program.

COSTS

The cost of achieving the reduction in dust exposure is an important part of this story. During the period 1948 through 1950, about $300,000 was spent on dust control measures. In addition, the realization that the old hydrofluorination and metal plant (Plant 4) could not be brought under adequate dust control was an important factor in the AEC decision to replace it with two new plants; and ≈15% of the cost of the new plants went into dust control. From 1951 through 1956, an additional $600,000 was spent directly on health measures, of which about $400,000 was for dust control and dust collection. All in all, the AEC spent more than one million dollars, directly and indirectly, in improving the dust conditions in a plant with a capital value of about 25 million dollars.

We are now in the process of starting up the last unit of a completely new feed processing plant at Weldon Spring, which has been designed with the objective of achieving such a high level of dust control that the air concentration will at no point exceed 50% of the permissible level. Dust concentration data are not yet available, but the cost data for health provisions in this plant are presented in Table 5.

MEDICAL

When Mallinckrodt agreed to begin the processing of uranium in 1942 and 1943, the Company insisted upon extensive medical surveillance of its employees. The Washington University School of Medicine and the University of Rochester participated in the employee protection program. Staff physicians from the former set up a thorough clinical examination program for all employees, which is still in operation. Each employee has had at least a pre-employment and an annual physical examination including blood counts, urinalysis, and chest x-ray. Laboratory tests are made on exposed workers more frequently, from once a month to twice a year depending upon the particular operation.

In summarizing the medical information, it is of interest that at the beginning of 1958 more than 100 of the original employees working during 1943 through 1946 were still on the payroll. These employees have been under continuous medical surveillance by physicians having complete latitude to study any abnormality or to carry out any desired investigation to establish whether there is demonstrable damage to any employee from working with uranium. To date, no abnormal clinical finding has been traced to uranium as a cause. Albuminuria in 18 cases has been followed up, and in every case the causative agent was

Table 5

Cost Data on Dust Control at the Weldon Spring Uranium Feed Processing Plant
(Prices are direct plant costs plus overhead; engineering fees are not included.)

	Health provisions									
Area	Dust collection	%	Fume collection	%	Other	%	Total	%	Total area cost	% Total area cost
101	$ 282,000	100	$ —	—	$ —	—	$ 282,000	100	$ 2,290,000	12.3
103	270,000	73	100,000	27	—	—	370,000	100	5,000,000	7.4
201	408,000	77	—	—	123,000	23	531,000	100	6,032,000	8.8
301	250,000	68	50,000	14	65,000	18	365,000	100	5,195,000	7.3
403 & 404	45,000	100	—	—	—	—	45,000	100	1,996,000	2.3
406	6,000	100	—	—	—	—	6,000	100	211,000	2.8
407	30,000	100	—	—	—	—	30,000	100	3,900,000	0.8
410	—	—	—	—	384,000	100	384,000	100	1,355,000	28.3
Totals	$1,291,000	64	$150,000	7	$572,000	28	$2,013,000	100	$25,980,000	7.8

found to be something other than the occupational environment. There is no case of lung tumor or fibrotic tissue or other lung change traceable to uranium dust. There is no medical evidence of demonstrable damage to any employee due to chronic exposure to uranium. We are not at all smug about this – only grateful. At the present low levels of exposure we do not expect to see evidence of damage, but, because of the past history of high concentrations, we intend to continue the extensive medical program for at least another 15 years in order to obtain the type of information needed to establish realistic control standards. Today we are just approaching the time when we might expect to see lung damage and kidney damage from chronic exposure if it is going to occur. We believe that it will not occur and hope that we are correct.

LONG-RANGE OBJECTIVES

We have had high hopes of one day achieving a minimal exposure plant, but it has been a constant battle just to keep the exposure under 1 MAC. Each time a health improvement is completed, there has been an increase in production rate to offset it; even in the new plant, rates have been about double those expected. To date there exists no complete design for a zero exposure uranium processing plant, and, based upon the technology of current processes, to prepare one would be difficult if not impossible. There are unknown factors in any chemical process; pilot attempts to produce some of the uranium products with a dust tight process have resulted in materials acceptable by chemical analysis but not very good as process materials. Some recently designed fluid-bed reactor techniques offer a good possibility of solving many of the routine operational dust problems. However, maintenance work is always a source of high exposure, and much of it cannot be done under zero exposure conditions. In the present state of the art, we must rely on personnel protective equipment for some maintenance work and some nonroutine operations, and must keep a close watch on operational procedures.

CONCLUSION

The fifteen years of experience with uranium production shows no clinical or medical evidence of damage to workers from chronic exposure, even though some workers had potential high chronic dust exposure in the early years. Dust control continues to be a major health protection problem. The uranium industry as a whole does not have well defined standards for health protection and is faced with continued controversy in the matter of protection versus cost. It is imperative that adequate standards be developed so that the industry can proceed in an orderly manner. Whether the data indicate a need for the program to be conservative or liberal is secondary to the need for uniformity and for a clear definition of minimum performance requirements.

Occupational Exposures to Uranium Air Contamination in Feed Materials Production Facilities, 1948 - 1956

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During the years 1947 to 1954, the responsibility for feed materials production was in the hands of the New York Operations Office of the AEC. As a consequence, the Health and Safety Laboratory, a division of the NYOO, was able to acquire, from the various production facilities, a body of first-hand information on occupational exposures to uraniferous materials that is nearly all-inclusive up to 1954 and extends over a few more years on a partial basis. Extensive records of exposures to uranium dust in all the plants constitute a comprehensive air hygiene history and a logical introduction to current exposure data. These exposure histories are meaningful if considered together with the history and development of the production facilities.

The sequence of feed material operations began with the crushing and sampling of ore as received from overseas (later from the United States) and terminated with one of two products, gaseous uranium hexafluoride (UF_6) or uranium metal. The UF_6 was shipped to Oak Ridge for further processing; the metal was furnished to other Commission offices in various forms from rough ingots to finished components, depending on the offices' requirements. The products were normal uranium exclusively.

Figure 1 is a schematic flow plan of the various operations. In the organization of the exposure data, the processes will be considered in the six steps shown, i.e., ore sampling, refining, reduction and recasting, rolling, fuel element fabrication, and scrap recovery.

The production facilities operating in 1947 were those activated by the Manhattan District. They had been constructed hastily and on a temporary basis. Certainly, dust control equipment was not given the consideration that it receives in present installations. Furthermore, at first industrial hygiene supervision was unsatisfactory in many cases. Adequate supervision was complicated by the fact that the production sequences were performed in widely scattered plants. Many were too small to support the competent health supervisory staffs that are associated with later, integrated facilities. The production network was complex, consisting of a sampling plant in Middlesex, New Jersey, refining steps in St. Louis, Buffalo, and Cleveland, reduction and recasting in St. Louis and Buffalo, and scrap recovery in Pittsburgh. Metal rolling and component fabrication were performed in a dozen plants in as many cities.

By 1954, this picture had changed materially. Basic processes were consolidated in two essentially

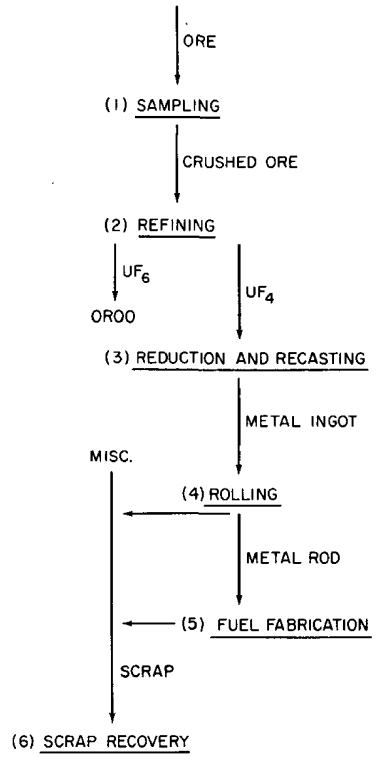

Figure 1. Uranium production sequence.

Table 1

Number of Production Workers by Year and by Process

Process	1948	1949	1950	1951	1952	1953	1954	1955	1956
Ore crushing and sampling	25*	25*	26	25*	23	23	20*	24*	28*
Refining	201	214*	267*	267*	235	212	167	230*	238*
Reduction and recasting	96	102*	122	135*	170*	232	310*	105*	135
Rolling	32	49	40	38	109	101	150*	223	—
Fuel fabrication	—	—	—	—	19	87*	100	123*	271
Scrap recovery	—	27	32*	37	50	50	34	43	—
Total	354	417	487	502	606	705	781	748	672

*Adjusted.

parallel production centers in St. Louis and Cincinnati. The Cincinnati plant carried out all consecutive steps from ore sampling through reactor component manufacture and scrap recovery, whereas at the St. Louis plant the steps included ore refining through metal recasting. The conversion of UF_4 to UF_6 had been transferred to Oak Ridge jurisdiction, as the other basic feed production steps were soon to be. Research and development and special production connected with fuel elements continued to be performed in a variety of installations throughout the United States, and still are.

As would be expected, the number of employees engaged in these operations increased during the stated period. The total number of workers in feed material production in 1948 was estimated as 350; it had increased to at least 780 in 1954, and was undoubtedly greater, as our information on fuel element fabrication and UF_6 production is incomplete for that year and for one or two prior years. In Table 1 are listed the numbers of production workers in each of the six process steps by year from 1948 to 1956. About half these numbers are totals taken directly from our records; the numbers marked with asterisks have been adjusted to compensate for missing segments of data in those years. The drop in numbers in several categories after 1954 reflects the Health and Safety Laboratory's decreasing participation in health programs at these production facilities after their transfer to Oak Ridge.

Throughout the period under discussion, the Health and Safety Laboratory's role was to provide health and safety supervision at the numerous facilities. The supervision was nearly absolute at some of the smaller plants which lacked the necessary health and safety skills. At other plants, supervision was only nominal, the Health and Safety Laboratory's function being to supplement the existing capabilities. But in all cases, periodic occupational dust exposure studies were conducted to evaluate hazards, to identify sources of exposure, and to provide information for the design of engineering controls.

The surveys were designed to obtain time-weighted average daily exposures. Representative replicate air samples were collected at all the jobs and in all areas to which each employee was assigned during the working day. Computations based on these sample data and time study data obtained during the surveys led to average exposure values for all employees in each plant. It should also be noted that the computed values neglect protection afforded by respirators. Thus, these are really potential exposures, but in very few cases are they substantial overestimates, as the use of respirators was inadequate and spotty. From these surveys, approximately 125 studies of occupational exposures at 25 plants were accrued over a period of 9 years. These studies are the sources of the data that follow.

In Figure 2 is plotted the exposure history for all uranium feed production employees by year from 1948 to 1956. For each year, exposures are represented as percent of total workers exposed to each of seven concentration ranges: 0 to 55, 55 to 110, 110 to 220, 220 to 440, 440 to 880, 880 to 1800, and >1800 d/m/m^3. Figure 2 reveals several interesting facts. For instance, in 1948, the number of production workers exposed to levels in excess of the present MAC, 110 d/m/m^3,

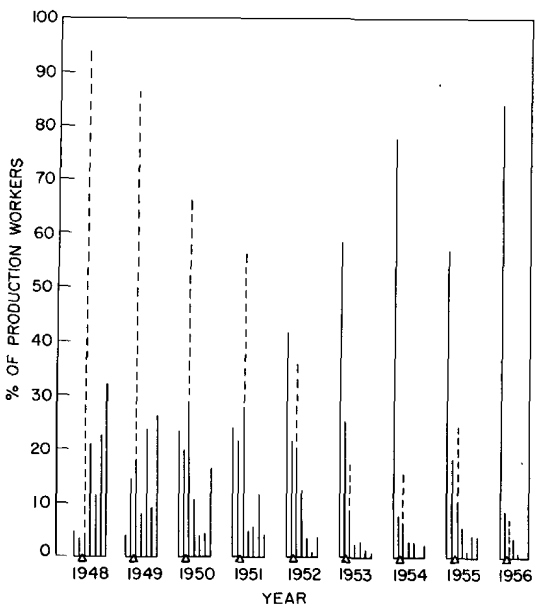

Figure 2. Dust exposure history of all uranium feed production workers. KEY: In each year, each vertical solid line indicates the percent of workers exposed to a different range (in units of d/m/m^3), the first being 0 to 55; the second, 55 to 110; the third, 110 to 220; the fourth, 220 to 440; the fifth, 440 to 880; the sixth, 880 to 1800; and the seventh, >1800. The broken vertical line in each year represents the percent of workers exposed to concentrations greater than the present MAC, which is 110 d/m/m^3, and is therefore the sum of the third through seventh solid lines.

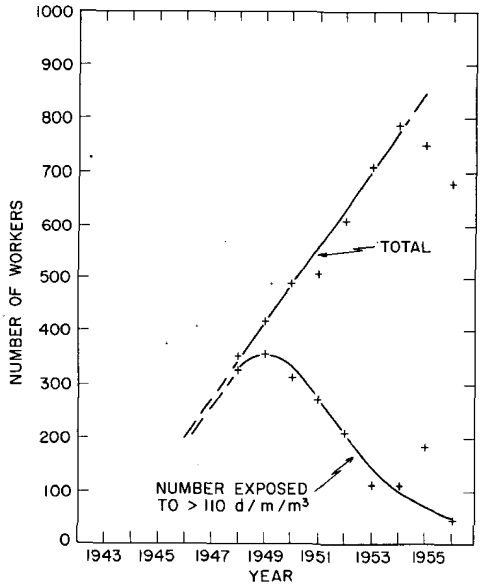

Figure 3. Number of uranium production workers by year, and number exposed to concentrations >110 d/m/m^3.

greatly exceeded the number with exposures less than this value. The percent exposed to >110 d/m/m^3, shown by the broken vertical line, was 91%, and 32% were exposed to average concentrations >1800 d/m/m^3. From 1948 on, this situation progressively improved at a fairly rapid rate. By 1951, approximately half the employees had exposures below the MAC, and only 4% were exposed to average concentrations >1800 d/m/m^3. By 1956, 94% had exposures below the MAC. Even so, a little less than 1% were still being exposed to >1800 d/m/m^3. Again, it is emphasized that these values represent the average concentrations in the working environment and do not take into account protection afforded by personal respiratory equipment.

In Figure 3 are plotted, by year, the total number of uranium production workers and the number of these who were exposed to >110 d/m/m^3. It is of interest to speculate about exposures prior to 1948, the year when our records begin. It would appear from the extrapolated curves that 90% or more of all workers prior to 1948 were exposed to >110 d/m/m^3. This is not unexpected in view of the fact that between 1945 and 1949 the recommended MAC for uranium was 500 γ/m^3 (700 d/m/m^3).

Figures 4 through 9 show the exposure distributions of workers in six sequential steps of the production process, plotted in the same manner as in Figure 2.

Figure 4 shows exposure data in sampling plants. It is interesting to note that all production workers were exposed to average concentrations >220 d/m/m^3 in 1950 and probably prior to that year. Little improvement occurred until 1955, when all exposures were less than the MAC. The explanation of this dramatic change is simple. The 1955 data are from an automatically equipped sampling plant, designated Plant B, that replaced Plant A, the facility that had been in operation since the early war years. The original plant was so antiquated that attempts at dust control were largely ineffective. The 1956 data apply to still another plant, designated Plant C, where the control was less effective than in Plant B.

Figure 5 is a plot of exposure data for workers in refining operations. For the purpose of this presentation, refining includes processing ore to uranium oxide, conversion of the oxide to tetrafluoride, and subsequent conversion of the latter to hexafluoride or its reduction to metal. That these data are quite similar to those in Figure 2, apply-

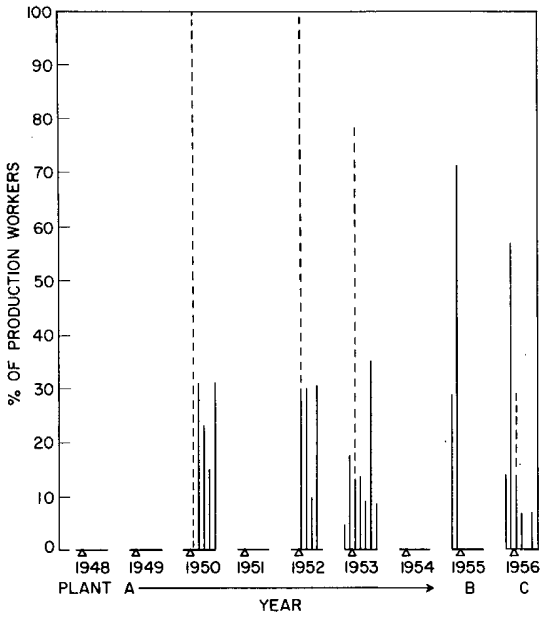

Figure 4. Dust exposure history of workers in sampling plants. KEY: See Figure 2.

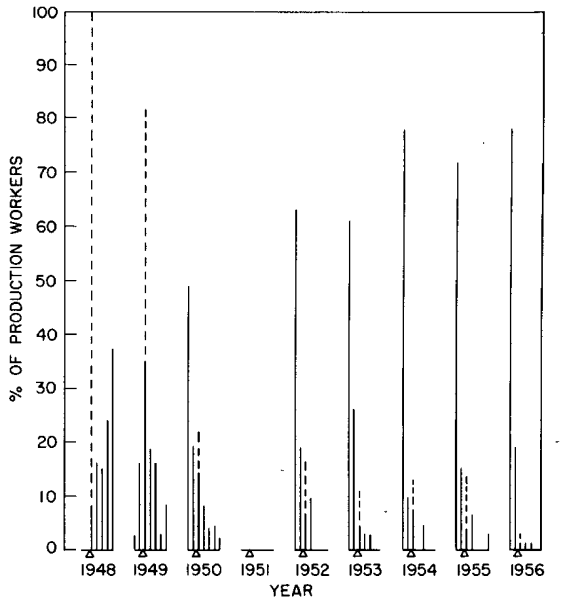

Figure 6. Dust exposure history of workers in reduction and recasting. KEY: See Figure 2.

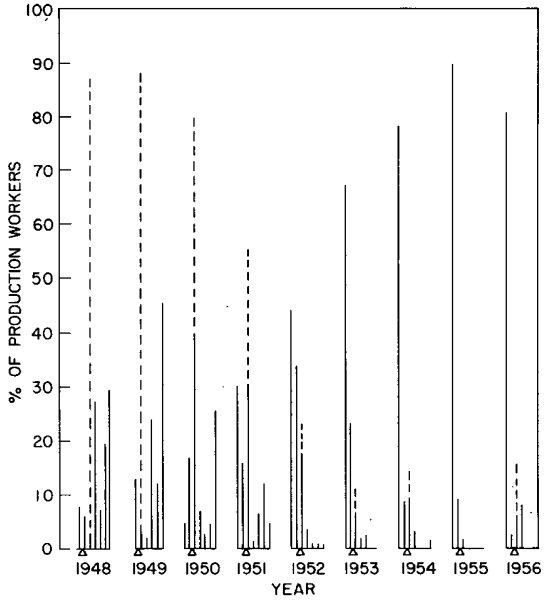

Figure 5. Dust exposure history of workers in refining operations. KEY: See Figure 2.

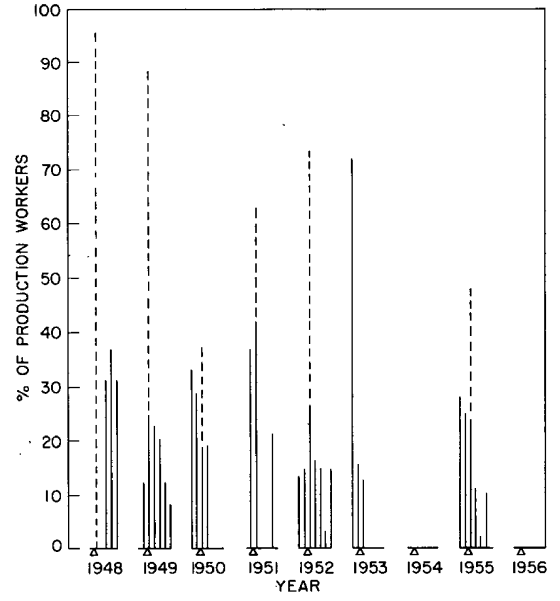

Figure 7. Dust exposure history of workers in rolling operations. KEY: See Figure 2.

ing to all production workers, is not unexpected, since a large proportion of all workers were employed in the refining industry. The figure shows that control was very nearly perfect by 1955; in 1956 there was a slight retrogression resulting from a large increase in production volume.

Figure 6 applies to reduction and recasting; and here again the general trends are similar to those for all workers taken together.

Figure 7 presents data for rolling operations. Through 1952, all production rolling was manual. Although improvement in dust control was realized

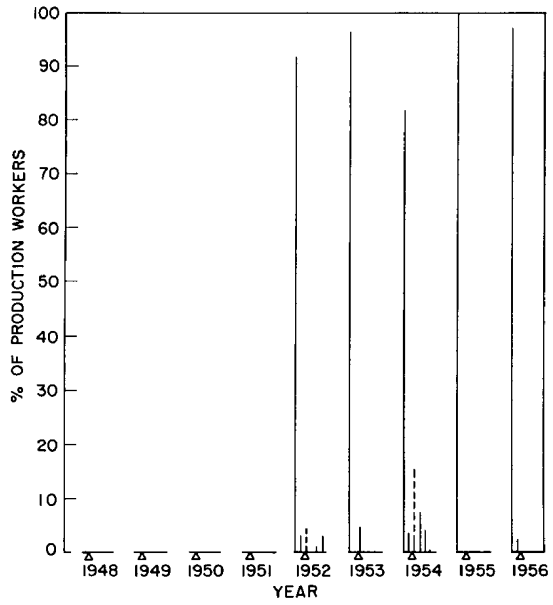

Figure 8. Dust exposure history of workers in fuel fabrication. KEY: See Figure 2.

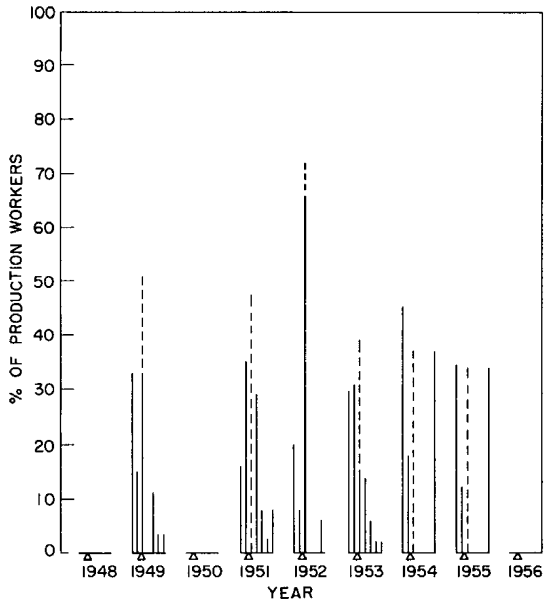

Figure 9. Dust exposure history of workers in scrap recovery. KEY: See Figure 2.

from year to year, it was not until 1953, with the change to automatic rolling equipment, that satisfactory control was achieved.

The record for fuel fabrication, as shown in Figure 8, is good. Our data for this production step are nonexistent prior to 1952 and are somewhat spotty after that time. However, most fuel

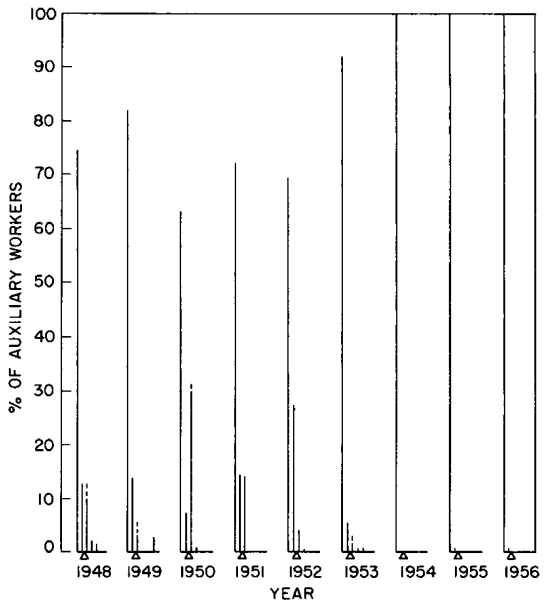

Figure 10. Dust exposure history of auxiliary employees. KEY: See Figure 2.

fabrication operations are performed on a smaller scale than any of the other processes under discussion, and control is less challenging.

Exposures in scrap recovery operations, presented in Figure 9, appear not to have changed materially over a number of years.

All the previous data apply exclusively to production workers. However, the Health and Safety Laboratory surveys frequently have included auxiliary workers; that is, employees not directly connected with production but located in or near production buildings. The auxiliary groups include chemists, engineers, office workers, garage mechanics, outside maintenance personnel, and the like. Some of these individuals had occasion to be in production areas part time, others had no direct contact with production; nevertheless, a small but significant number were exposed to uranium dust. These data are presented in Figure 10. These data are too spotty to be representative of the entire industry; however, it is of interest that in 1948, $\approx$13% of the auxiliary workers studied were exposed to concentrations >110 d/m/m^3 and >1% were exposed to >440 d/m/m^3, whereas by 1954 no exposures >110 d/m/m^3 were found.

In summary, between 1948 and 1956, there was a radical change in occupational exposures in the uranium production processes. In 1948, and probably before that time, 90% of production

workers were exposed to average concentrations in excess of the present MAC of 110 d/m/m^3, and 32% were exposed to average concentrations $>$1800 d/m/m^3. By 1956, conditions had improved remarkably, with only 6% of production workers exposed to average concentrations higher than the MAC. The improvement was brought about mainly through the construction of modern processing facilities to replace those that had been put into operation during the war.

Survey of Air-Borne Normal Uranium From Various Operations at Los Alamos Scientific Laboratory

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Over the past few years personnel of the Los Alamos Scientific Laboratory have done a great deal of work with normal uranium. This work has involved a variety of operations and conditions ranging from strictly laboratory type to small-scale semi-industrial. In order to protect operating personnel and to learn what uranium air concentrations arise from particular operations, Laboratory Health Division personnel have collected air concentration data from many of these operations. These data are presented to give a comprehensive view of normal uranium concentrations in air for some of the work areas of this Laboratory.

The data to be presented are listed in tables according to the operations to which they pertain. All concentrations are for alpha radiation and are given in disintegrations per minute per cubic meter of air. No corrections were made for filter paper counting efficiency.

The samplers used in collecting the data were as follows: Filter Queen at 4 cfm with HV-70 paper, Gast pump at 20 liters/min with glass or HV-70 paper, HiVol sampler at 20 cfm with glass paper, central sampling system at 2 cfm with HV-70 paper, Sutorbilt pump at 16 cfm with HV-70 paper, and Giraffe at 2 cfm with HV-70 paper.

Table 1 shows general air concentrations for three foundry rooms of semi-industrial nature (see Figures 1 and 2). Filter Queens were placed about 5 ft from the furnaces for one set of samples and about 3 ft from where breaking and stripping was done for another set. There was no local ventilation except in the case of the furnaces listed in the last two lines in the table. General room air change was about once every 10 min. Sampler running time was $\approx$8 hr.

In Table 2 are listed general air, breathing zone, and stack concentrations for a semi-industrial machine shop containing about 15 lathes (see Figures 3 and 4). There are three principal sets of samples for this shop: (1) those taken in the old shop building before local ventilation was installed, (2) those taken in the old shop after local ventilation was installed, and (3) those taken after the shop was relocated in a new building. Ventilation and housekeeping in the old shop were poor; however, local ventilation on the machines brought about a marked improvement. In the new shop ventilation is good and housekeeping improved. Samplers used were Filter Queen, Gast pump, central sampling system, and Sutorbilt pump. The sampler running time was 3 to 8 hr.

*Data were collected by CHARLES D. BLACKWELL, JOHN W. ENDERS, EDWIN C. HYATT, ROBERT N. MITCHELL, AND DONALD A. MCKOWN.

Table 1

General Air Concentrations in Three Foundry Rooms (d/m/m^3)

Location	No. of samples	Maximum	Average	% Below 18 d/m/m^3	% Below 70 d/m/m^3
Breaking and stripping	546	266	16	74	94
Furnace loading, unloading	546	99	9	86	98
Furnace loading, unloading	468	119	8	88	99
Breaking and stripping	434	289	15	78	98
Furnace loading, unloading	434	388	7	93	99

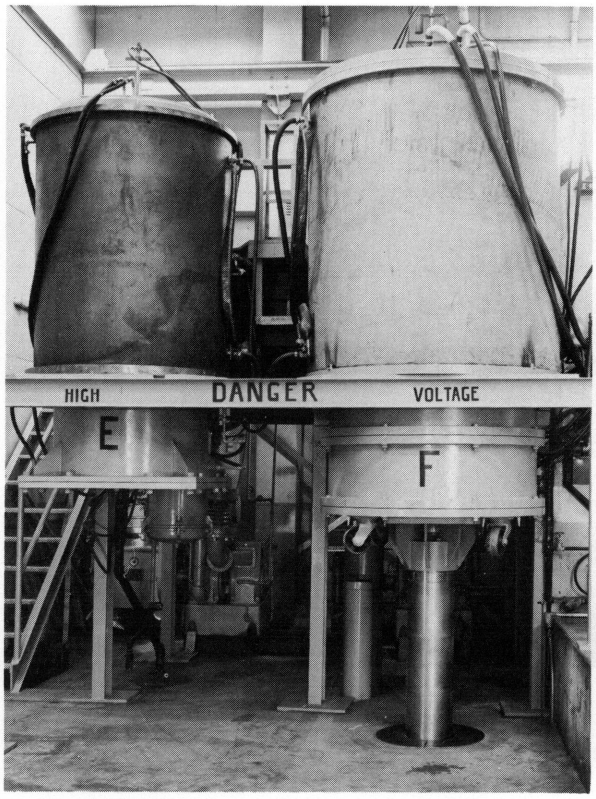

Figure 1. Normal uranium furnaces (see Table 1). Two furnaces in a LASL foundry room are shown. There is a Filter Queen sampler behind the furnace, ≈4 ft away, and another on the floor at the left.

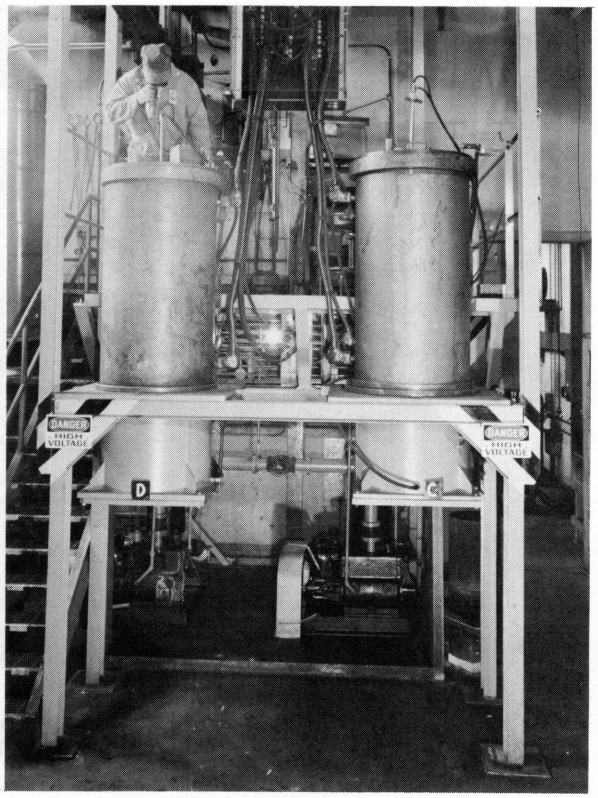

Figure 2. Normal uranium furnaces (see Table 1). Another foundry, with several small furnaces. One sampler was used in this case, located approximately behind the worker.

Table 2

General Air, Breathing Zone, and Stack Concentrations in a Semi-industrial Machine Shop (d/m/m^3)

Location	No. of samples	Maximum	Average	% Below 18 d/m/m^3	% Below 70 d/m/m^3
		Old Shop Before Ventilation Installed			
General air (Filter Queen)	742	407	46	24	80
Breathing zone (Gast pump)	105	1,760	207	7	29
		Old Shop After Ventilation Installed			
General air (Filter Queen)	232	18	4	99	100
Stack (Gast pump)	38	1,330	178	28	50
		After Shop Moved to New Building			
General air (Filter Queen)	157	23	2	99	100
Breathing zone (central sampler)	572	202	6	97	99
Stack (Sutorbilt pump)	80	22	4	98	100

Figure 3. Normal uranium machine shop (see Table 2). This shows the old shop after local ventilation was installed; note the flexible ducts extending down to the machines.

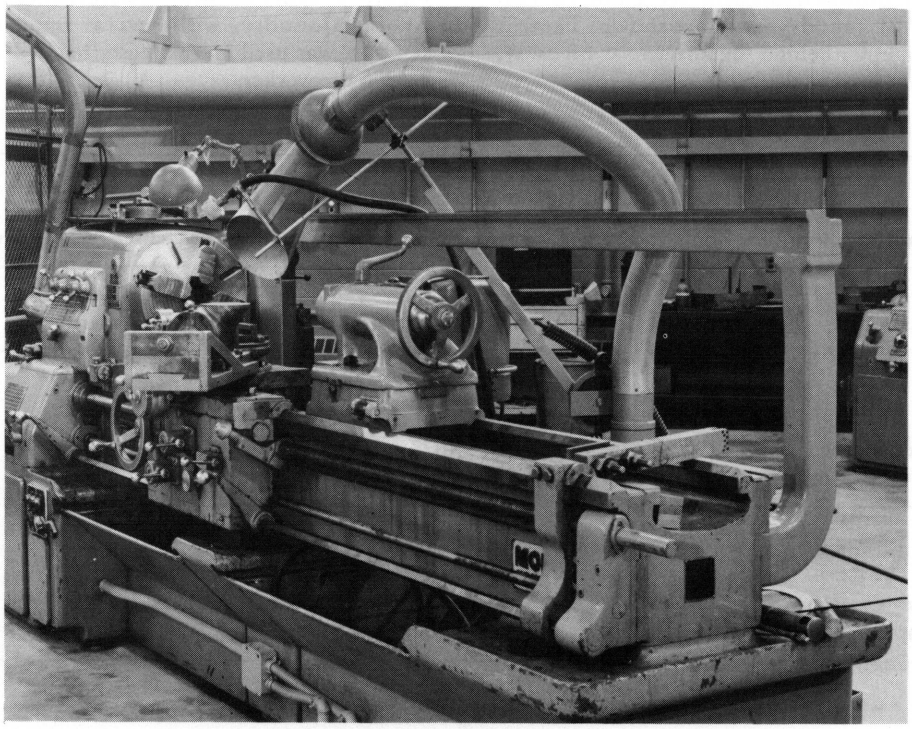

Figure 4. Normal uranium machine shop (see Table 2). One of the lathes in the new shop building, with local ventilation. Just above the chuck is one of the sampler heads; its hose runs back down to the floor where it joins the central sampling system. This sampler operates at 2 cfm.

Table 3

General Air Concentrations Near Some Scrap Processing Operations (d/m/m³)

Operation	No. of samples	Maximum	Average	% Below 18 d/m/m³	% Below 70 d/m/m³
		Before Vacuum Transfer Unit Installed			
Chip pressing	45	5,700	168		
Oxide handling	11	300,000	10,500		
		After Vacuum Transfer Unit Installed			
Scrap burning, oxide handling	669	597	13	88	97

Table 3 shows general air concentrations near some of the operations of scrap processing before and after a vacuum transfer unit was installed (see Figure 5). Some of the operations to be noted here are chip pressing, scrap burning, and oxide handling. Before the vacuum system was installed, oxide was handled with a shovel and the ventilation in the small building where this was done consisted of only a small vent fan. The sampler used here was a Filter Queen running ≈8 hr.

In Table 4 are listed breathing zone concentrations for a group of special operations, some of which may be termed semi-industrial (see Figures 6 and 7). The samplers used were Gast pump, for the samples in the first two lines, and Giraffe for the balance. The average running time was 30 min.

Table 5 shows general air concentrations for some of the operations listed in Table 4. The data in the first two lines were collected with a HiVol sampler, and those in the third with a Giraffe. There was no local ventilation on any of these operations.

In Table 6 are listed both breathing zone and general air concentrations from a welding rod extrusion operation, the former sampled with a Giraffe and the latter with a Filter Queen. Running time was ≈30 min. There was local exhaust ventilation on the press only.

Originally this operation was performed with essentially bare metal, which was put into the furnace, heated to the proper temperature, loaded into the press, and extruded. The operators found that putting a plug of glass wool into the die ahead of the billet increased the life of the die and also

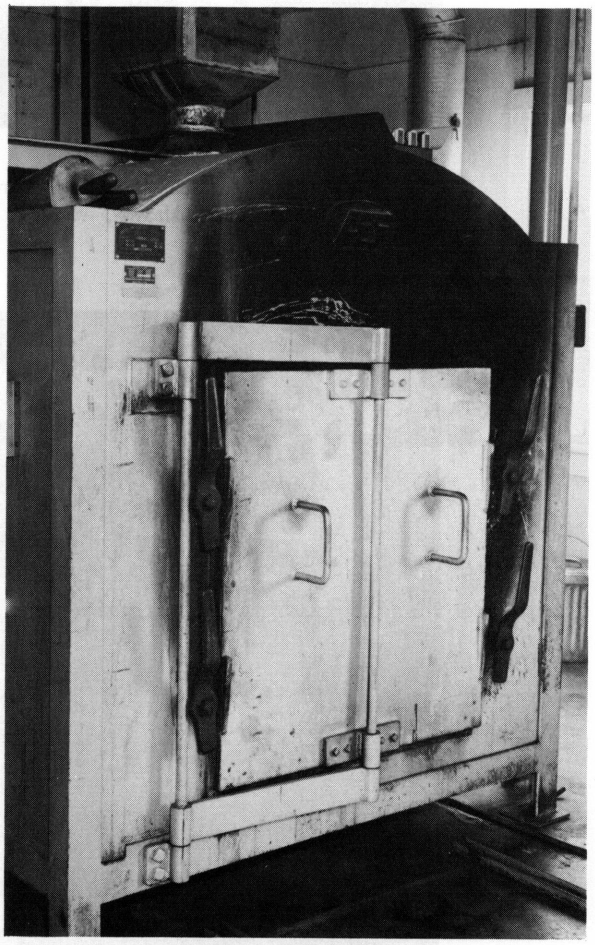

Figure 5. Gas fired furnace for normal uranium scrap burning (see Table 3). Note tracks on floor on which a hood with exhaust ventilation can be rolled up to the front of the furnace, so that the trays in the furnace can be pulled out into the hood.

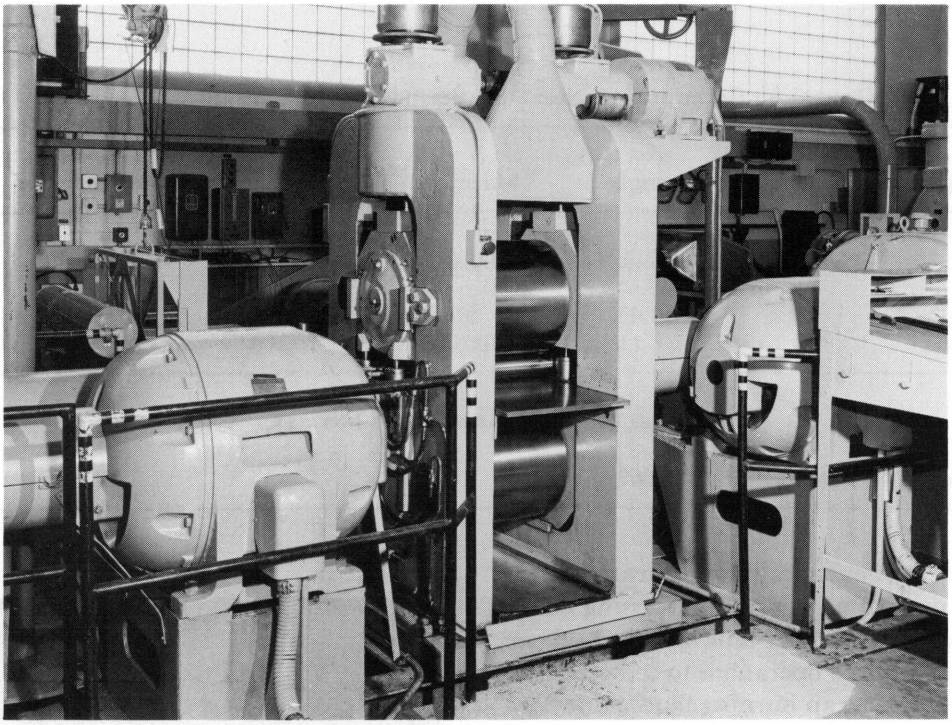

Figure 6. Normal uranium oil rolling (see Table 4). A mill with local ventilation is shown, which is used for rolling out of an oil bath.

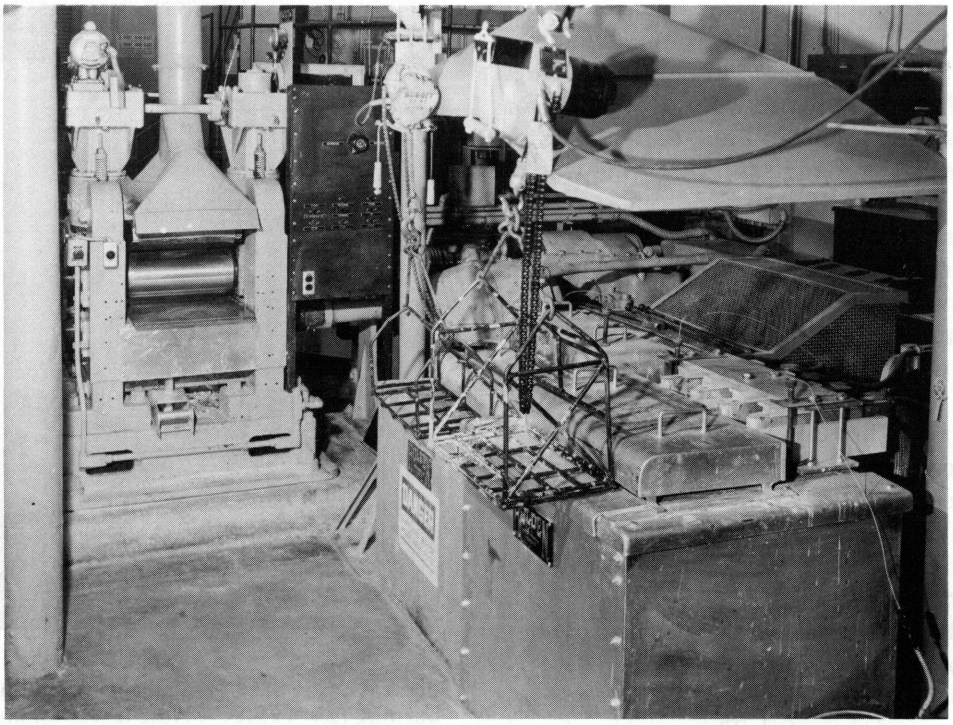

Figure 7. Normal uranium salt rolling (see Table 4). A mill for rolling out of a salt pot, with ventilation on the mill and a hood over the salt pot.

Table 4

Breathing Zone Concentrations for Special Operations (d/m/m³)

Operation and location	No. of samples	Maximum	Average	% Below 18 d/m/m³	% Below 70 d/m/m³
Rolling From Salt Pot					
Roll mill operator	13	68	24	38	100
Roll mill operator* (small mill)	49	91	11	85	95
Shear operator*	12	53	310	0	16
Rolling From Oil					
Roll mill operator*	37	5	5	100	
Impact extrusion	15	1,645	254	26	60
Unloading furnace	8	927	197	0	25
Stripping casting	37	310	73	24	70
Crucible cleaning	31	760	159	0	12
Breaking press	21	38	10		
Spot welding (inert gas)	6	6	2	100	
Continuous welding*	13	32	12	69	100

*Indicates local ventilation on the operation.

Table 5

General Air Concentrations for Special Operations (d/m/m³)

Operation	No. of samples	Maximum	Average	% Below 18 d/m/m³	% Below 70 d/m/m³
Impact extrusion	12	55	6	91	100
Rolling from salt pot	32	38	9	81	100
Deep drawing	42	63	8	85	100

Table 6

Breathing Zone and General Air Concentrations for a Welding Rod Extrusion Operation (d/m/m³)

Sampler location	No. of samples	Maximum	Average	% Below 18 d/m/m³	% Below 70 d/m/m³
Salt and Glass Wool Not Used					
General air	8	300	162	0	25
Exit end of die	7	12,800	3,206	0	0
Breathing zone, operator handling extruded rod	4	1,235	616	0	0
Salt and Glass Wool Used					
General air	5	54	26	40	100
Breathing zone, heating and loading press	11	220	69	0	63
Breathing zone, press operator	6	39	26	16	100
Breathing zone, operator handling extruded rod	12	180	66	8	83

Table 7

General Air Concentrations in Four Typical Laboratory Operations (d/m/m³)

Operation	No. of samples	Maximum	Average
Analytical chemistry, wet work	1,046	27	<1
Analytical chemistry, dry work	1,046	7	<1
Furnace room (10 small furnaces)	1,453	21	<1
Small foundry and machine shop	437	12	1
Polishing and cutoff wheel	247	1,969	6
Polishing	745	10	1

very greatly reduced air concentration levels. Immediate covering of the extruded rod, which was wound up in a circular drum, with salt also greatly reduced air-borne concentrations.

Table 7 shows general air concentrations in four typical laboratory operations. Filter Queens, running ≈8 hr, were used. Local ventilation was generally used in these operations. Concentrations in a room where an unventilated cutoff wheel was used are shown in the next to the last line, and concentrations in the same room after the cutoff wheel was discontinued are shown in the last line. General room air change was about once every 3 min.

In conclusion, it can safely be said that since 1953, when collection of these data started, there has been a general trend downward in air concentrations in most areas, even in some areas where no special efforts to reduce concentrations have been made.

Air Sampling for the Control of Internal Exposure From Enriched Uranium at Y-12

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Abstract

This paper describes very briefly the program for determining the concentration of uranium in the air being breathed by persons working in the enriched uranium processing areas of the Y-12 Plant of Union Carbide Nuclear Company at Oak Ridge, Tennessee. The aim of the program, ways to accomplish this aim, and the methods used to process the data accumulated are mentioned. The Y-12 Plant limit for permissible concentration of air-borne uranium and its origin or derivation are given.

The sampling program and techniques are rather standard, but mention is made of three unique features: the use of a cardboard planchet to simplify handling of samples, the use of automation to expidite sample counting, and the use of several twin-head sample collectors and statistical analysis to determine over-all program precision and optimum sampling frequency.

Figures illustrate air sampling and counting equipment and graphs show uranium concentrations determined by the program.

The program for monitoring the atmosphere in the enriched uranium processing areas of the Y-12 Plant of Union Carbide Nuclear Company is not unique. We use the rather standard procedure of drawing the air through a filter to collect the suspended particulate contamination and then counting the disintegrations from the radioactive material collected on the filter in a suitable counting instrument. The general philosophy or aim of the air monitoring program at Y-12 is to see that the average concentration of radioactive contaminants in the air remains within reasonable limits and is not hazardous to employees. To accomplish this aim, one must collect many samples representative of the air being breathed by the majority of the persons, but one also must monitor many individual operations which may result in localized high concentrations of air-borne uranium. It follows then that our air samples are classified as either general air, breathing zone, or operational samples. These descriptive classifications indicate that the samples were collected to determine either the average concentration of air-borne uranium in an area, the concentration in the air being breathed by a particular operator, or the amount being released to the atmosphere by a particular operation.

Figure 1 shows a typical sample head used for both portable units and permanent systems. Shown are the two-piece quick-opening paper holder, the rubber gasket, wire "spider" to support the paper, and the 1¼-in.-diameter sample disc of Hollingsworth and Vose No. 70 filter paper. Each of these sample heads is connected to its source of vacuum through a rotometer and valve to permit adjustment of the air flow rate.

Figure 2 shows the basic sampling equipment used in most locations at Y-12. The majority of our air samples are collected by means of permanently installed systems of several sample collection units connected by pipe manifolds to a special vacuum pump or perhaps the house vacuum system. The primary consideration in determining the locations for these sample collection units is that the samples be representative of the average atmosphere breathed by the majority of persons in the area. Thus, they must not be biased or unduly influenced by high concentrations localized at any one operation. It can be seen that one very high activity sample averaged in with the results from many very low samples may suggest that the atmosphere being breathed by the majority of persons in a given work area is highly contaminated when such definitely is not the case. On the

other hand, close scrutiny of the levels of activity shown by these individual samplers is necessary for the control of both local and average contamination levels.

Portable units similar to the one shown in Figure 2 are used for collecting breathing zone or operational samples to locate sources of air contamination, or to evaluate new operations or the effects of operator technique, changes in equipment or ventilation, etc., on the levels of air contamination. Each unit has its own vacuum pump and air flow adjustment with the collection head mounted on a telescoping boom for versatility. These units also may be used to collect general air samples in temporary situations or where permanent systems have not been installed.

Equipment, such as the Hi-Vol sampler shown in Figure 2, is used for collecting special samples which require a large air volume in a short collection time. Several different types of filter collectors and an adaptor to permit use of an annular impactor are available for use with this device.

One rather unique feature of our sample handling technique is the doughnut shaped cardboard planchet on which our samples are mounted for handling and counting. The planchets of ⅛-in.-thick cardboard are $\approx$ 2⅝ in. in diameter with a 1⅜-in.-diameter hole in the center. Masking tape stuck onto one side of the planchet leaves a gummed surface exposed in the hole. The health physicist collecting air samples carries a supply of the planchets in a special dispensing container similar to a bus driver's coin changer. Using forceps he removes the paper disc from the sampler head and places it in the planchet so that it adheres to the masking tape. Pertinent sampling and identification data can be recorded on the planchet itself.

Another unique feature of our air sampling program is the use of statistical analysis of past experience to determine the frequency with which sampling must be done to provide the desired reliability of results. In two areas we have installed a pair of twin sampling heads side by side. The analytical results on the samples collected simultaneously during a quarter year by these twin heads are compared statistically to obtain a percent limit of error for the mean concentration. Our statisticians examine the data from samples collected over an extended period and apply the limit of error suggested by the twin sample data to determine the minimum number of samples which should be collected in a given area in order that the average shown by the data may have the desired reliability.

Air samples together with printed data forms are sent to a central counting laboratory. To permit the decay of the short-lived alpha-emitting daughters of radium and thorium and to be sure

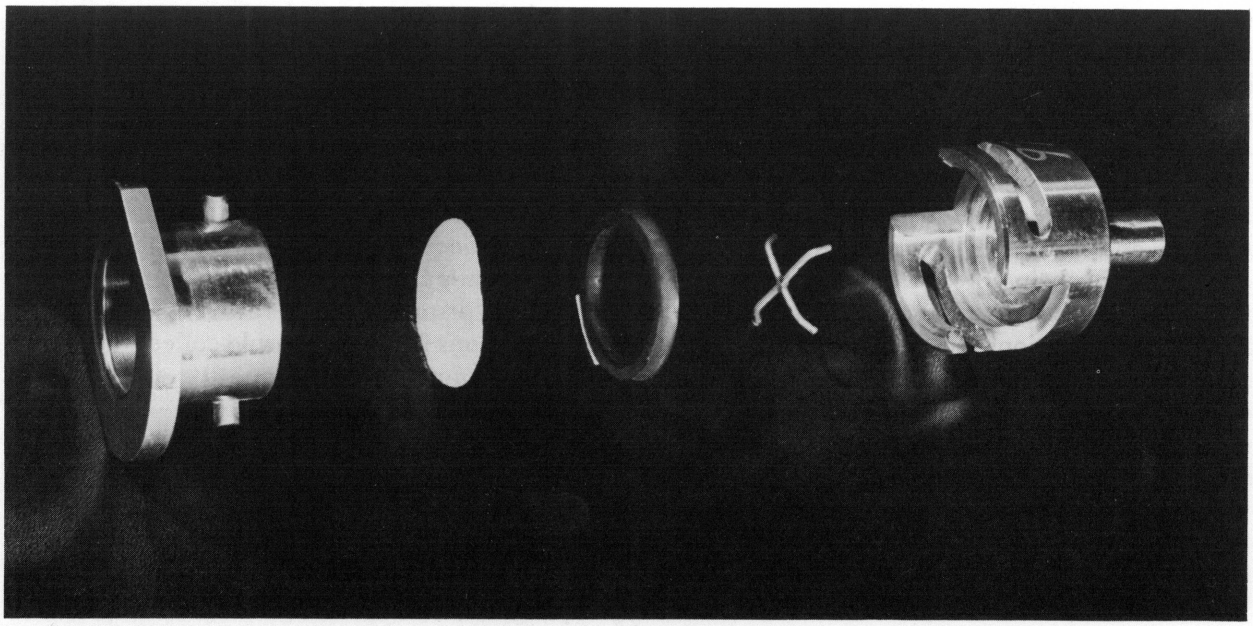

Figure 1. Air sampler head.

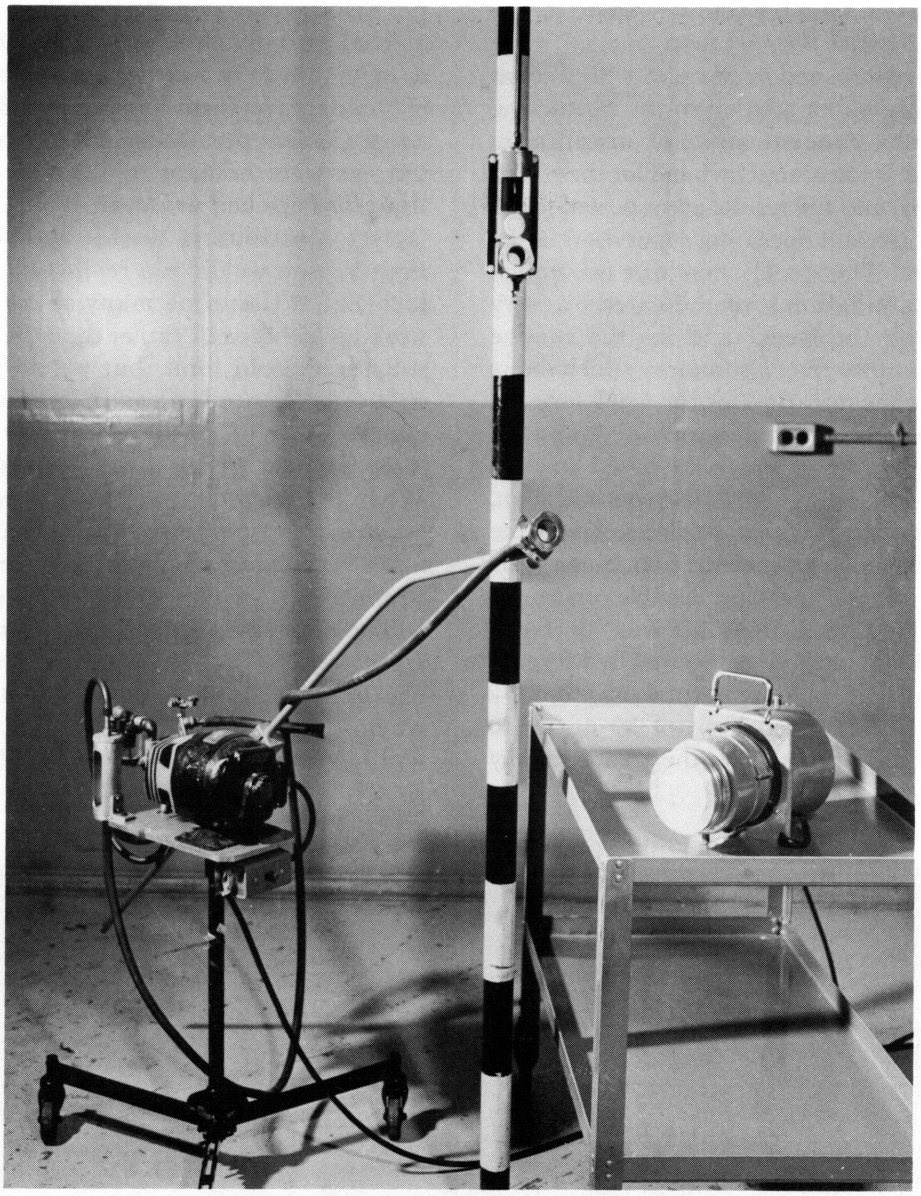

Figure 2. Air sampling equipment.

that the activity counted is that from the long-lived uranium, samples are not counted until at least 16 hr after the end of the sampling period.

Routine general air samples are counted in an automatic alpha scintillation counter (Figure 3). The counter has twin scintillation detectors which permit each sample to be given two 4-min counts. The two counts and an identification number for each sample are printed on a tape. The counter usually is loaded with 125 to 130 samples (about an 8-hr supply). Often it is loaded in the afternoon and left to count overnight. In the morning the counted samples are found stacked in the tube on the right, and sample counts have been printed on the paper tape. Upwards of 300 samples can be loaded into the sample tubes, but the weight of that many samples sometimes causes the mechanism to jam. Breathing zone, operational, or other special samples are counted in Nuclear Measurements Corporation gas proportional counters.

Considerable use is made of machine computers in the calculation and tabulation of data from the large numbers of air samples collected during a week. However, the completed data

cards are first sent from the counting laboratory to the Health Physics Department central office where they are screened to check any high count samples before being sent on to the tabulating machines. The concentration of uranium in $d/m/m^3$ of air is calculated by hand for these high count samples, and the results are reported to the health physicists and operating supervisors in the areas affected. The weekly machine tabulation lists the contamination level indicated by every sample during the week; and for the routine general air samples gives a summary which shows the average concentration for the week for each area, the number of samples included, and the percent of the samples which indicated levels in excess of the maximum permissible concentration.

At this point it is pertinent to discuss briefly the maximum permissible concentration. Some years ago the limit we use for the permissible concentration of enriched uranium in air was "derived" from the preliminary limits suggested by Morgan[1] and Neuman[2] for insoluble normal uranium. By converting the suggested limit of 50 $\mu g/m^3$ to $d/m/m^3$ and adjusting this for the greater energy per disintegration of the enriched uranium, we arrived at a maximum permissible limit of 70 $d/m/m^3$ air. When in 1953 the National Bureau of Standards published its *Handbook 52*[3] suggesting a more lenient permissible limit for natural uranium, we were hesitant to adopt it or revise our limit for enriched uranium. There were several factors contributing to this hesitancy, but the main reason we did not revise our limit was the fact that at that time many of our people were working 56 hr/week rather than the usual 40. We still use the old limit, but we do not take any stringent action unless a given location consistently exceeds the MPC or the average for a whole area is too frequently close to or in excess of the MPC. When the revised version of *Handbook 52* is published in the near future, we will change our plant limits to conform to the ones recommended therein. Prepublication information suggests that there will be no really drastic change from the present *Handbook 52* MPC for uranium in air; thus, the new limit likely will be more lenient than the one we now use. If this is the case, we undoubtedly will have to revise our present policy and take

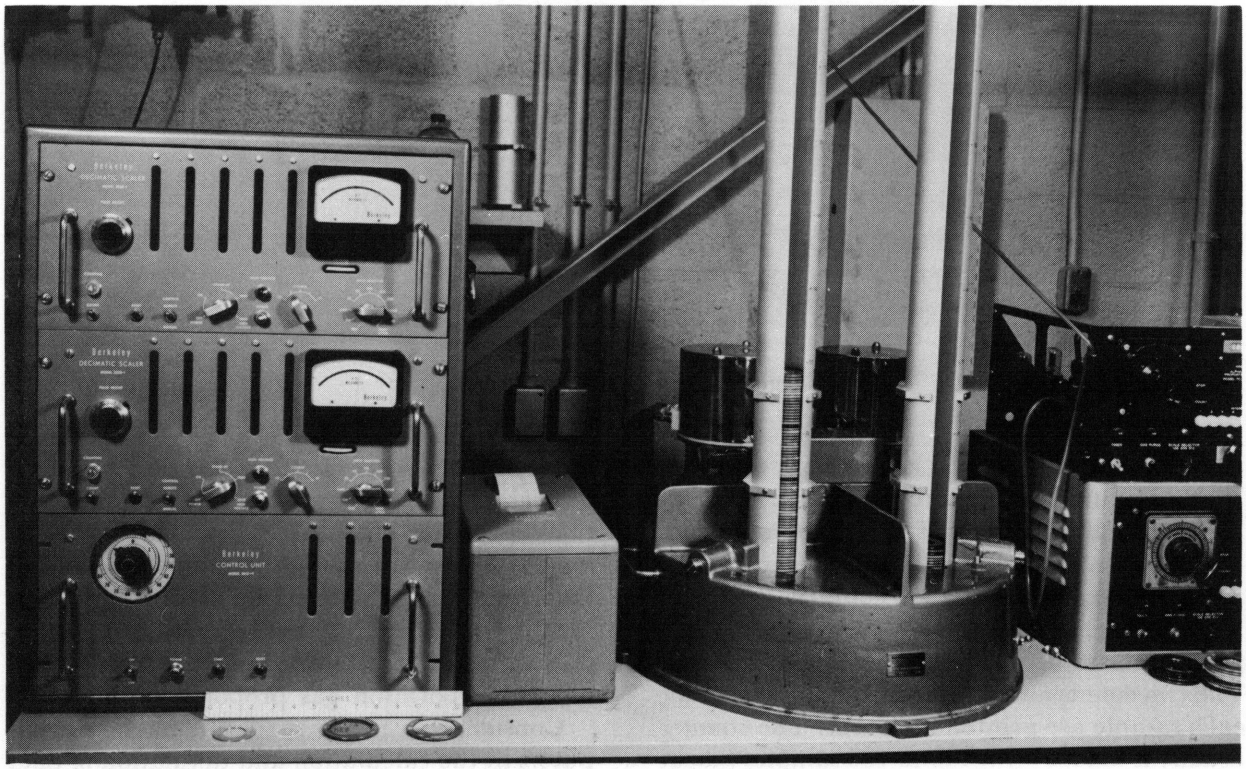

Figure 3. Automatic air sample scintillation counter.

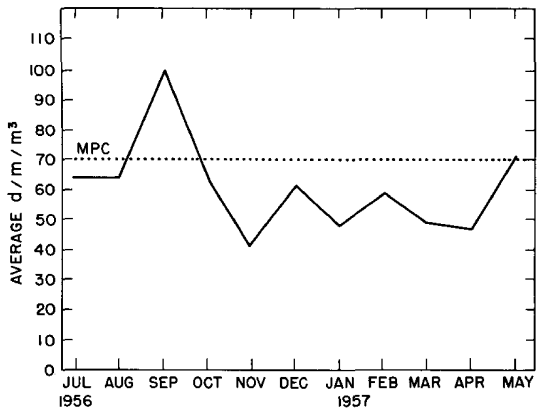

Figure 4. Average concentration in general air samples (≈1950 samples/month).

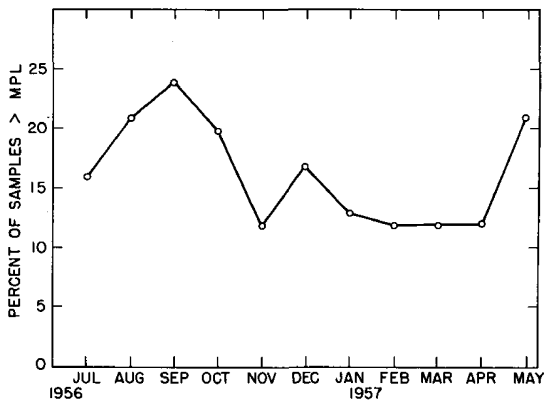

Figure 5. Percent of general air samples which exceeded plant maximum permissible level (≈1950 samples/ month).

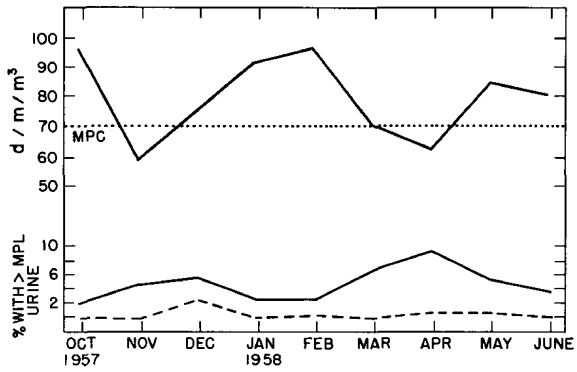

Figure 6. Correlation of average uranium concentration in air (upper curve, ≈1550 samples/month) with percent of persons having urinalyses exceeding the maximum permissible limit (solid lower curve; total number of persons, ≈250) and with percent of persons having urinalyses exceeding the action point (broken lower curve; same group of persons).

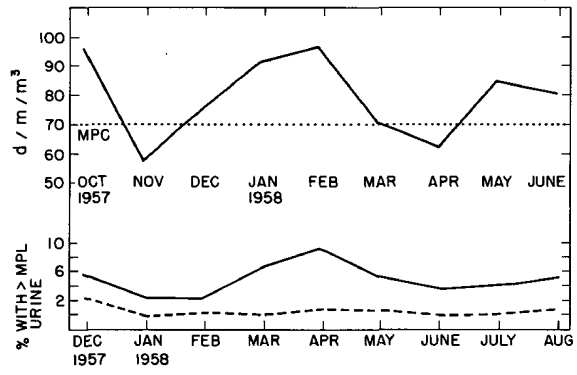

Figure 7. Same curves as in Figure 6, but with lower time scale shifted two months ahead of upper time scale.

more positive action when either localized or general concentrations approach the permissible limit.

Figures 4 through 10 illustrate some of the atmospheric uranium concentrations determined by our air sampling program.

Figure 4 shows, for each of the first 11 months of fiscal 1957, the average of the concentrations indicated by some 1950 samples per month in the entire enriched uranium processing facility. For the most part, the average concentration shown is less than our plant maximum permissible (dotted line), and the peak concentration shown (September 1956) is less than 40% of the limit which may be suggested in the new *Handbook 52.*

Figure 5 shows the percent of the same 1950 samples per month which exceeded our plant maximum permissible level. This illustrates that a relatively few high samples can cause average concentrations to be rather high and suggest a possibly biased or untrue average.

The upper curve of Figure 6 shows the average concentration for several, but not all, of the enriched uranium processing areas during the last 9 months of fiscal 1958. The solid lower curve indicates the percent of the persons breathing this air (total ≈250) whose urinalyses suggested an internal dose in excess of the maximum permissible limit. The lower broken curve indicates the percent of those exposed whose urinalyses exceeded our action point calling for restricting their exposure to uranium. As one might suspect, the urinalysis peaks do not quite coincide with the air peaks. However, Figure 7, in which one time

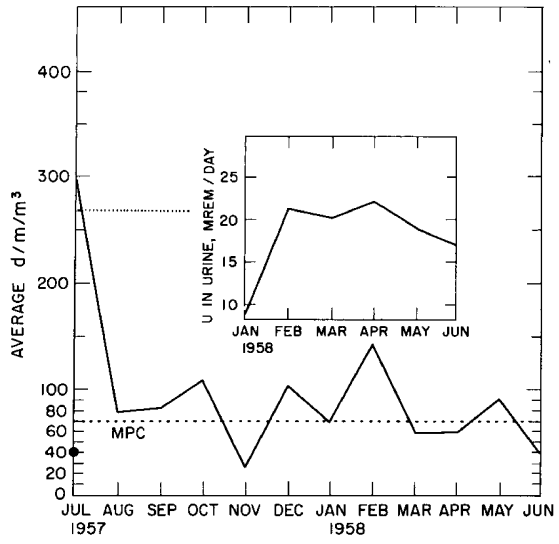

Figure 8. Average air concentration in an enriched uranium machine shop. Inset shows urinalyses for comparison.

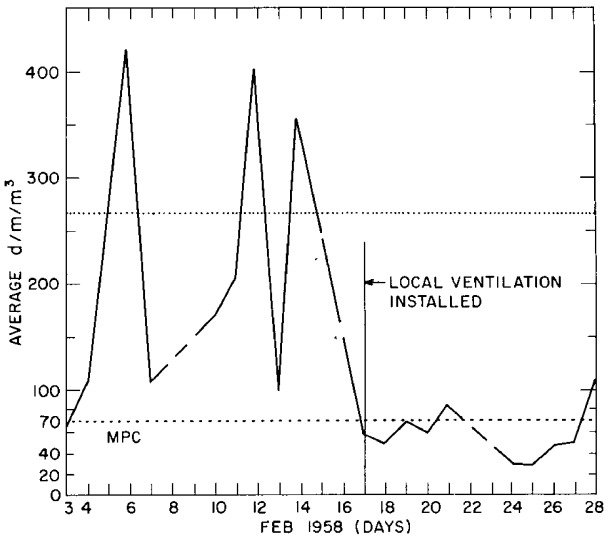

Figure 9. Daily average air concentration in the same shop as that in Figure 8. The over-all average dropped from 216 d/m/m³ before local ventilation was installed to 55 d/m/m³ afterwards.

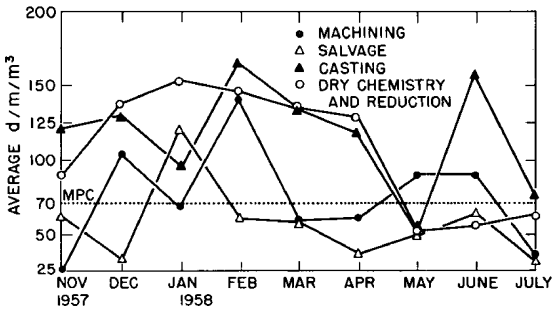

Figure 10. Comparison of monthly average air activity levels in four different types of processing operations.

scale has been shifted with respect to the other, shows that the urinalysis peaks really do follow the air peaks, but with a lag of about two months before changes in average air concentration are reflected in the average internal dose picture of a large group. The main reason the urinalysis curves do not correspond more closely to the air curves is that urinalyses are averaged over three-month periods, whereas air sample averages include data for only one month. Factors not reflected in air sampling, such as occasional use of respirators or area evacuation during high air activity releases, also tend to flatten the peaks of the urine curves and prevent their correlation with air data curves.

Figure 8 shows the monthly average air concentration in an enriched uranium machine shop. The high concentration shown for July was the result of a chip fire which caused extremely high air activity on only one day. If the results for this one day were ignored, the average for July would be only 40 d/m/m³. Even including this one day, the July average is not greatly above that which may be the new *Handbook 52* limit (upper dotted line). For the sake of comparison, the small insert graph is the urine picture for the approximately 33 persons working in the machine shop. In this case, the urine points are the monthly averages as are the points on the air curve. It can be seen that the internal dose suggested by urinalyses follows the trends of the average air activity curve quite closely, but the 2-month lag is still evident.

Figure 9 shows how the level of air activity in the same machine shop may fluctuate from day to day. The average for the month shown, February 1958, was seen in Figure 8 to be 140 d/m/m³. Individual daily averages vary from 24 to 419 d/m/m³. Figure 9 also illustrates the effectiveness of an active air sampling program in getting unsatisfactory conditions corrected. The peaks during the early part of the month reflect heavy work load days in a uranium rod sawing operation which was inadequately ventilated. Note that after more efficient ventilation was provided the average level for the whole shop was much more reasonable and less subject to large fluctuations.

Figure 10 shows the monthly average air activity levels in four different types of processing opera-

tions. Note that dry chemistry, reduction, and casting operations seem to generate higher average concentrations of air-borne contamination than do machining and salvage or uranium recovery operations.

In closing, it is felt that any program short of having a full time breathing zone sampler for each employee is incomplete; however, an active, well-planned air sampling program is an invaluable tool in the control of hazards to persons working with uranium.

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2. W.F. NEUMAN, in Voegtlin and Hodge, *Pharmacology and Toxicology of Uranium*, Part IV, p. 2409, McGraw-Hill, New York, 1951.
3. National Bureau of Standards, *Handbook 52, Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water*, 1953.

Toxicology and Pharmacology - Animal Data*

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Four volumes in the National Nuclear Energy Series, entitled *The Pharmacology and Toxicology of Uranium Compounds*,[1] deal in great detail with the assigned topic. This extensive work, a compilation of the research on the biological action of uranium under the general editorship of Drs. Carl Voegtlin and Harold C. Hodge, represents the efforts of a large group of workers at Rochester over a period of four years. Indeed, in his prefatory remarks to these volumes Dr. Voegtlin states: "It is fair to say that the study of the uranium compounds herein described represents the most comprehensive experimental investigations of an industrial poison ever carried out by any group of scientific workers in such a short time."[2]

This paper has two objectives: 1) to review briefly what is known about the toxicity of uranium and 2) to describe in some detail current studies under way at Rochester involving the inhalation of atmospheres containing uranium dioxide.

Uranium, when absorbed, is a highly toxic substance. When soluble uranium compounds are given by intravenous injection, the lethal dose varies from 0.1 mg U/kg to 20 mg U/kg depending upon the species used. Uranium is thus in a class with such industrial poisons as mercury and arsenic. When soluble uranium compounds are ingested, only very small amounts (0.02 to 0.07%) are absorbed by the gastro-intestinal tract. A dietary level of 2% uranyl nitrate hexahydrate (UNO_3) will produce renal tubular injury in rats within 21 days. Generally 50 to 80% of the rats so injured recover from the kidney damage. Uranium penetrates the skin very slowly and in very small amounts. When soluble uranium compounds are inhaled, prompt absorption from the lung takes place. The amounts absorbed are quite small, particularly if the atmospheric concentration does not exceed the maximum allowable concentration (50 $\mu g/m^3$). Nevertheless, it is clear, particularly with respect to the less soluble uranium compounds, that inhalation exposures might be hazardous.

How is uranium handled by the body? It exists in the body as the hexavalent form as the uranyl ion (UO_2^{++}). The uranyl ion reacts with proteins and the bicarbonate ion to form complexes which are anions. Protein binding occurs at the carboxyl groups, and the protein complex is nondiffusible. The bicarbonate complex, which accounts for $\approx$60% of the UO_2^{++} bound, is diffusible and is the source of the injurious uranium. The bicarbonate complex is filtered by the glomerulus and enters the proximal convoluted tubule of the kidney. In the tubule, dissociation of the complex occurs, and the uranyl ion is released following resorption of the bicarbonate ion. The cellular proteins are complexed by the uranyl ion, and the cell is injured and sometimes dies. If cellular death occurs, certain cytoplasmic materials such as proteins and enzymes are released in the urine. This is the only histological damage produced by small or moderate doses of uranium.

The ultimate fate of all absorbed uranium is storage in the bone or excretion in the urine. Within one hour following the intravenous injection of a soluble uranium compound, 30% of the dose is found in bone, 30% has been excreted in the urine, 20% is found in the kidney, 19% is in soft tissues, and $<$1% remains in the blood. One month after injection 20% of the dose remains in the bone, and the balance has been removed almost entirely by urinary excretion. The work of Neuman and co-workers[3] demonstrated clearly that uranium is deposited on the surface of the bone in competition with calcium for the sites of deposition.

Let us now consider in some detail the data obtained on the inhalation toxicity of uranium compounds. For purposes of comparison, we will use data obtained with the soluble salt, uranyl nitrate hexahydrate (UNO_3), and the relatively insoluble uranium dioxide (UO_2). A number of species were exposed, but emphasis will be given to the

*Based on work performed for the Atomic Energy Project Contract W-7401-eng 49.

responses of two species in particular, the rat and the dog.

Exposures of dogs to UO_2 dust at concentrations of 20 mg U/m³ for one month or 10 mg U/m³ for one year produced the characteristic renal tubular injury. A one-year exposure of dogs to an atmospheric concentration of 1 mg U/m³ was without effect. No renal injury was observed in the rats exposed for the same period at any of these atmospheric concentrations.

Renal pathology was observed in dogs and rats exposed to atmospheric concentrations of UNO_3 ranging from 0.3 to 9.0 mg U/m³ for periods up to one year. No kidney injury was noted in rats or dogs exposed for one year to UNO_3 at an air concentration of 0.15 mg U/m³.

Small amounts of uranium were found in the lungs of rats and dogs exposed to UNO_3. For example, the lungs of rats exposed daily for one year to an atmospheric concentration of 2 mg U/m³ contained only 1.3 μg U/g lung. The femurs of these rats had an average content of 6 μg U/g bone. It has been estimated[4] that a total skeletal burden of nearly 4 g uranium would be required to constitute a radiological hazard to man. Thus, radiological injury to the bone appears unlikely.

On the other hand, rats exposed for one year to atmospheres containing UO_2 in a concentration of 10 mg U/m³ had lung concentrations ranging from 400 to 600 μg U/g. Dog lungs under similar exposure conditions contained 400 to 1200 μg U/g. Because of the high concentrations of uranium found in the lungs of the rats and dogs exposed to UO_2, the attention of the people at Rochester was shifted from their interest in the renal effects of uranium (which appear unlikely to occur at the present MAC) to the possibility that uranium storage in the lung might constitute a radiological hazard.

Table 1

U Content of Rat Tissues at the End of One-Year Exposure to UO_2 Dust (5 mg U/m³)
(Values in μg U/g fresh tissue)

Tissue	Mean*	Range
PLN	1760	60-2600
Lung	910	790-1030
Kidney	0.8	0.44-1.30
Femur	0.4	0.3 -0.7
Liver	0.07	0.03-0.09

*Mean of pooled samples from 5 rats.

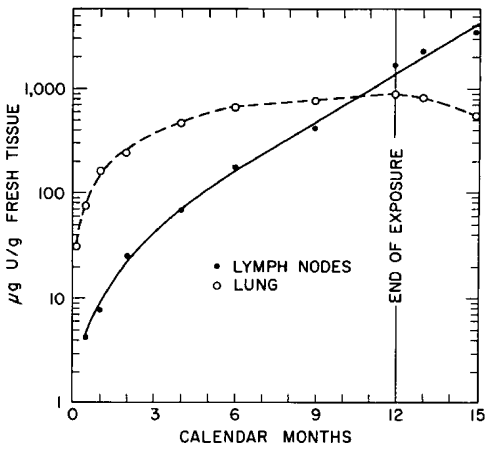

Figure 1. Uranium content of rat tissues.

In order to study the effects of continued storage of uranium in the lung and bone, a five-year inhalation experiment with UO_2 was started at Rochester. Currently the study is in its third year. The source of the data presented below is a progress report by Maynard and co-workers.[5] The construction and operation of the exposure chambers used in this study have been reported by Leach and co-workers.[6] Thomas and Hodge[7] have reported on the radiological implications of data obtained from these studies.

Rats, monkeys, and dogs were exposed daily (6 hr/day) for periods up to two years to atmospheres containing UO_2 dust. The average air concentration over the two-year period (nearly 2000 samples) was 5.7 mg UO_2/m³. The mass median diameter of the UO_2 particles was 0.98 μ. The UO_2 was dispersed into the inlet air of four chambers by means of a Wright Dust Feed; a fifth chamber was used for the control animals.

During the exposure period, serial sacrifice of the dogs was made to determine the rate of retention in the lungs and other tissues. A number of rats were sacrificed after a one-year exposure, and a number of rats were removed from the chambers and reserved for later sacrifice to obtain post-exposure data.

The uranium content of tissues of rats sacrificed after a one-year exposure is given in Table 1. The uranium content of the lungs and tracheobronchial lymph node (PLN) is markedly higher than that found in the kidney, femur, and liver. The uranium content of the rat lung and PLN at

Table 2

U Content of Dog Tissues at the End of Two-Year Exposure to UO_2 Dust (5 mg U/m^3)
(Values in μg U/g fresh tissue)

Tissue	Mean*	Range
PLN	29,000	22,000-44,000
Lung	2,200	1,800- 2,700
Femur	7.3	4.6-9.3
Kidney	6.3	6.1-6.5
Liver	1.3	0.9-1.6
Bone marrow	0.3	0.2-0.4

*Mean of pooled samples from 3 dogs.

various time periods is shown in Figure 1. The concentrations in the tissue are plotted as the logarithm of the uranium content (ordinate) against time (abscissa) plotted on a linear basis. Each point represents the average of 5 rats. As seen in this figure, the lung burden builds up fairly rapidly to 900 μg U/g and levels off at about 6 months. The uranium content of the PLN appears to lag behind for some time, but at about the 11th month it reaches the lung value. The limited postexposure data indicate that the biological half-time of uranium in the lung is of the order of 6 months. The PLN values during this period indicated continued clearance from the lung.

The uranium content in dog tissues after a two-year exposure to UO_2 is given in Table 2. Here again extremely high values are seen in the lungs and PLN. In comparison, the uranium content of the femur, kidney, spleen, and bone marrow is very low. An interesting pair of curves (Figure 2) is obtained when the lung and PLN concentrations of uranium are plotted logarithmically (ordinate) against the exposure time on a linear basis (abscissa). A very rapid build-up of uranium occurs in the lungs and PLN within the first 3 or 4 months. The lung build-up begins to level off after 6 months of exposure, whereas the uranium content of the PLN continues to increase at a markedly higher rate. The lag in the build-up of uranium in the lymph nodes observed in the rat does not occur in the dog.

It should be noted at this point that, although complete histopathological studies were performed on both the rats and dogs, no tissue changes were observed in any of the animals examined. In spite of the fact that uranium can be seen in histological preparations of the lymph node, and at times makes up as much as 15% of the weight of this organ, no histopathological changes were seen.

It was not planned to sacrifice the monkeys exposed in this study until the five-year exposure was completed. Unfortunately two monkeys became ill; these were sacrificed after approximately two years of exposure. For purposes of comparison, the mean lung and PLN values obtained from the monkeys are given in Table 3 along with the values obtained from dogs sacrificed after two years of exposure. While the uranium burden of the monkey lung is only $\approx$1.5 times as great as that of the dog, the uranium content of the monkey PLN is nearly 5 times as great. In spite of their large uranium content, the monkey lymph nodes do not show any evidence of histological injury.

In Table 4 are listed the radiation doses to the lung and pulmonary lymph nodes of dogs and monkeys. These were calculated by Thomas and Hodge and are discussed in detail elsewhere.[7] Assuming the tolerance dose to be 0.3 rem/week, the doses to the dog and monkey lung exceed the tolerance by factors of 40 and 60, and the doses to the PLN by factors of 400 and 1800. However, it should be noted again that no pathological effects were observed in any of these tissues. Thomas and Hodge have suggested that a proportionality seems to exist between the uranium burden of the

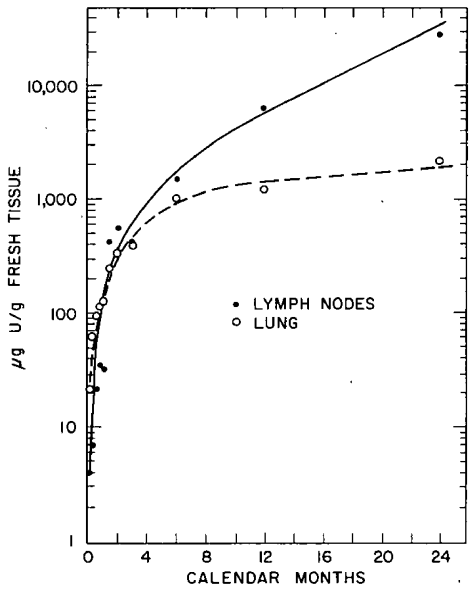

Figure 2. Uranium content of dog tissues.

dog lung and the atmospheric concentration, i.e., for a 10-fold increase in air concentration there is an approximate 10-fold increase in the lung burden. On this basis it is calculated that the lung of the worker inhaling UO_2 in the atmosphere at the MAC (50 $\mu g/m^3$) would receive a radiation dose of the order of 0.1 rem/week, assuming that man handles uranium in the same manner as the dog.

For the benefit of those who are interested in the urinary excretion of uranium, the limited data obtained in this study are given in Table 5. In inhalation exposures of this type, contamination of samples is a real problem. These animals were sacrificed under operating room conditions, including the use of surgical gowns and a triple set of instruments. The urinary concentrations of uranium reported are those obtained from samples of bladder urine taken at sacrifice. The data indicate that the urinary concentrations remain fairly constant after the first year of exposure despite the fact that the lung burden has increased by a factor of 2. The kidney content of uranium is also fairly constant after the first year of exposure, which indicates that a saturation point has been reached. Thus it appears that, under the conditions of this experiment, urinary uranium cannot be utilized as an indicater of the body burden of uranium, at least after the first year of exposure.

To summarize, we can say that when uranium is absorbed it exhibits marked toxicity. The characteristic lesion due to the chemical action of uranium is a renal tubular injury. At the present time, the data support the present MAC figure insofar as chemical toxicity is concerned. A two-year exposure of dogs and monkeys to inhaled UO_2 at $\approx$100 times the present MAC has not produced kidney injury or shown any damage that can be attributed to radiation. Urinary uranium appears to be of questionable value as a measure of the body burden of uranium, when the exposure is primarily to insoluble compounds such as uranium dioxide.

Table 3

U Content of Lung and PLN of Dogs and Monkeys at the End of Two-Year Exposure to UO_2 Dust (5 mg U/m^3) (Mean values in μg U/g fresh tissue)

No. and species	Lung	PLN
3 Dogs	2,200	29,000
2 Monkeys	2,950	142,000

Table 4[7]

Radiation Dose to the Lung and PLN of Animals Exposed for Two Years to UO_2 Dust (5 mg U/m^3)

	Rate over 2 years, rem/week	Rate at 2 years, rem/week	Calculated 2-year dose, rem
Dog lung	14	23	1,400
Dog PLN	115	310	11,500
Monkey lung	19	33	1,900
Monkey PLN	560	1510	56,000

Table 5

U Content of Lungs and Kidneys Versus Urinary U of Dogs Exposed to UO_2 Dust (5 mg U/m^3)

Exposure time, years	Total lung U, μg	Urinary U, μg/ml	Kidney U, $\mu g/g$
1	100,000	0.65	5.9
2	134,000	1.0	6.3
2.5	188,000	0.58	5.2

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Studies of Human Exposure to Uranium

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Uranium has been processed in large quantities in this country since 1942. This has resulted in varying degrees of exposure to several thousand humans. The information available on the biological effects of such exposures has been obtained to a large degree, in fact almost exclusively, from experimental work on laboratory animals. Those who have been closely associated with these humans realize that working with them is quite different from working with dogs and rats in the laboratory. Humans cannot be confined or expected to void into bottles; nor can they be put

Table 1

General Information

	Case I	Case II	Case III	Case IV
General class of work	Production	Production	Maintenance	Technical
Specific type of work	Machining	Metal casting	Painter	Supervisor
Duration of employment, years	3.7	3.8	4.8	5.2
Estimated air dust exposure, $\mu g/m^3$	35	30	30	<5
Tissues obtained from autopsy		Lung	Lung	Lung
	Kidney	Kidney	Kidney	Kidney
		Bone (rib)	Bone (vertebra)	Bone (vertebra)
	Liver	Liver	Liver	Liver
	Spleen	Spleen	Spleen	Spleen
		Adrenal fat	Heart	Testes

External Radiation Record
Exposures are in mrem determined from film badge readings.

	Case I 192 weeks*	Case II 198 weeks	Case III 250 weeks	Case IV 270 weeks
Total exposure				
beta	12,530	10,180	2,710	120
gamma	175	1,140	0	70
Average weekly exposure				
beta	65	51	11	<1
gamma	<1	6	0	<1
Highest exposure any week				
beta	220	380	110	65
gamma	80	160	0	70
Highest exposure any 13-week period				
beta	1,495	2,220	350	65
gamma	80	1,140	0	70

*Period of employment.

on controlled diets. I consider it quite proper for tolerance levels to be established on the basis of animal experiments, at least until such time as sufficient human data can be obtained. To date it appears that insufficient effort has been expended in obtaining and analyzing human data.

About two years ago we reviewed the medical and environmental data that we had obtained up to that time. This reassured us that adequate control measures were in effect to protect the health of our employees. This information had been collected for control purposes only and was satisfactory for those purposes; it was not collected with the thought of establishing exposure limits, and we believe that this older information cannot properly be put to such use. We realize that we have missed many golden opportunities to obtain human data which would be of use in establishing or confirming exposure limits.

We have now completely revised our method of obtaining information on employees working with uranium. We expect these data to be useful not only for control purposes but also for the purpose of establishing exposure limits. Conclusions based on the revised urinary excretion program will be discussed in a paper by R.C. Heatherton in Session II. The present paper will be concerned with information obtained from studies on tissues obtained from autopsies.

To date we have obtained tissue samples from six subjects, four males employed at our plant* whose deaths were unrelated to their occupations and two females who died at a neighboring hospital. The latter two had, as far as we can determine, no exposure to uranium or other radioactive materials in excess of natural background; we do not have complete medical histories on them.

On the four males we have complete medical records from their date of employment, as well as complete autopsy reports, data on exposure to external radiation and air-borne uranium, and some urinary excretion data. The available information is presented below. We realize that it is insufficient for the drawing of definite conclusions, but we feel that it is useful as a start. We hope to obtain more information of this type in the future, and by our presentation today we hope to encourage others to obtain and to present similar information.

*AEC Feed Materials Production Center operated by the National Lead Company of Ohio.

CASE I

The first case was a 36-year-old white male who worked as a machine tool operator handling normal uranium. He was employed in our fabrication plant from October 6, 1952, until his death on June 19, 1956. It was not until after his death that we learned that this man had been an alcoholic. The cause of death was assumed by the coroner to be "Antabuse reaction." Microscopic examination of the tissues did not disclose any anatomical changes which would definitely establish the cause of death. Kidney sections were read by two pathologists and reported as follows: "(1) The nephrons are well preserved; only a few fibrosed scattered glomeruli are found; (2) sections of the kidney reveal a normal architecture and hyperemia."

Complete physical examinations had been made in September 1952, August 1954, and January 1956. These revealed no significant abnormalities. The dispensary record was not remarkable except for an absence from work, April 28 to May 22, 1956, for the treatment of a "nervous condition." As we learned later, it was during this period that the patient had been placed on Antabuse therapy. Unfortunately, we did not learn of this man's death or of an autopsy until most of the tissue had been discarded. However, we did obtain blocks of liver, kidney, and spleen, upon which we attempted to do radioautographs and a chemical analysis for uranium content. As might be expected, the radioautographs were negative. The tissue analyses for uranium were technically unsatisfactory.

The estimate of this individual's exposure to air-borne uranium appears in Table1, and the laboratory findings in Table 2.

We obtained no information of the type sought from this case; however, it was not a total loss, since the lessons learned enabled us to do better on subsequent cases. We learned, for example, that it is most desirable for one of our own physicians to be present at the autopsy to obtain specimens. This means that we must be ready to report on short notice to the coroner's facilities or to the pathology sections of the hospitals. We also think it advisable to obtain fresh tissue that has not been placed in preservatives; this is important for the determination of accurate fresh tissue weights. If the tissue is placed in preservatives, it should be reweighed prior to ashing. The ashed weight must,

Table 2

Case I Laboratory Data

Urinalysis							
Date	Sp. gr.	*p*H	Albumin	Sugar	WBC/hpf	RBC/hpf	U, mg/l
9/26/52	—	alk.	neg.	neg.	0	0	—
3/ 5/54	—	6.0	neg.	neg.	rare	rare	0.018
3/ 8/54	—	5.5	neg.	neg.	rare	0	.007
4/ 9/54	—	5.5	neg.	neg.	1 - 3	1 - 2	.027
8/30/54	1.011	6.0	neg.	neg.	rare	0 - 2	.039
1/21/55	—	5.5	neg.	neg.	rare	0	.019
1/20/56	1.021	5.5	neg.	neg.	rare	0	.185

Other urine results (3/4/54 to 3/26/56): 0.042 to 1.957 U mg/l.

Hematology: Normal

of course, be recorded. In this way we can determine the amount of uranium in the ashed sample, in the preserved sample, and in the fresh tissue, the last being the real goal.

CASE II

The second case was a 54-year-old white male who worked in the metal production plant for $\approx$3½ years and in the Pilot Plant for a few weeks prior to his death. He had previously been employed for 5 years on construction at the Los Alamos Scientific Laboratory in New Mexico. He started work at our site in September 1953 and was apparently in good health until early in 1957.

On March 12, 1957, he reported to the dispensary complaining of generalized aches and pains, coughing, diarrhea, and abdominal cramps. In the course of a subsequent examination, a 14× 17 chest film was made. The report by the radiologist states: "No change from previous film. Hylar shadow on left is enlarged. Recheck film should be made in three months."

Eleven days later he reported to the dispensary at 1:05 A.M. and was seen by the nurse. On the basis of her findings and a telephone consultation with one of our physicians, it was determined that this man probably had a peptic ulcer and was hemorrhaging. He was therefore sent by ambulance to a neighboring hospital. He returned to work one month later, at which time he was transferred to the Pilot Plant and placed on limited duty. Twenty-seven days later he was brought to the dispensary, this time by ambulance, complaining of acute pain through his chest and upper abdomen. Pulse was 110, but strong; skin dry; blood pressure, 178/96. He was put to bed in the ward at the request of his own personal physician. His condition worsened, and $\approx$ 2 hr later he was transferred to a neighboring hospital where he underwent emergency surgery for a ruptured peptic ulcer. (These two episodes, in my opinion, are a high recommendation for well-trained industrial nurses. It should be noted that in these two instances this patient was seen by two different nurses.)

The perforation found at surgery was closed without difficulty. A biopsy was taken of nodules found in the liver. On the basis of the histological findings a question of metastatic bronchogenic carcinoma was raised. This was subsequently proved. It is noteworthy that these incidents occurred during the interval between March 12, 1957, and the scheduled date for a recheck x-ray examination. The patient died at 10:20 A.M. on June 19, 1957.

An autopsy was performed shortly after and was witnessed by two of our physicians. Microscopic sections of the kidney were read as follows: "Nephrons on the whole are well preserved; however, here and there one finds a partially or completely fibrosed glomerulus with dilated tubules. There is considerable cloudy swelling in the contorted portions of the tubules; pelvic epithelium is normal." Based on an examination of the tumor tissue, a diagnosis of bronchogenic oat cell carcinoma was made.

Table 3

Tissue Analyses
Units are μg uranium per g wet (preserved) tissue.

	Case II		Case III		Case IV	
	NLO*	HASL**	NLO	HASL	NLO	ANL†
Lung	0.084	0.600	1.65	0.61	0.043	0.068
	.679	.535		.78		
	.441					
	.207					
Average	0.424		1.02		0.056	
Kidney	0.085	0.057	0.023	0.026	0.006	0.010
			.077	.027		
Average	0.071		0.038		0.008	
Bone	0.0167	0.0195	0.078	0.018	—	0.034
						.036
Average	0.018		0.048		0.035	

*Analysis by National Lead Company of Ohio.
**Analysis by Health and Safety Laboratory, AEC, NYOO.
†Analysis by Argonne National Laboratory.

Sufficient amounts of tissue were obtained for us to analyze these in our own laboratory and also to send specimens to the Health and Safety Laboratory in New York. We were also able to do analyses on samples taken from several portions of the lung. The estimate of this individual's exposure to air-borne uranium is given in Table 1, the tissue analyses in Table 3, and the laboratory findings in Table 4.

CASE III

The third case was a 46-year-old white male, a painter working in all areas of the plant from August 25, 1952, to June 3, 1957. He received complete examinations in June 1952, June 1953, July 1955, and November 1956. Except for an attack of cellulitis in his left foot in 1956, he was essentially in good health until late in February 1957, when he reported to the dispensary with sciatic pain in his right leg. He was referred to his family physician. Since his condition failed to improve, he went to a chiropractor early in May 1957. He was seen again by us in the dispensary in late May and early June 1957. At about this time he was referred by his family physician to a neurosurgeon. On July 5, 1957, a laminectomy was performed and an extradural tumor removed. A second operation was performed on July 15, and a third on August 6. He remained in the hospital until his death at 10:40 A.M., August 31, 1957.

An autopsy was done shortly thereafter, attended by one of our physicians. The final diagnosis was as follows: "Extradural angioblastoma of the spinal cord" (pheochromocytoma). The autopsy report indicates a left pyelonephritis. The report on the microscopic examination is not available, but I think it will be obtained. Tissue samples were analyzed in our laboratory and in the Health and Safety Laboratory in New York. Exposure information is given in Table 1, tissue analyses in Table 3, and laboratory findings in Table 5.

CASE IV

The fourth case was a 38-year-old white male who worked for 5 years in an administrative capacity in our Technical Division. He had previously been employed in a chemical laboratory at Oak Ridge. He received complete physical ex-

Table 4

Case II Laboratory Data

URINALYSIS

Date	Sp. gr.	*p*H	Albumin	Sugar	WBC/hpf	RBC/hpf	U, mg/l
6/ 8/53	1.003	alk.	neg.	neg.	0	0	—
9/ 8/53	1.005	6.5	neg.	neg.	1 – 2	0	—
9/ 2/55	—	7.5	neg.	neg.	rare	rare	0.029
1/27/56	1.025	5.0	neg.	neg.	1 – 2		0.006

HEMATOLOGY

Date	Hb	RBC	Micro hemat.	WBC	PMN	Lymph	Mono	Eos	Baso
6/ 8/53	14.8	5.27	—	6,000	58	37	2	3	0
9/ 8/53	14.4	4.80	—	9,900	67	24	8	1	0
1/27/56	12.7	4.43	—	10,700	49	32	11	7	1
4/24/57	10.9	—	39	—	—	—	—	—	—
5/ 1/57	10.6	—	38	—	—	—	—	—	—
5/ 6/57	11.9	—	42	—	—	—	—	—	—
5/15/57	12.4	—	43	—	—	—	—	—	—

Table 5

Case III Laboratory Data

URINALYSIS

Date	Sp. gr.	*p*H	Albumin	Sugar	WBC/hpf	RBC/hpf	U, mg/l
5/23/52	1.016	<7	neg.	neg.	0	0	—
5/28/53	—	<7	neg.	neg.	rare	0	—
6/ 8/55	1.014	5.5	neg.	neg.	rare	rare	0.092
11/15/56	1.018	5.5	neg.	neg.	0 – 2	0	0.028

HEMATOLOGY: Normal

aminations in August 1952, July 1954, November 1955, and May 1957. The findings at all these examinations were essentially negative, except that in May 1957 a reversal was noted on his differential count. He had no complaints at that time. Twice during the summer of 1957 he suffered from acute upper respiratory infections for which he was treated by his family physician.

On October 30, 1957, he was referred to our Medical Department by his immediate supervisor. He was obviously acutely ill. The laboratory findings appear in Table 6. He was referred at once to a hematologist. Investigation showed marked granulocytopenia. Subsequent hospitalization and examination indicated an aplastic anemia. Terminally, he had pancystopenia. Death occurred at 12:30 A.M., December 6, 1957.

The autopsy, performed later that morning, was witnessed by one of our physicians. The final diagnosis was aplasia of the bone marrow. Microscopic examinations were reported as follows: "Kidneys – Focal extravasations of blood are shown. Here as elsewhere there is no inflammatory cell response. There is an area of hemorrhage and necrosis, and at the junction with viable tissue there is a zone of bacteria. Bone Marrow – This is essentially fatty. Some areas show practically only fat while others show a mixture of lymphocytes and possibly a few granulocytic cells. Also present in some areas are fatty macrophages. No fibrous tissue increase."

Tissue samples were analyzed in our laboratory and at the Argonne National Laboratory in Chicago. The following quotation is from the re-

Table 6

Case IV Laboratory Data

URINALYSIS

Date	Sp. gr.	*p*H	Albumin	Sugar	WBC/hpf	RBC/hpf	U, mg/l
8/11/52	—	<7	neg.	neg.	0	0	—
6/25/54	1.015	5.5	neg.	neg.	1 - 2	0	0.010
11/23/55	1.020	6.0	neg.	neg.	0 - 2	0 - 2	—
12/ 7/55	1.020	5.0	neg.	neg.	rare	rare	—
5/14/57	1.014	5.5	neg.	neg.	rare	rare	—
10/30/57	1.010	5.0	neg.	neg.	rare	rare	—
10/31/57	1.013	6.0	neg.	neg.	2 - 4	0 - 2	—
					(Casts: Rare hy.)		

HEMATOLOGY

Date	Hb	RBC	Micro hemat.	WBC	PMN	Lymph	Mono	Eos
8/11/52	14.8	4.88	—	10,800	55	42	—	3
6/25/54	14.4	4.55	—	9,800	51	40	6	3
11/23/55	14.4	4.67	—	9,800	52	45	2	1
5/14/57	14.4	—	48	9,800	36	57	2	5
10/30/57	8.1	3.32	25½	2,700	13	80	5	2
10/31/57	8.1	—	26½	1,900	10	88	2	—
11/11/57	11.2	—	33	3,500	6	92	2	—
11/13/57	—	—	—	3,300	6	90	4	—
11/15/57	10.6	3.72	—	3,100	1	99	—	—
11/18/57	9.1	3.54	29	3,400	4	96	—	—
11/20/57	9.4	3.81	31	2,500	4	96	—	—
11/21/57	8.9	3.05	27	2,900	1	99	—	—
11/22/57	8.4	3.49	28	3,800	4	96	—	—
12/ 2/57	10.9	3.43	33	2,300	—	100	—	—

port by Dr. Austin M. Brues of Argonne of their analysis of this case. "These results clearly indicate that the amount of alpha, beta and gamma emitters in the organs are within the limits of amounts normally found, and this is further verified by the uranium analyses. . . . All these examinations indicate far less than toxic amounts."

Table 7

Tissue Analyses From Unexposed Subjects
Units are μg uranium per g wet (preserved) tissue.

Tissue analyzed	Case V	CaseVI
Lung	0.089	0.006
Kidney	.026	.020
Bone (vertebra)	.028	.004
Liver	.093	.008
Spleen	.080	.005
Adrenal	.202	.006
Heart	.067	.008

Exposure data are given in Table 1, results of tissue analyses in Table 3, and laboratory findings in Table 6.

CASE V

The fifth case was a 46-year-old white female who had no known exposure to radiation or radioactive materials above background levels. Medical history and autopsy findings were not available. Tissue analyses appear in Table 7.

CASE VI

The sixth case was a 62-year-old white female with no known exposure to radiation or radioactive materials above background. Again, no medical history or autopsy report was available. Tissue data appear in Table 7.

RESULTS AND CONCLUSIONS

The amount of uranium found in analyses of the kidneys is well below the level at which we would expect to find kidney damage. The microscopic sections indicate no kidney damage which could be attributed to uranium. Analyses of lung and bone showed concentrations far below those which would be expected to cause radiation injury to these tissues. We believe that these findings substantiate the opinion given earlier that employee's deaths were in no way related to their occupations.

Earlier we indicated that there is insufficient human data upon which to base definite conclusions. However, if the assumptions in *Handbook 52* used for calculating air limits for insoluble uranium are correct, much more uranium should have been found in the lungs of Cases II and III. It appears to us that the biological retention half-time is considerably less than the assumed 120 days; probably it is of the order of 30 to 60 days. Urine results obtained in our new program support this opinion. It is interesting that Dr. Miller at Argonne has reached about the same conclusions by using the body counter. We believe that with the use of body counters at the uranium production centers, a wealth of reliable information could be obtained much more quickly and at much less cost than by the techniques now in use.

It appears to us that the kidney may be the critical organ for the types of exposures we encounter. We have come to the conclusion that far too little is known about the solubility of various uranium compounds in body fluids, particle size, and deposition and retention as functions of particle size. We are now preparing for publication a paper dealing with our findings and assumptions in greater detail.

Human Data on Uranium Exposure

A. BUTTERWORTH
U.K. Atomic Energy Authority, Springfields, England

This paper will be a practical exposition of our findings at Springfields about the effect of uranium on human beings working 8 hr per day. First, we have no evidence whatsoever to indicate that any of our several thousand employees have suffered any injury to the kidneys or other organs as a result of working with uranium at the Springfields Works. Second, we have a considerable amount of information on the relationship between uranium exposure, as air contamination, and any resultant effect on the kidneys. We have no evidence to indicate that the exposure to high air concentrations of uranium hexafluoride causes any permanent kidney damage. We have had many instances in the past, in one particular period of 12 months, when considerable numbers of individuals did have rather transient proteinuria. Immediately upon removal from exposure, the urine reverted to normal. There were probably over 50 individuals involved in this series of cases.

We have obtained some information from a human volunteer, one of my colleagues, who offered to drink a solution of uranyl nitrate and subsequently collect his urine for analysis. We made a very careful calculation of the risk involved to make sure the amount ingested would not cause any significant injury. We forgot that the stomach will rebel against such insults. However, in spite of his being made violently ill, and other symptoms, we did find that within ½ hr after ingestion of the solution he was excreting in his urine $\simeq$10 mg U per liter. At this time there was also protein in the urine. He had one attack of sickness and two attacks of diarrhea within the next 12 hr. We estimated subsequently that something of the order of 1% of uranium had been absorbed from his intestine, which must have been pretty severely stirred up. Therefore, this case does not represent in any way the type of absorption that may occur in a normal healthy nonirritated intestine. However, the results were useful inasmuch as we did find that proteinuria was absent on the third day and that the uranium excretion fell off rapidly.

We had another case of exposure to UF_6 in which the organ of absorption was the lung. Again, we found in the urine several mg U/l. On this occasion there was also proteinuria, transient as in the previous case; this individual had not previously had protein detected in the urine.

In the case of an individual who had a uranyl nitrate burn of the back, the amount of uranium absorbed through the injury was considerable. Again, we found just over 1 mg U/l in the urine, but in this case there was no proteinuria.

The above were cases dealt with routinely; the information obtained was from accidents and from normal industrial experience. This is the only usual way of obtaining information in the production plant. However, in 1953 we decided to obtain post-mortem material from employees autopsied after death. We arranged with our two main local hospitals to be informed if a death occurred among our employees, so that we could be present at any appropriate post-mortem. By 1954 the system was working fairly well. In 1954 we had the first case (D/46, Table 1) in which we were confident that the analytical data were sufficiently accurate to be used as a reasonable basis for calculations. This was an employee who had worked in the uranium hexafluoride plant for 5 years. He died of coronary thrombosis 12 hr after leaving work, and so had been exposed up to a few hours before death. His previous medical history had been perfectly satisfactory. The kidney and lung were analysed for uranium. The tissues were not preserved, but either used fresh or frozen for not more than two days. The kidney uranium content, in μg U/g wet tissue (i.e., ppm) was 0.08; in the lung it was 0.15. Subsequently we did some calculations based on these figures, which will be discussed below.

The next case (D/75, Table 1) was a man who had been an electrician at our plant for 3½ years. He died of lung cancer. Unfortunately, he was

seriously ill for 6 months before death, and therefore was not at work for that period. Analysis for uranium in the lung showed 0.05 ppm and 0.05 ppm, and in the kidney 0.04 ppm and 0.05 ppm.

The next case (D/87, Table 1) was an employee who had worked for about 9 years as a changeroom attendant in UF_4, UF_6, and UO_2 areas. The results were of the same order in the kidney, 0.04 ppm U. The lung contained 0.20 ppm U, one of the highest results obtained.

To date we have had 7 post-mortems; all the results are given in Table 1. The last one was an individual who worked for us up to the time of his death. He was a changeroom attendant for 5½ years, working in UF_4 and UO_2 areas. In this case the lymph glands were also analysed because quite recently we became interested in the lymph glands as a result of reports received from the United States. We found in the lung 0.05 ppm and in the lymph nodes 0.12 ppm. These results are interesting although inconclusive.

We are at present in the process of reviewing our standards in this field. Obviously much more work must be done, and we will have to cooperate more closely with our local hospitals in order to continue to obtain this type of information. It does appear that our orders of magnitude are the same as those Dr. Quigley has presented in the preceding paper.

We have also done analyses on a number of post-mortem samples obtained from individuals who had never worked with uranium or been exposed to it. These are our control figures, and are shown in Table 2. The concentration of uranium in the kidney of nonexposed individuals appears to be of the order of 0.02 ppm wet weight, and in the lung of the order of 0.04 ppm. This investigation is also being continued; however, it is interesting that these orders of magnitude are also similar to Dr. Quigley's.

In view of the uranium content of the nonexposed kidney, it is probable that a proportion of the uranium detected in the kidney specimen of case D/46 was of nonoccupational origin. Although this fact will be ignored in subsequent calculations, it should nevertheless be kept in mind. The uranium exposure of this individual was regular and of long duration. It is therefore reasonable to assume the existence of equilibrium conditions in the kidneys, since the biological half-time of renal uranium is relatively short.

With an average daily exposure of 14 $\mu g/m^3$, 28.5 ± 11.4 μg uranium were found in the kidneys. If the average daily exposure had been at the maximum permissible concentration of 270 $\mu g/m^3$, then we would have expected $28.5 \times 270/14 = 550 \pm 220$ μg U. The recommendations of the International Commission on Radiological Protec-

Table 1

Analysis of Uranium Content of Human Pathological Material, Springfields Cases

Case No.	Age, years	Sex	Cause of death	Uranium content, ppm wet weight of organ specified				Occupation	Duration, years	Occupational exposure to radioactivity
				Kidney	Liver	Lung	Heart			
D/46	52	M	Coronary thrombosis	0.08	0.04	0.15		Process worker with UF_4 and UF_6	≈5	Yes
D/75	43	M	Lung cancer	0.04		0.05		Electrician	3½	Yes
				0.05		0.05				
D/87	58	M	Coronary occlusion	0.04		0.20		Changeroom attendant,		
				0.03		0.18		UF_4, UF_6, U metal	8½	Yes
D/91	60	M	Cirrhosis of liver	0.02		0.03		Stocktaker	11½	Yes
				0.02		0.02				
D/95	54	F	Coal gas poisoning	0.02	0.01	0.06	0.01	Sewing room assistant	11½	No
				0.03	0.01	0.07	0.01			
D/97	48	M	Coronary occlusion	0.04	0.02	0.05	0.02	Laboratory attendant	3	Yes
				0.03	0.02	0.05	0.02			
D/99	59	M	Postoperative septicemia	0.06	0.04	0.05		Changeroom attendant,		
				0.06	0.05	0.05	0.12*	UF_4, U metal	5½	Yes

*Mediastinal lymph glands.

Table 2

Analysis of Uranium Content of Human Pathological Material From Hospital Cases With No Occupational Exposure to Uranium, Causes of Death Not Known

	Uranium content, ppm wet weight of organ specified				
Case No.	Kidney	Lung	Liver	Heart	Spleen
C.1		0.05 0.06	0.02 0.02		
C.2	0.02			0.13	0.01
C.3-C.9	0.02, 0.03 0.01, 0.01 0.01, 0.01 0.01, 0.01 0.01 0.01, 0.01				
C.10-C.12		0.02, 0.02 0.02, 0.02 0.04, 0.04			

tion specify the maximum permissible body burden of soluble uranium as 0.04 μC = 120 mg, of which 6.5% = 7800 μg is located in the kidneys. In the above calculation, however, the expected amount of uranium is considerably less.

Assuming that 10 m^3 air are breathed during a working day, the average daily lung intake of uranium was $14 \times 10 = 140$ μg/day. The recommendations of the International Commission on Radiological Protection specify that 8% of soluble uranium inhaled is retained in the kidneys. Therefore the quantity of renal uranium that should have been found in this case may be calculated thus:

Mass of uranium inhaled per day = 140 μg
Fraction retained in kidneys = 0.08
Biological half-time of renal uranium = 30 days (I.C.R.P. value)
Mean half-time of renal uranium = $\approx$45 days
$\therefore$ Equilibrium value of renal uranium = $140 \times 45 \times 0.08 = 504$ μg

The quantity of uranium actually found on analysis was certainly many times smaller. It would seem, therefore, that the renal retention of soluble uranium is less than that suggested in the recommendations of the International Commission on Radiological Protection.

A lung uranium content of 150 ± 60 μg arose from a daily exposure to 14 $\mu g/m^3$. If the daily exposure had been at the maximum permissible concentration (270 $\mu g/m^3$), the lung would have contained $150 \times 270/14 = 2850 \pm 1156$ μg U.

The recommendations of the International Commission on Radiological Protection specify the maximum permissible body burden of *insoluble* uranium to be 0.01 μC = 30 mg, the lung being the critical organ in this instance. This figure is 10 times greater than that given above, which suggests that a maximum permissible lung burden of soluble *natural* uranium cannot occur, since renal damage from chemotoxicity would necessitate removal from exposure before such a quantity of uranium could accumulate.

The time of death in case D/46 was a mere 12 hr after the last exposure to uranium. An average of 140 μg uranium were inhaled per day, and 150 ± 60 μg were found in the lungs; hence it appears that the biological half-time of soluble uranium in the lungs is of the order of one day.

We have another investigation in progress with some rabbits. We are not directly experimenting with these rabbits; they live in our factory in an area where old uranium residues are stored. They are sacrificed at intervals for analysis of various tissues. This investigation has been in progress for only 9 months, and as yet only a few analyses have been done. The results are shown in Table 3. No particularly significant histology was found in the rabbit tissue of the heart, lungs, liver, kidneys, or bone. However, there were two interesting analytical findings in the case of kidney and bone, which we have not yet tried to explain. The concentration of uranium in the kidney of these rabbits is quite high compared with that of the single control rabbit, and also compared with that of our human cases. It varies between 0.46 and 1.4 ppm, the lower value being about 10 times as high as in human cases; the control rabbit showed 0.05 ppm. The uranium content of the bone ranged from 0.22 to 1.1 ppm, the control showing 0.03 ppm. We do not have comparative figures for bone in humans. These figures are interesting, but we have not had time to consider them in detail or to analyse a sufficient number of specimens.

Finally, I would like to quote some of the conclusions reached in the report of case D/46 prepared by my colleague Dr. McLean and myself. (This was our first post-mortem case, who had died of coronary thrombosis within a few hours of ceasing work.) Note that the conclusions are drawn only from this one case. We said, "Soluble uranium in the form of UO_2F_2 does not appear to accumulate in the kidneys to any significant extent. Soluble natural uranium should be con-

Table 3

Chemical and Histological Examination of Rabbit Tissue, Springfields Works, 1958

	Rabbit No. 1	Rabbit No. 2	Rabbit No. 3
Heart			
Total weight of organ	7.3 g	9.0 g	5.0 g
Weight taken	6.1 g	7.2 g	4.1 g
Uranium content	0.02 ppm	0.10 ppm	0.02 ppm
Histology	There are occasional small foci of round cells in the intramuscular septa, mural blood vessels show sclerosis and occasionally dilatation.	Appearance normal.	Low grade chronic interstitial myocarditis.
Lungs (2)			
Total weight of organs	12.4 g	11.5 g	12.1 g
Weight taken	10.7 g	7.2 g	10.3 g
Uranium content	0.06 ppm	0.10 ppm	0.11 ppm
Histology	Alvedi are dilated and in some cases have ruptured so that several together form large irregular spaces with remnants of alveolar walls projecting into lumen; bronchioles are dilated, denuded of epithelium, the walls edematous and sparsely infiltrated with round cells. Arteries and branches show fibrous thickening of the walls with some perivascular round cell infiltration.	Appearances of alveolar emphysema, passive congestion and chronic bronchitis.	Appearances of alveolar emphysema with edema, chronic bronchitis and passive congestion.
Liver			
Total weight of organ	57.5 g	68 g	61.2 g
Weight taken	26.5 g	32 g	30 g
Uranium content	0.04 ppm	0.10 ppm	0.04 ppm
Histology	Parenchymal cells appear normal. Vascular changes are present as in other organs.	Appears normal.	Passive congestion only.
Kidneys (2)			
Total weight of organs	10.5 g	9.6 g	10.9 g
Weight taken	8.5 g	8.5 g	7.9 g
Uranium content	0.52 ppm	1.4 ppm	0.62 ppm
Histology	There may be a slight dilatation of the pelvices and there is slight passive congestion, proximal and distal tubules and glomeruli appear normal. There is sclerosis of arterial walls. Interstitial tissues do not appear increased nor is there significant evidence of inflammation.	There is some passive congestion, but interstitial tissue does not appear increased nor is there significant evidence of inflammation.	Some dilatation of pelvis and calyces, there is passive congestion and sparse round-cell infiltration. Interstitial tissue does not appear increased nor is there marked inflammatory change.
Bone			
Total weight	16.5 g	15.5 g	13.2 g
Weight taken	13.5 g	13.1 g	12.1 g
Uranium content	0.22 ppm	1.1 ppm	0.26 ppm
Histology	Development of all cell series appears normal.	Development of all cell series appears normal.	Development of all cell series appears normal.

Table 3

Chemical and Histological Examination of Rabbit Tissue, Springfields Works, 1958

	Rabbit No. 4	Rabbit No. 5	Control Rabbit
Heart			
Total weight of organ	10.4 g	8.7 g	7.9 g
Weight taken	8.7 g	6.8 g	6.7 g
Uranium content	0.03 ppm	0.28 ppm - 0.21 ppm	0.07 ppm
Histology	Low grade chronic interstitial myocarditis.	No evidence of disease.	Appears normal.
Lungs (2)			
Total weight of organs	8.2 g	15 g	12.5 g
Weight taken	7.1 g	13.5 g	10.3 g
Uranium content	0.06 ppm	0.12 ppm	0.05 ppm
Histology	Appearances of alveolar emphysema and broncho-pneumonia.	Appearances of alveolar emphysema with congestion; chronic bronchitis. Bronchiolitis and interstitial pneumonia. There is infestation with an animal parasite, probably helminthic.	Appearances of alveolar emphysema with some congestion, also low-grade chronic interstitial pneumonia.
Liver			
Total weight of organ	62.5 g	62.2 g	56 g
Weight taken	30 g	30 g	27 g
Uranium content	0.02 ppm	0.07 ppm	0.03 ppm
Histology	Passive congestion and chronic cholangitis.	Some increase of fibrous stroma in portal tracts with round cell infiltration in places. There is one pyogenic focus of obscure origin.	No evidence of disease.
Kidneys (2)			
Total weight of organs	10.6 g	12.8 g	12.6 g
Weight taken	9.5 g	11.6 g	11.1 g
Uranium content	0.46 ppm	0.65 ppm	0.05 ppm
Histology	Appears normal apart from passive congestion.	Some degeneration of proximal and distal tubular epithelium, glomeruli appear normal. Interstitial connective tissue is not increased nor is there inflammatory change.	Appear normal. Interstitial connective tissue is not increased nor is there inflammatory change.
Bone			
Total weight	13.0 g	15.7 g	16.8 g
Weight taken	11.2 g	14.8 g	16.2 g
Uranium content	0.33 ppm	0.43 ppm	0.03 ppm
Histology	Development of all cell series appears normal.	Development of all cell series appears normal.	Development of all cell series appears normal.

sidered as a chemotoxic rather than a radiotoxic agent. Radiotoxicity is probably of importance only when the specific activity is increased by a factor of 10 or more. It is improbable that either the kidneys or lungs can retain a maximum permissible amount of soluble natural uranium, since the limiting factor is the renal sensitivity to the chemotoxic effect. It appears that the proportion of UO_2F_2 absorbed into the circulation from the lungs is greater than the 25% specified in the recommendations of the International Commission on Radiological Protection. The biological half-time of UO_2F_2 in the lungs is of the order of one day. The biological half-time of soluble uranium in the kidneys appears to be considerably less than 30 days and probably does not exceed 15 days."

In conclusion, as Dr. Quigley has said, it is important for all production establishments to obtain as much information as possible on human as well as on animal material, and in order to do this it is of vital importance to plan in advance. Had we planned better in 1953 and 1954, we might have more information available now.

Discussion of Session I Papers

Chairman, W.B. HARRIS

QUESTION: On the seven cases you have autopsied, Dr. Butterworth, do you have any idea of the average air dust exposure of these people?

BUTTERWORTH: We haven't attempted to calculate these air dust exposures, except in one case (the first autopsy), since the sampling procedures were such that not even a good estimate could be made. We have the information for the one case because this man had worked in a plant where the air sampling program was virtually on a 24-hr per day, 7-day per week basis.

I have a question for Mr. Harris. In your introduction, when you were talking about the onset of carcinogenesis, you said one doesn't have to wait 15 years. I did not understand the point you were making.

HARRIS: It probably is not a very important point. I was merely attempting to show that people who have radioactive material deposited in the body in many cases show first symptoms at a very early date. It does not appear, from the information obtained from radium exposures, that it is necessary that the radioactive exposure of tissue be for 15 or 20 or 25 years; some of the people are more sensitive and therefore will show symptoms within a short time. Granted that the data presented were sketchy, it is still interesting that the people who showed symptoms at the earliest time had material in their body at the lowest level of all the people involved. I am sure this is not statistically significant, but it was observed in this particular study.

QUESTION: It seemed to me that Mr. Harris implied that there was a relationship, perhaps an inverse relationship. However, later experiments completely contravene this implication as far as carcinogenesis is concerned in the lung. I can speak from my own results along these lines. It seems to me that you have to study a subject over a period of years to exclude the possibility of late development. Therefore, I would say that I reject what was said.

HARRIS: I accept the rejection of the implication. I merely stated that in this study the people who showed the earliest symptoms were not in the highest exposure group.

STOKINGER: I am disturbed by the relatively high uranium values in tissue obtained from supposedly unexposed individuals compared to the values for people who had been exposed. Could Dr. Quigley explain how these values were obtained?

QUIGLEY: These were samples obtained on autopsy.

STOKINGER: Do you have any idea of the medical history?

QUIGLEY: No; we were a little surprised ourselves by the results. There was no medical history or autopsy report available except for those I mentioned. The results we have now are not complete. We don't know whether they came from people with upper or lower possible exposures.

I noticed that in the talks exposures were given in various units. At our plant we consider 70 $d/m/m^3$ to be equivalent to 50 $\mu g/m^3$ in air. Mr. Breslin spoke of an air MAC of 110 $d/m/m^3$. I think there is some confusion in the reporting of human data, and we should attempt to make it uniform.

MASON, M.: I would like to hear from the physicians who are here. I am not a medical man, but I handle the Workmen's Compensation cases as well as the health problems in our plant. During the last 5 years we have had a great deal of trouble with the healing of fractures of the bones in the feet and hands. It seems that every time we have had a clean break of any of the bones, we have been unable to get it to heal without having the surgeon put a spike in it.

Have you found it to be true for people who have had relatively long exposure, that after some kind of trauma they have developed symptoms of arthritis? I have talked this over a great deal with our physicians. They have examined each individual case and considered the cumulative cases, and they feel that there is no relationship, but frankly they are not sure. I just suggest that industrial physicians should watch such cases to see if there

is any such indication. We watch ours quite closely, but I have no explanation to offer. However, it is interesting that in 4 to 5 years of experience we find that we have more trouble in getting bones to knit properly.

BUTTERWORTH: Maybe this is connected with the age of the employees.

MASON, M.: I think the average age is probably in the range of 38 years, which is not very old.

HARRIS: Has this observation been checked against normals in the local population?

MASON, M.: A statistical study has not been made. This is a question of a general impression. The physicians agree with me that it looks a little unusual, but we haven't been able to draw any conclusions from it.

QUESTION: Is there a lack of callus formation?

MASON, M.: We have had about 5 cases in the last 3 years in which the bone did not knit at all and we have had to use bone grafts. In one or two cases we have spiked the bones together.

QUESTION: In long bones?

MASON, M.: In the hands and feet, the long bones, yes. We have not had any breaks of the arm, so I have no information about that.

QUESTION: It seems to me that we have good enough analyses from the urine, but I have been concerned with the weight of body tissue. By the time we get the tissue from the lab, it has usually been preserved in formaldehyde. How does analysis of such tissue compare with that of fresh tissue? All the results have been given in terms of $\mu g/g$ wet tissue. What is wet tissue? I think we should standardize our definition of that.

HARRIS: It would be useful if we could standardize everything.

QUIGLEY: The results we quoted were on the basis of weight taken after preservation. In other words, the samples sent to you were preserved, and the results were not comparable with those on wet tissue. I think this is one of the things we should look into a little more. Some of the reports were on wet tissue and others were not; therefore, we do not have comparable results.

HENRY: I would like to comment on one aspect of the earlier papers, those giving large numbers of air concentrations, MAC's, etc., namely on the method by which these were determined. One report refers to averages of 10,000 or 20,000 or 2000 samples, another to averages of personal exposures, and others perhaps to averages in given areas; it would be helpful if those who gave these figures would give some indication of how their results were obtained. Are they using the normal industrial hygiene method of weight averaging, or air samples, or average breathing zones, or what?

HARRIS: I can answer for two sets of data, one collected by the Mallinckrodt organization and the other by the Health and Safety Laboratory. Both of these were collected on the basis of average exposures, weighted with time, collected from both breathing zone and general air sampling.

QUESTION: Mr. Patterson indicated that at Y-12 they use two different methods of detection, one involving a gas flow proportional counter. What was the other method of detection?

PATTERSON: In counting air samples we use an automatic alpha scintillation counter having twin scintillation detector heads, which is very convenient for processing the large number of routine general air samples. The gas flow counters are used in processing most of our special purpose samples, such as those from breathing zones.

In answer to Dr. Henry's question, the averages I show are obtained only from the routine general air program. They do not include any breathing zone or operational or other special sample results.

QUESTION: You mention special purpose samples. Do you feel that proportional counting is the better method for these?

PATTERSON: Not entirely; our choice of method is based on the numbers of samples that have to be handled in the different programs.

QUESTION: I believe Mr. Mason said he had about 100 people in his original group. Do you recall how many others there were, and the analytical results on them?

MASON, M.: The size of the original group was between 100 and 200. I don't know why we have such a stable population, but we still have about 100 of these people with us today. There has been no attempt to follow up those who have left us. This was given very serious consideration, but for many reasons, which we can all appreciate, we decided it could not be done. Some of those still with us are no longer on the uranium controlled project but are working at the main plant. Of these, 33 have been there since 1951 and have been followed on an annual examination basis. So, I would say that we have about 75 or 76 people still on uranium operations, still being exposed, and 33 who are still with us but no longer working with uranium. The remainder have not been followed.

QUESTION: When did you get the information on those you mentioned?

MASON, M.: In 1954; that is the last date for which I have any data on these.

DOWNS: When we closed the plant at Harshaw, we gave much consideration to arranging for the continuation of examinations of these people in the years to come. We examined all the possibilities that we could think of, but after talking it over with the legal people at the AEC and at Harshaw, we decided against it. First of all, the contractor who originally employed these people would have financial difficulties in making arrangements for examinations. We had thought of handling that through the AEC's New York Office. Second, it would be very difficult to follow people dispersed over the entire United States. The physical examinations would be given by different physicians with different backgrounds, and there probably would be a great lack of uniformity. We tried to work through the Veterans Administration, since many employees at Harshaw had been veterans. Again, there seemed to be a barrier we couldn't break down, people said they would be happy to help, but we just couldn't get started.

I think it is very important to get follow-up information for purposes of comparison. I think Mallinckrodt is the only place where individuals are employed who have been exposed to high levels for an appreciable period of time.

QUESTION: Don't you run into some legal problems in getting autopsies?

QUIGLEY: We have discussed this with our legal department. In the cases we had, we have worked with the pathologist who has permission to do the autopsy; this is done either by coroner's order or by the relatives giving permission at the hospital where the individual dies. The pathologist is the one who has the permission to do any examinations that are necessary, and we feel that we are operating strictly as his agent in doing chemical analyses that his laboratory is not equipped to do; as such we send copies of our results to the pathologist. Our system as a whole has never been questioned. In one case the question was brought up of whether a woman had to give permission for us to send samples to a laboratory other than our own. I don't know whether our lawyer advised her that this point had been covered, but the question was dropped.

QUESTION: I was wondering about the possibility of incurring a law suit, should high levels be found.

QUIGLEY: I think that is a chance we have to take. We were fortunate that in the four cases we had the results were low. We are not trying to select our cases; we are trying to get information. We must realize that some day the information might be used against us; that is a problem for the legal people. We as health people should try not to create problems for them, but, to do our own job, we do have to obtain the material.

BUTTERWORTH: In the United Kingdom, our arrangements are identical with those of Dr. Quigley; that is, the pathologist is the responsible person in law, and when he is given permission for a post-mortem, it is up to him to decide what information is required in order to diagnose not only the immediate cause of death but also any other possible contributing causes. In this respect, as Dr. Quigley has said, we are merely acting as a laboratory to do an analysis which he is unable to do himself.

As far as court is concerned, we have already been in court, not because of our analyses of human tissue, but in relation to a claim against the Authority by an individual that one of the deaths was caused by the deceased having worked in the Springfields plant. In the course of this inquiry in court, it came to light that these analyses had been done, and, far from being frowned upon, they were favorably regarded as being much needed in the future to supply information about the effect of uranium on humans. I have no fear of medical-legal problems concerning the taking of samples from human beings. We could, as Dr. Quigley has said, have to make admissions in court if we found higher uranium levels in some than in others.

SESSION II

the number of persons we must monitor and the frequency with which we must sample because of the chronic nature of our exposure, have almost dictated our use of this type of program.

When submitting the sample, the participant enters the times of the present and last voidings in spaces provided on the bottle identification label. From these times and the volume of the sample, the laboratory calculates a rate for the excretion of uranium. Collection stations are usually located in clothing change rooms. Samples remain under refrigeration in the collection stations for a period of from 2 to 20 hr before delivery to the laboratory. They are analyzed within 24 hr after receipt at the laboratory.

Analysis for uranium in urine is accomplished by electrodeposition of uranium from raw or undigested urine onto silver disc cathodes and subsequent counting of the alpha disintegrations from the silver discs in low background proportional counters. (See Figures 1, 2, and 3.) An index of incidental sample contamination during laboratory procedures is provided for by the plating of 3 "blanks" of synthetic urine (tribasic sodium phosphate) with each run of samples (a run being all the samples plated at one time or a maximum of 35 urine aliquants, 3 synthetic blanks, and 2 urine aliquants spiked for recovery factor control).

The technique of plating from undigested urine was developed by experimentation after it was determined some years ago that samples were becoming contaminated in the laboratory during the 16-hr acid digestion step. Again, we do not recommend this procedure. Its adoption originally was a matter of expediency; however, with the present laboratory load of several hundred samples per week, the time and laboratory space required to provide for a digestion step make the inclusion of such a step a practical impossibility. We realize and admit that this method of analysis gives rise

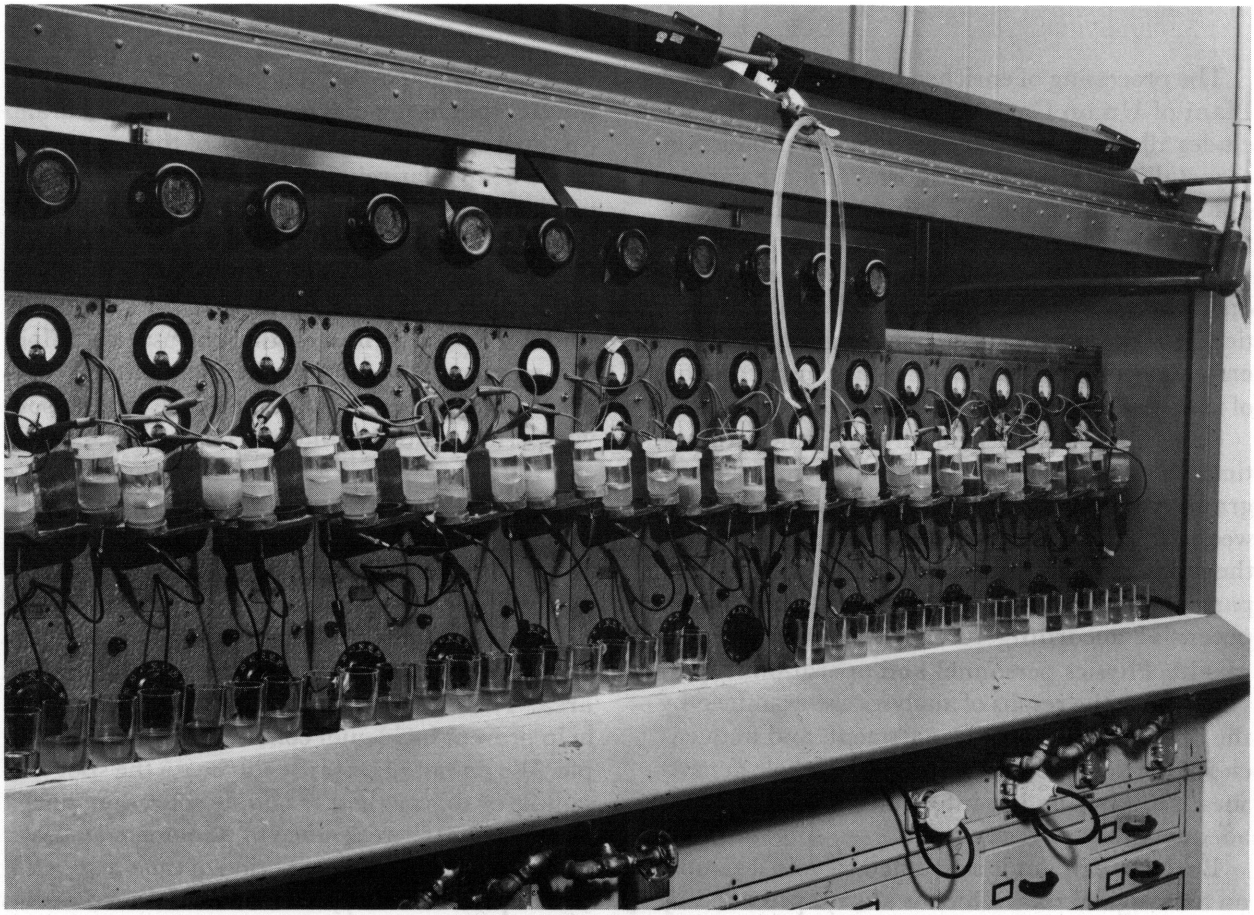

Figure 1. Plating unit in operation.

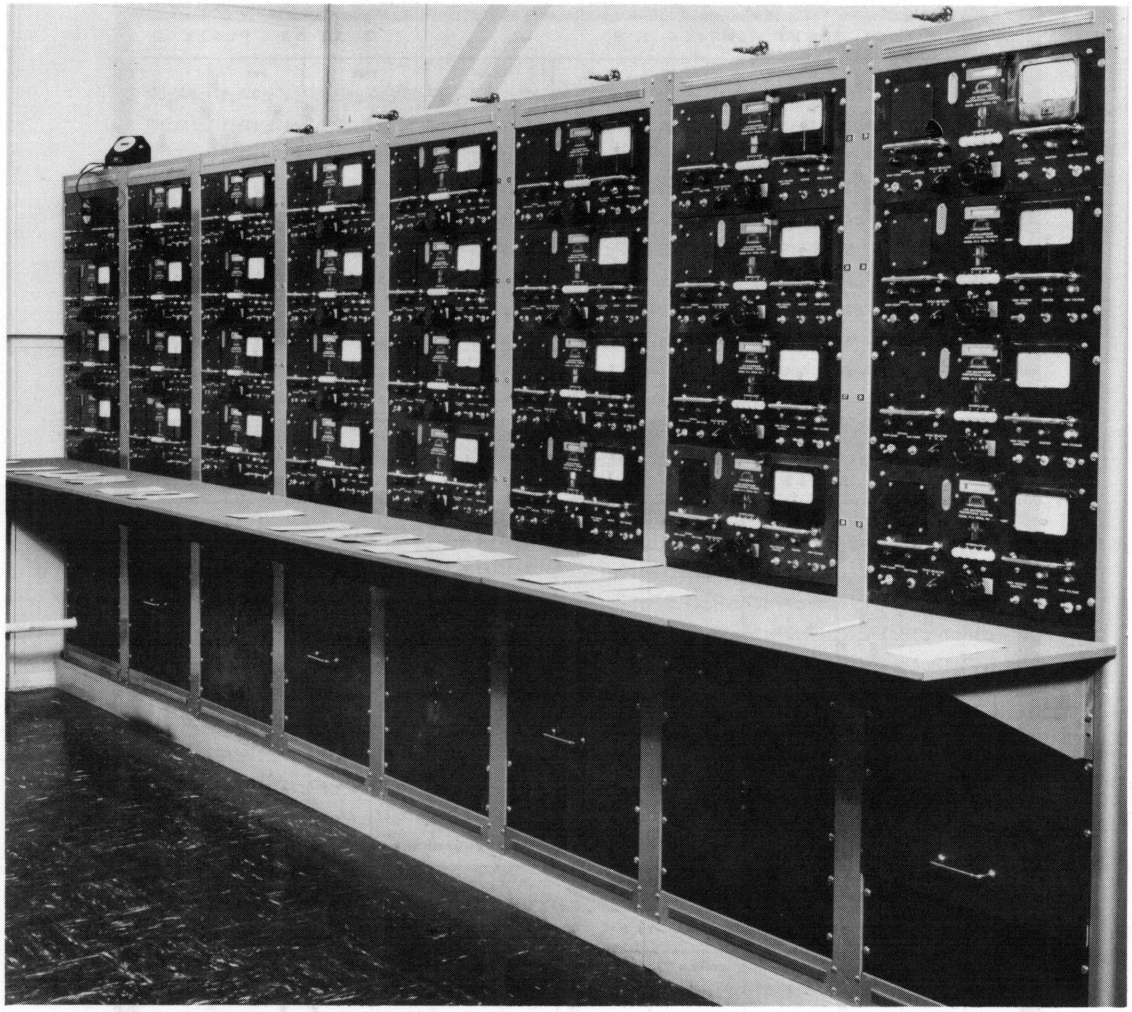

Figure 2. Urinalysis plate proportional counters.

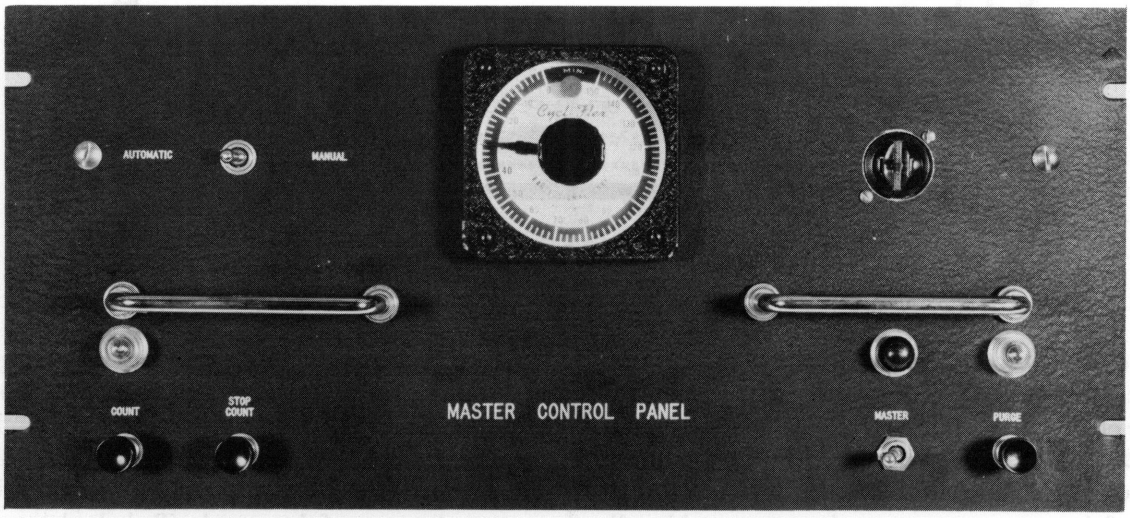

Figure 3. Master panel for control of all proportional counters simultaneously.

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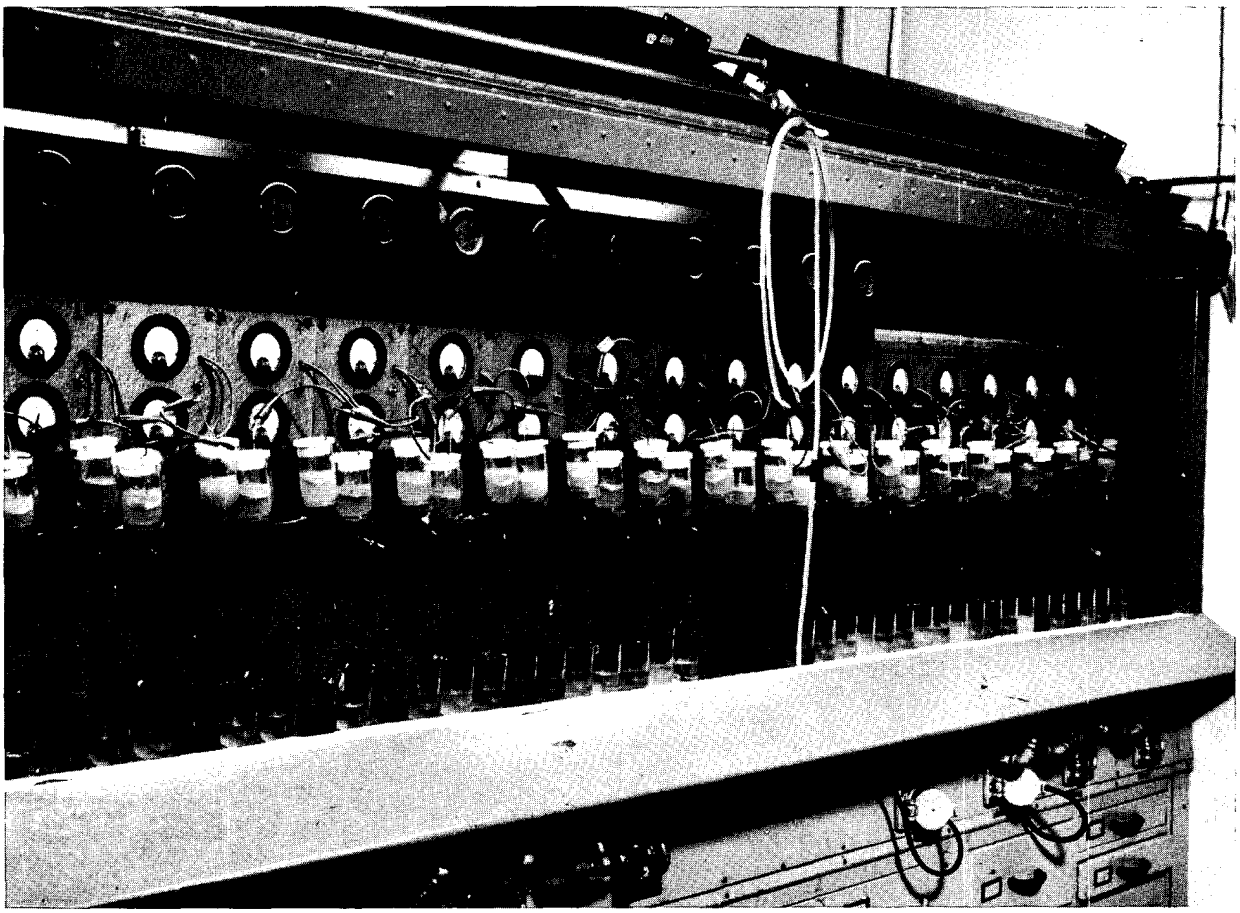

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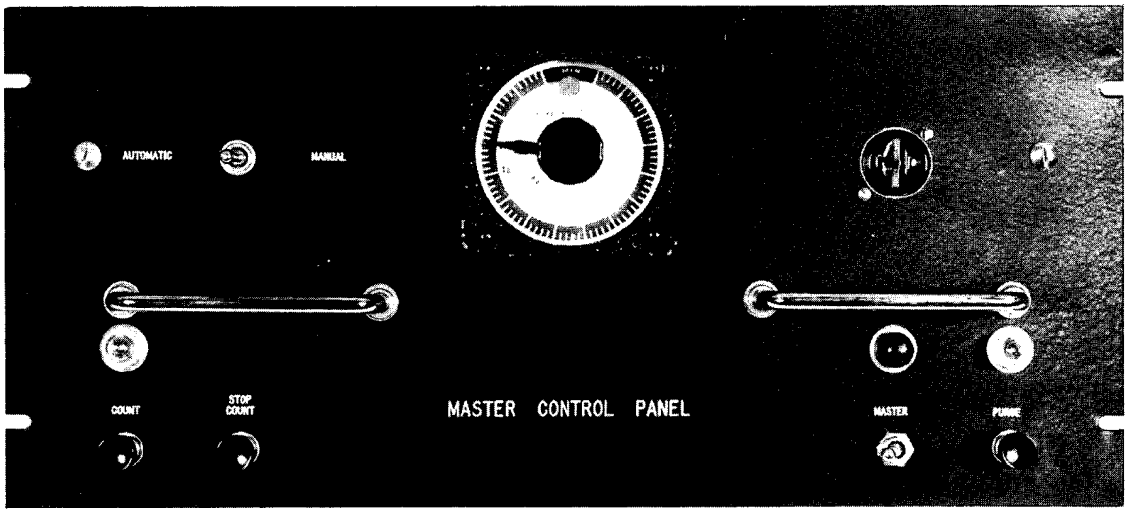

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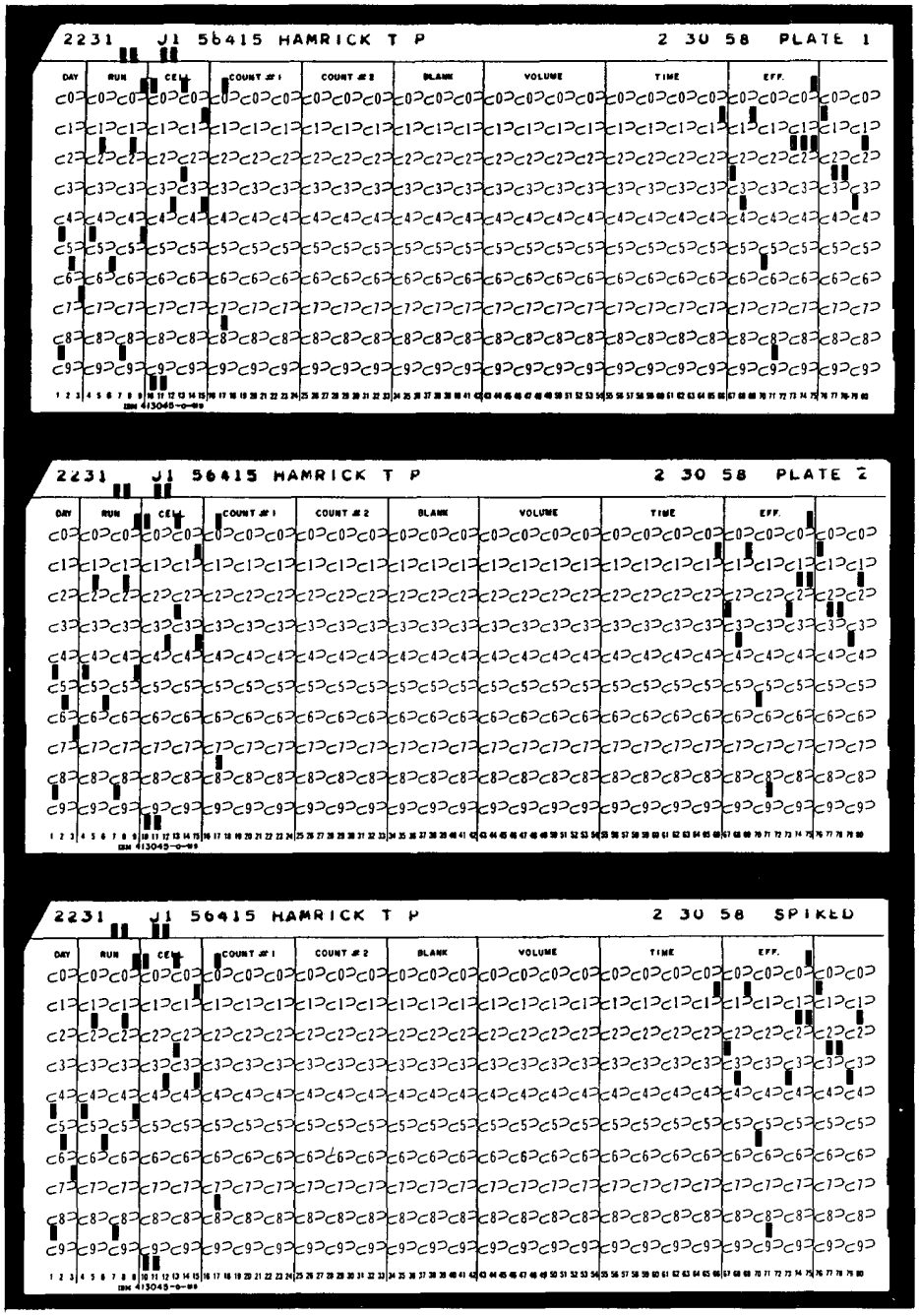

Figure 4. Urinalysis mark-sense cards.

to a relatively low order of precision in any one result; the average plating efficiency or recovery factor of a known "spike" from urine by this method is $\approx$40%. We compensate for this by the use of a ratio in our sample result calculations, i.e., the ratio of the average recovery demonstrated by all samples spiked during any one week to 100% recovery. Also, to help improve precision, our procedure calls for the plating of two discs from each sample and the counting of each disc in each of two separate counters; thus, the count determined for any urine sample is the result of four countings on two aliquants of the sample. To help determine laboratory precision, we operate an extensive

DEPT	SHIFT	BADGE	NAME	MREMS	DATE	VOL.	TIME	RATE	EFF.	PA	P1	P2	PS
				19	6-23-58	30	1.17	26	40.0	6	6		
				75<	6-30-58	65	1.17	56	40.0	11	11	11	
				70<	7-07-58	20	.67	30	40.0	19	19		
				33	7-14-58	45	1.67	27	40.0	10	9	10	
				32	7-21-58	180	6.25	29	40.0	9	10	8	
				5	7-28-58	85	2.00	43	40.0	1	1	1	
				44<	8-04-58	40	1.00	40	40.0	9	7	11	
					8-11-58NP								
				9—	8-18-58	20	1.08	19	40.0	4—	4—		
				57<	8-25-58	280	1.25	39<	40.0	12	12	11	10
				77<	*8-28-58NS	70	1.00	70	40.0	9	9	9	
				19	9-01-58	105	4.08	26	40.0	6	6	6	16
				11—	9-08-58	110	1.25	88	40.0	1—	1—	0	
					9-15-58PO								

CUM. MREM	NO. SAMPLES	AVG MREM		AVG RATE
2730	11	30	9-18-58	41

Figure 5. Urinalysis report machine tabulation sheet as sent to Health Physics Department.

quality control program and make considerable use of statistical evaluation of both laboratory and quality control determinations. Further, realizing the over-all lack of precision in any one sample result, we normally recommend restrictions only on the basis of the 13-week or quarterly cumulative internal dose as indicated by from 2 to 13 samples. This point will be discussed later in more detail.

For a more detailed treatment of our analytical and counting procedures, see the Appendix, which is an excerpt from the Y-12 Laboratory Division Procedure Manual.

In operating a bio-assay program of this size, one must have considerable help. By making maximum use of the Tabulating Services Department and IBM computers, we are able to handle with a small staff the large quantities of data involved each week.

The Health Physics Department provides the Tabulating Services Department with a participation schedule for the ensuing quarter-year showing which departments and which shifts are scheduled to submit samples on each participation day during the quarter. Once each week the Health Physics Department notifies Tabulating Services of any additions, deletions, or changes in the participation schedule. From this information the Tabulating Services Department prepares three IBM mark-sense cards for each sample scheduled to be submitted, one card for each of the two aliquants normally plated from each sample, and one card to be used if a spiked aliquant is prepared for recovery factor control. The third card is used only for 4 samples out of about 35. These cards all have the person's name, badge number, department, and shift prepunched before they are sent to the monitoring laboratory. (See Figure 4.) The prepunched cards are sent to the monitoring laboratory each week and provide a check on scheduled and nonscheduled participation. Blank cards are prepared by hand at the laboratory for samples which have been submitted out of schedule and thus have no prepunched card. All the necessary identification and analytical data are recorded on the cards, and the cards are returned to the Tabulating Services Department for calculation and tabulation of internal dose estimates. The IBM machines subtract from the four gross counts on each sample a "blank," which includes both the average incidental contamination factor and the average counter background for the week; figure the average of the four net counts; calculate the internal dose indicated by the sample; and tabulate the results of all the analyses.

The IBM tabulation sheet sent to the Health Physics Department each week gives for every person on the program the internal dose to the critical organ in units of mrem/24-hr day as indicated by each sample submitted during the most recent 13-week period, the cumulative dose for the same period, the average dose per day, and the data necessary to calculate these doses. Participation irregularities are indicated by code letters. At the end of each departmental listing totals are given for de-

partmental cumulative dose, number of participants, number of samples submitted; and number of indications greater than 43 mrem/day. (See Figure 5.) The Health Physics Department makes this same tabulation, in slightly condensed form, available to operating supervision on request.

At this point, it is pertinent to consider the derivation of our limits and internal dose calculations and their application in our program. Adjustment of Equation G-4 of *Handbook 52*[1] by use of the currently accepted value of 10 rather than 20 for the alpha RBE, and 100 ergs/gram-rad rather than 93 ergs/gram-rep, gives the formula accepted at present for the maximum permissible body burden (q). This essentially is the equation for q used in ICRP[2] recommendations and in prepublication editions of the revised *Handbook 52*. Thus, the insoluble lung-contained burden which will result in a dose to the critical organ of 0.3 rem/week is

$$q = \frac{8.4\times10^{-4}\, m}{f_2 \sum E(\mathrm{RBE})N} = 0.017\ \mu\mathrm{C} \qquad (1)$$

in which the usual notations apply: q is the body burden which contributes the given dose to the critical organ; m is the mass of the critical organ in grams (in this case 10^3 g for the lung); f_2 is the fraction of the total body content (q) which is contained in the critical organ (in the case of the lung, $f_2=1$), E is the energy (Mev) absorbed in the critical tissue per disintegration; RBE is the biological effectiveness of the alpha radiation relative to gamma (10 for alpha); N is the relative damage factor or distribution factor (1 for all organs other than bone); and the constant 8.4×10^{-4} includes the numerical constants 0.3 rem/week, d/m/μC, min/week, ergs/Mev and ergs/gram-rad.

Application of the distribution and excretion parameters of Neuman[3] and Bernard[4] gives the amount of uranium in 24-hr urine excretion indicative of this body burden as follows:

$$0.017\ \mu\mathrm{C} \times 2.22\times10^{6}\frac{\mathrm{d/m}}{\mu\mathrm{C}}\times f_2 \times\frac{0.693}{120}\times\frac{1}{2}\times\frac{2}{3}$$

$$=73.7\ \frac{\mathrm{d/m}}{24\ \mathrm{hr}}. \qquad (2)$$

In this equation, again $f_2=1$; the factor 0.693/120 is the biological decay constant or fraction of q eliminated from the lung each day; 1/2 is the fraction[3,4] absorbed in the blood of the amount eliminated from the lung; and 2/3 is the fraction[3] excreted via urine in 24 hr of the amount absorbed in the blood.

It can be seen from our application of distribution and excretion parameters that our interpretation of urinalysis results and our assignment of internal dose assume an exposure under equilibrium conditions of intake and elimination. We must make this simplifying assumption, since ours is a chronic 5-day/week exposure to atmospheres bearing uranium in small but significant quantities. Having made this assumption, we further assume that a sample which suggests an excretion of 73.7 d/m/24 hr is indicative of a dose to the critical organ of $\approx$43 mrem/24 hr ($0.3\times10^3/7=42.85$ mrem/24 hr). Thus, our estimates of cumulative internal exposure or dose are arrived at according to the following relationship:[5]

$$\mathrm{rem} = \frac{0.3}{7\times73.7}\sum \Delta t_{i-1} d_i \qquad (3)$$

in which d_i is the d/m from total uranium in 24-hr excretion; Δt_{i-1} is the time interval between measurements; and the expression $0.3/(7\times73.7)$ merely says that the MPC for urine (73.7 d/m/day) corresponds to the amount eliminated in 24 hr from a body containing a burden which delivers each day a dose 0.3/7 rem to the critical organ; in this case, the lung.

In actual practice the mrem dose/24 hr indicated by any one urine sample is estimated according to the following equation:

$$\frac{\mathrm{mrem}}{24\ \mathrm{hr}} = \frac{\mathrm{P.A.}}{30\ \mathrm{min}}\times\frac{2\ \mathrm{dis}}{\mathrm{count}}\times\frac{\mathrm{vol}}{20}$$

$$\times\frac{24\ \mathrm{hr}}{\mathrm{T.I.}}\times\frac{100\%}{\mathrm{P.Eff.}}\times0.5814 \qquad (4)$$

in which P.A. is the average net 30-min count determined from the two plates for each sample minus the corrected blank divided by 30 to give counts/min; 2 disintegrations/count is the counter efficiency; the factor volume/20 adjusts the count obtained on the 20-ml aliquant to that from the whole sample volume; the next factor adjusts the sample result from that for the time interval (T.I.) covered by the sample to that in 24-hr excretion; the factor 100%/P.Eff. adjusts to 100% recovery the count obtained from a plate at the demonstrated efficiency of our method; and the last factor converts from the d/m/24-hr excretion indicated by the sample to mrem/24 hr to the critical

organ. Essentially this factor is the number (A) necessary to satisfy the expression

$$\frac{73.7 \text{ d/m}}{24\text{-hr excretion}} \times A = \frac{42.85 \text{ mrem}}{24 \text{ hr}}.$$

However, granted the distribution and excretion parameter assumptions we make, this number (0.5814) can be shown to be mathematically correct for any given burden (q) and the corresponding dose calculated according to *Handbook 52*[1] methods.

All the constants in Equation (4) can be combined to give the simplified equation

$$\frac{\text{mrem}}{24 \text{ hr}} = \frac{\text{P.A.} \times \text{vol} \times 4.6512}{\text{P.Eff.} \times \text{T.I.}}$$

$$= \frac{\text{P.A.} \times \text{rate}}{\text{P.Eff.}} \times 4.6512. \quad (5)$$

Substituting for P.Eff. the average efficiency which has been demonstrated for our method of plating from undigested urine gives the basic equation

$$\text{mrem/24 hr} = \text{P.A.} \times \text{rate} \times 0.11628. \quad (6)$$

The 13-week cumulative then becomes

$$\frac{\text{cumulative mrem}}{\text{13 weeks}}$$

$$= \frac{\sum \text{mrem/24 hr}}{\text{No. of samples included}} \times \frac{\text{91 days}}{\text{quarter}} \quad (7)$$

or the average mrem/day as indicated by all the samples analyzed during the quarter times the number of days in the quarter.

In spite of the neatness of these convenient equations for calculating dose, we realize that the weakest link in the chain, and possibly the largest source of error in our dose evaluation (other than variances in plating and counting procedures) is the assumption we make of stabilized distribution and excretion parameters. Probably the next largest source of error is our assumption of an equilibrium condition of intake and excretion, when actually we know that the average concentration of uranium available for intake is quite variable even when that average comes from many samples over a fairly extended period. Since we cannot evaluate or very easily compensate for the error inherent in these simplifying assumptions, we attempt to compensate for the errors introduced by plating and counting variances with our considerable use of quality control and statistical analysis. Further, we may, to a certain extent, compensate for both laboratory and physiological variances by the fact that normally we report dose and take action only on the basis of the 13-week cumulative which includes results of from 2 to 13 samples. At the least, this approach makes for better statistical reliability and a more ready acceptance of our recommendations by supervisors.

At this point it will be useful to discuss our applications of statistical analysis and quality control in more detail. We evaluate our laboratory precision on the basis of both variations in plating efficiency and agreement between the counts of the two plates made from each sample.

Evaluation of plating efficiency is provided for by the processing each week of from 55 to 60 aliquants taken from regular urine samples and spiked with a known amount of uranium. As mentioned earlier, these spiked aliquants are processed along with the regular samples at the rate of 4 spiked aliquants for every 35 urine samples. Samples for spiking are chosen at random during the processing cycle, but each one chosen must have a volume adequate to provide 3 aliquants of 20 ml each. The use of prepunched mark-sense cards to record data from spiked aliquants as well as both unspiked aliquants makes it easy for the IBM machines to calculate plating efficiencies and tabulate the results for the whole week. Plating efficiency or recovery of the uranium spike is estimated according to the relationship

$$\text{P.Eff.} = \frac{\text{PS} - \text{PA}}{\text{Value of spike}} \times 100 = \% \text{ recovered} \quad (8)$$

in which P.Eff. is the plating efficiency or recovery factor; PS is the average net count of the plate from the spiked aliquant; PA is the average net count of the plates from the two unspiked aliquants; Value of spike is always 10 counts/30 min; and the factor 100 converts the fraction to percent recovered.

Every week the Statistical Services Department computes the variance and the mean of the plating efficiencies shown by the 55 to 60 spiked samples. A control chart is kept to be sure that the average plating efficiency for the week is not significantly different from the norm of 40%. (See Figure 6.) At the end of the quarter the Statistical Services Department computes the quarter mean, which is the arithmetic average of the plating efficiencies shown by some 700 or more spiked samples during the quarter. The variances found each

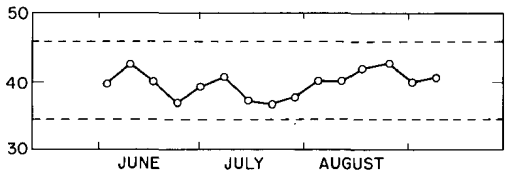

Figure 6. Plating efficiency control card.

Table 1

Bio-analysis Sample Control

No. of samples spiked	Value of spike, counts per half hour	Mean of analyses	Percent bias
699	10	4.12±0.35	3±8
793	10	4.12±0.34	1±8

week are pooled to give a better estimate of the grand variance for the quarter, the limit of error of the mean is computed, and the mean is examined for any statistically significant bias of mean efficiency to expected efficiency. The results of this inspection by the statisticians are reported each quarter in the manner shown in Table 1 for the first two quarters of 1958.

As noted in our laboratory procedure manual (see Appendix), both the two separate counts on each plate and the average net counts on both plates from any one sample must agree within the limits of good statistics, or additional plates and counts are made if sample volume permits. Further statistical analysis is given the results of all samples plated during the quarter to establish the limits of error for any one sample analysis in each of four ranges of average net counts. Our statisticians analyze the variance between the two plates for every sample submitted, calculate the limit of error which can be attached to the results of one analysis in each of the four counts ranges, and report these as percent limit of error. (See Table 2).

These values for the statistical limits of error which can be assigned to any given sample result are used to establish a confidence interval around an employee's quarterly cumulative internal dose. The calculation of this confidence interval is performed only for those whose internal dose is estimated to be greater than the 13-week maximum permissible and who are therefore being considered for removal from exposure to uranium atmospheres. Effective removal from exposure is recommended for the employee if the 67% confidence interval calculated for his quarterly cumulative dose lies entirely above the maximum permissible. The formulas for finding the limit of error and confidence interval for the cumulative are

$$\%\ \text{LE cumu.} = \frac{\sqrt{\sum(\text{LE}_{\text{individ.}})^2}}{N} \tag{9}$$

and

$$67\%\ \text{confidence interval} = \frac{\text{cumu. mrem}}{13\ \text{weeks}} \pm \left(\frac{\sqrt{[N_{0\text{-}5} \times (\text{LE}_{0\text{-}5})^2] + \ldots + [N_{>15} \times (\text{LE}_{>15})^2]}}{N_{\text{total}}} \times \frac{\text{cumu. mrem}}{100} \right). \tag{10}$$

An example taken from our urinalysis records illustrates how these are applied. The IBM sheet tabulation lists the pertinent data for the most recent 13 weeks, as shown in Table 3, from which the 13-week cumulative mrem is calculated. In this tabulation there are 6 samples in the first count range, 2 in the second, 1 in the third, and 2 in the fourth. If the number of samples in each of the count ranges and the % LE for one analysis

Table 2

Agreement Between Two Plates From Each Sample

mrem per 24 hr	Counts per half hour	No. of samples	No. of plates	Average counts per half hour	Percent LE of one analysis at 67% conf. interval level
0 - 30	0 - 5	4196	8392	2	±99
31 - 60	6 - 10	700	1400	7	±30
61 - 90	11 - 15	132	264	12	±22
>90	>15	93	186	64	±12

Table 3

mrem per 24 hr	Date	Volume	Time interval	Average counts per half hour
	6-16-58	NP		
61<	6-23-58	190	1.92	5
16	6-30-58	85	2.00	3
41	7- 7-58	235	3.50	5
40	7-14-58	690	10.67	5
—8	7-21-58	120	1.92	—1
56<	7-28-58	675	10.33	7
94<	8- 4-58	140	2.00	11
39	8-11-58	85	1.83	7
24	8-18-58	100	2.00	4
144<	8-25-58	550	9.25	20
135<	9- 1-58	350	7.00	22
	9- 8-58	NP		

Average mrem per 24 hr = 58
No. of samples = 11
Cumulative 13-week mrem = 5278

Table 4

No. of samples	Count range	% LE of one analysis	(% LE)²	$N \times (LE)^2$
6	0- 5	99	9801	58,806
2	6-10	30	900	1,800
1	11-15	22	484	484
2	>15	12	144	288
—				—
11				61,378

$$5{,}278 \pm \left(\frac{\sqrt{61{,}378}}{11} \times \frac{5{,}278}{100} \right) = 5{,}278 \pm 1{,}189$$

$$5{,}278 \quad \begin{matrix} 6{,}467 \\ 4{,}089 \end{matrix}$$

Table 5

Internal Dose Control Points

	Action point	Action taken
1.	10 counts/30 min (average net count of 2 plates)	Convert to mrem/day units immediately and report to Health Physics without waiting for normal channels of IBM tabulation.
2.	100 mrem/day	Oral report to supervisor of employee. Investigation of incident to determine probable cause, etc. Action taken depends on level of dose indicated and findings of investigation.
3.	3000 mrem cumu. per 13 weeks (arbitrary figure)	List of all persons >3000 mrem/13 weeks circulated within Health Physics Dept. Close check on work assignment of all persons on list.
4.	3900 mrem cumu. per 13 weeks	Special record kept to permit close follow-up of subsequent samples. Recommend participation with increased frequency for those participating less than once a week.
5.	4500 mrem cumu. per 13 weeks	Calculate limit of error and 67% confidence interval for 13-week cumulative. If lower limit of confidence interval is >3900 mrem, written notice to supervisor recommending removal of employee from further exposure.

in each range are applied as called for in Equation (10), the result is the 67% confidence interval for this estimate of internal dose. (See Table 4.) In this case the lower limit of the confidence interval (4089) is in excess of 3900, the maximum permissible for the quarter. The recommendation was made that this employee be assigned to a job outside uranium processing areas.

Our action or control points and some of the actions taken are listed in Table 5 and discussed briefly below.

1. The first action point is one at which the laboratory notifies the Health Physics Department in advance of routine reporting of results for samples which may indicate higher than normal exposures. Follow-up action will depend on the magnitude of the exposure indicated and other factors.

2. When the exposure indication of a sample reaches the level of action point No. 2, investigation and consultation with supervisors are indicated. (This does not, however, preclude the possibility of such investigation at levels <100 mrem/day.) Again action taken is dependent upon the magnitude of the exposure and other factors; it may include a request for special samples, use of take-home kits for longer time interval samples, investigation of operating conditions, special air samples, or any of the many follow-up actions

often used rather universally to investigate indicated exposures.

Note that the next three action points are on the basis of the 13-week or quarterly cumulative exposure. Although it has not been specifically mentioned before, our internal exposure records are kept and reported on the basis of a most recent 13-week quarter rather than a fixed calendar quarter. Thus, the action may be suggested at any time during a calendar quarter rather than only at the end.

3. The purpose of the list of persons exceeding 3000 mrem cumu./quarter is to alert the health physicists. Operating supervisors may take some action, such as changing work assignments within the department, on their own initiative after receipt of the regular IBM tabulation. A list of all persons exceeding this action point is included in the monthly Health Physics Department reports to supervision. At this level, it is usually recommended that the employee's exposure be limited or reduced.

4. The special record of results of subsequent samples is kept until the 13-week cumulative has dropped to <2000 mrem, an arbitrary figure approximately half the MPL, but one at which statistically one is confident that the exposure is below the maximum permissible limit.

5. The figure used for the action point in this case is approximately the upper limit of the 67% confidence interval for 12 to 13 samples if they have all been about the maximum permissible level. An employee who has been removed from further exposure is requested to continue submitting samples. He may be permitted to resume work in uranium processing when his 13-week cumulative has dropped to 2000 mrem if his total cumulative is no more than that which technically

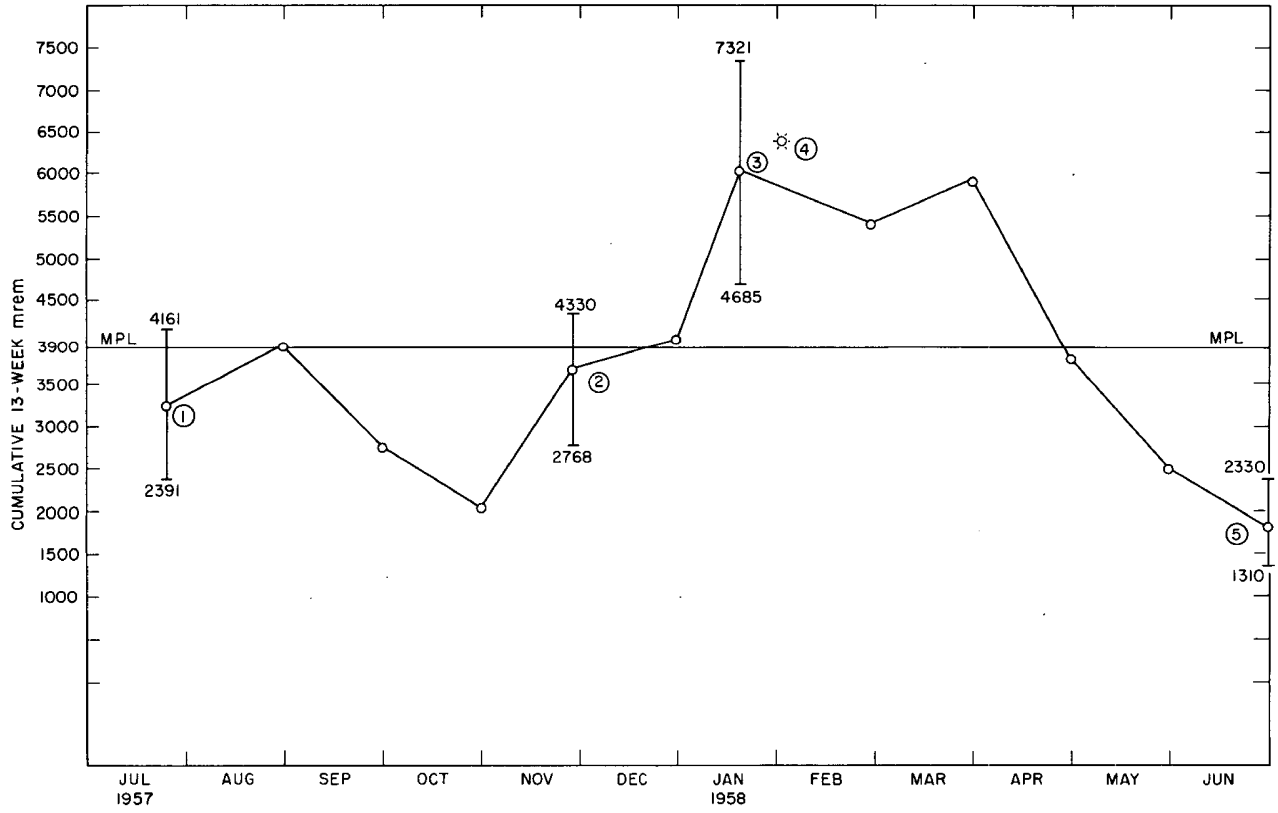

Figure 7. Urinalysis experience chart of a chemical operator in uranium salvage operations. Confidence intervals are indicated. The circled numbers indicate action points as follows: (1) Recommendation that exposure to air-borne contamination be minimized. (2) Second recommendation that exposure to air-borne contamination be minimized. (3) Recommendation for effective removal from further exposure in uranium processing. (4) Employee transferred from uranium processing areas Feb. 3, 1958. (5) One criterion met for return to uranium processing areas, i.e., 13-week cumulative is <2000 mrem.

would be the maximum permitted for his period of exposure. Only in rare cases is an employee permitted to return to uranium processing in less than 6 months from the time of his restriction.

Figures 7 to 10 are graphic presentations of both typical average experience and several atypical cases.

Figure 7 illustrates the application of action points in one case taken from our files. Sixty-seven percent confidence intervals are indicated for all points at which action was taken. Each point shown represents the cumulative mrem for the most recent 13 weeks as of that particular date; those points at which action was taken indicate the day of the month and the cumulative as of the date on which the recommendation was made, all other points are the cumulative as of that particular month. The last point shows that the employee has met the <2000-mrem/quarter criterion for returning to uranium processing work. Actually the employee was not returned to work with uranium because he had not met the two other criteria; he had been on restriction only 5 months instead of the usual minimum of 6 months, and his total cumulative exposure was still 103% of what would be the maximum permitted for his period of employment at the rate of 3900 mrem/quarter. He will not be permitted to resume work in uranium processing until his exposure record meets all three criteria.

Figure 8 shows graphically the average 13-week cumulative exposure of several departments for each month during fiscal 1958, including a total of about 250 people. It should be noted that in each of these departments the average exposure is well below the maximum permissible level. The bar graphs along the abscissa indicate the percent of all those included above who during the month exceeded the MPL (solid bar), and those whose level of exposure met the criterion for removal from further exposure (shaded bar). The numbers immediately above the bars give the actual num-

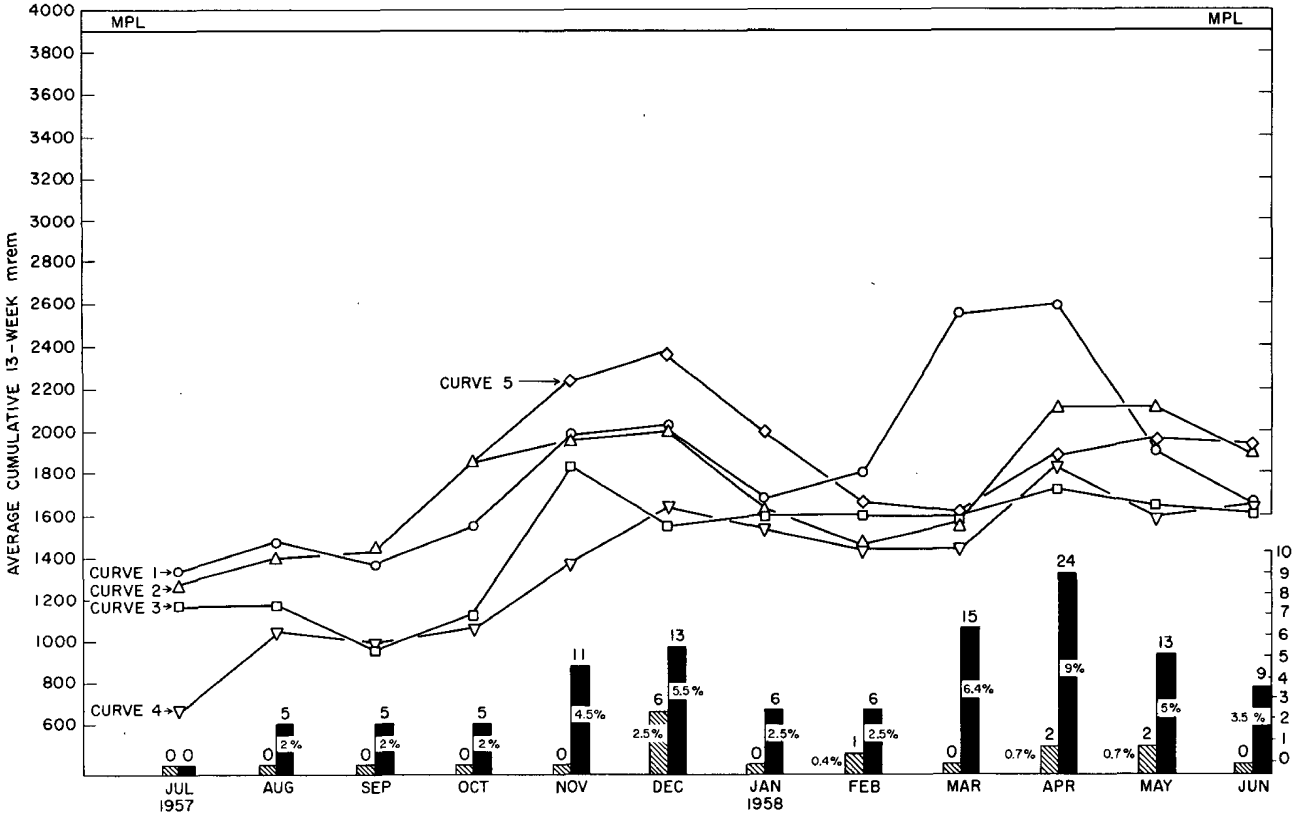

Figure 8. Average 13-week cumulative exposure for workers in salvage operations (curve 1); casting operations (curve 2); special processing (curve 3); machine shop (curve 4); and dry chemistry and reduction (curve 5). Solid bars represent persons with a 13-week cumulative >MPL (percent of all included), and shaded bars represent persons removed from uranium processing (percent of all included).

bers of persons in these categories. It can be seen that, for the most part, no persons had to be removed.

Figure 9 shows the excretion patterns demonstrated by two machinists who had inhaled insoluble uranium dusts. Investigation has suggested that the larger exposure was probably the result of failure of the employee to make proper use of the ventilation provided. It also is likely that this failure on the part of one employee contributed to the exposure experienced by the other employee, who himself may have been more conscientious. Particle studies indicated U_3O_8 of a size permitting good retention. Note how closely the two curves follow each other, even though the exposure in the upper case is some ten times the exposure in the lower case. The flattening portion of the curves indicates almost exactly the 120-day theoretical lung half-time. In the upper exposure we have had unbelievable agreement in the estimates of amount of uranium eliminated as determined by urinalysis on one hand and *in vivo* counting both at Chicago and at Y-12 on the other.

Figure 10 illustrates the excretion pattern demonstrated by a chemical operator following an exposure to soluble compounds of uranium. The initial exposure resulted from an accident in which uranyl nitrate solution spilled on the employee, causing superficial burns of the head and arm. After medical treatment the employee returned to work at normal assignments and continued to receive whatever low level exposure to atmospheric contaminants her job entailed for some 12 or more work days following the initial exposure. Thus, the excretion pattern is a rather complicated one probably involving inhalation of soluble fumes or mists and possibly some initial absorption through skin or burned tissue, further complicated by the continued low level chronic intake with the remote possibility of some continuing absorption through skin of residual contamination. Even with these complicating factors, all of which would tend

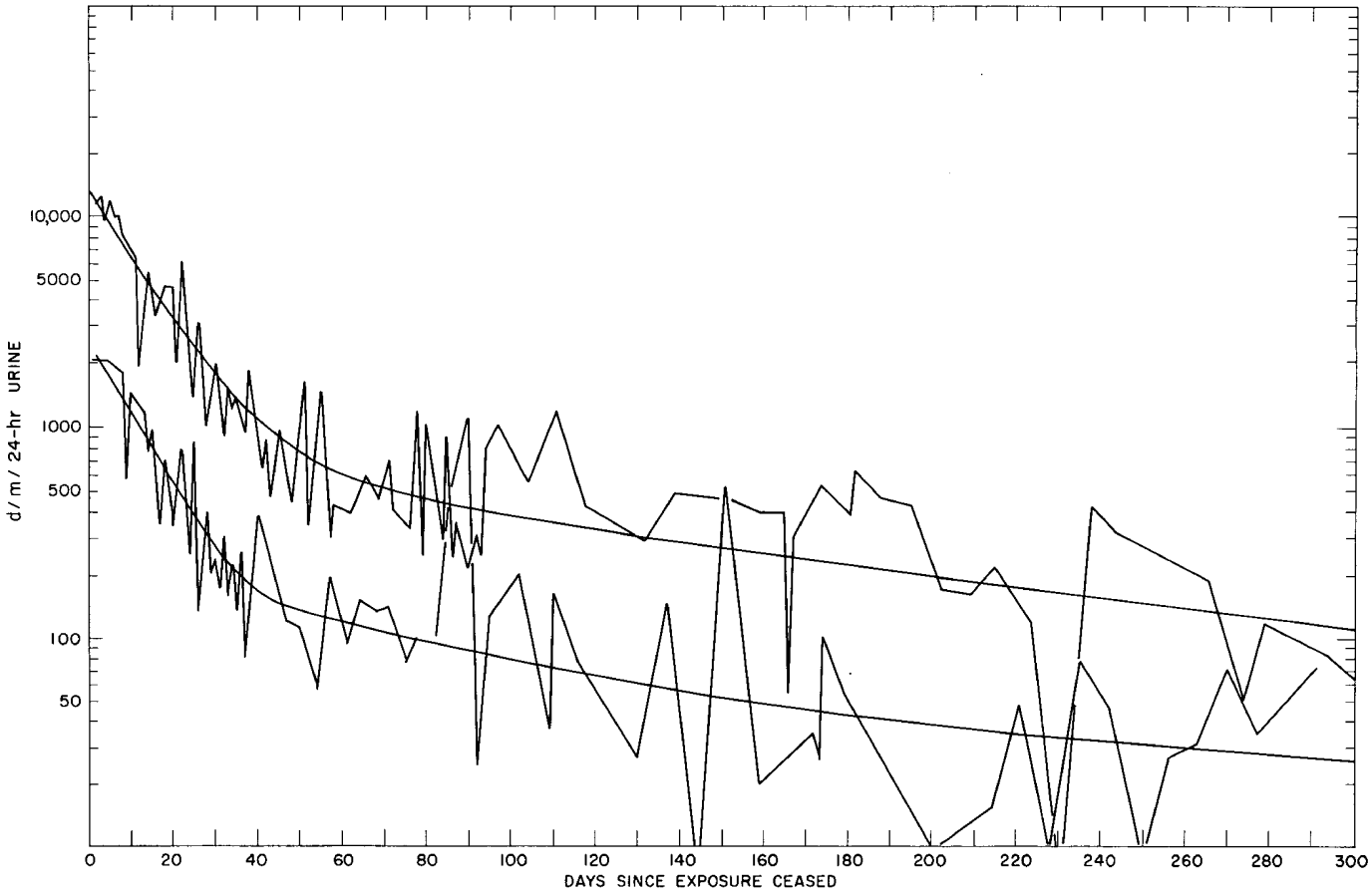

Figure 9. Elimination of a deposit of insoluble uranium from the lung by two machinists. The smooth curves were fitted visually.

to slow down the rate of decrease in sample count level, it can readily be seen that the decrease in level of urinary uranium in this case is much more rapid than in the case of the two machinists.

It is obvious that all three of these atypical cases represent more nearly single exposures than the chronic equilibrium exposure which we assume in our dose calculations. Thus, the calculation of dose for such cases by our routine methods results in an ultraconservative estimate or considerable overestimate of exposure. Evaluation of internal dose for the benefit of permanent records has been made by Mr. B.R. Fish of the Applied Radiobiology Section, Health Physics Division, Oak Ridge National Laboratory. We at Y-12 are most fortunate to have the willing assistance of such an organization.

In conclusion we realize that the compromises one must make to provide an adequate program in an operation such as ours result in a slightly less than optimum situation. However, we contend that our estimates of internal dose are quite conservative and therefore are in keeping with the philosophies of national and international authorities. Further, we feel that our urinalysis program, together with our air sampling and other programs for controlling exposure potential, provide quite adequate protection for our employees. Nevertheless, we shall continually re-evaluate our programs and strive to improve them wherever it is feasible.

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3. W.F. NEUMAN, *Urinary Uranium as a Measure of Exposure Hazard,* UR-82, The University of Rochester, 1949.
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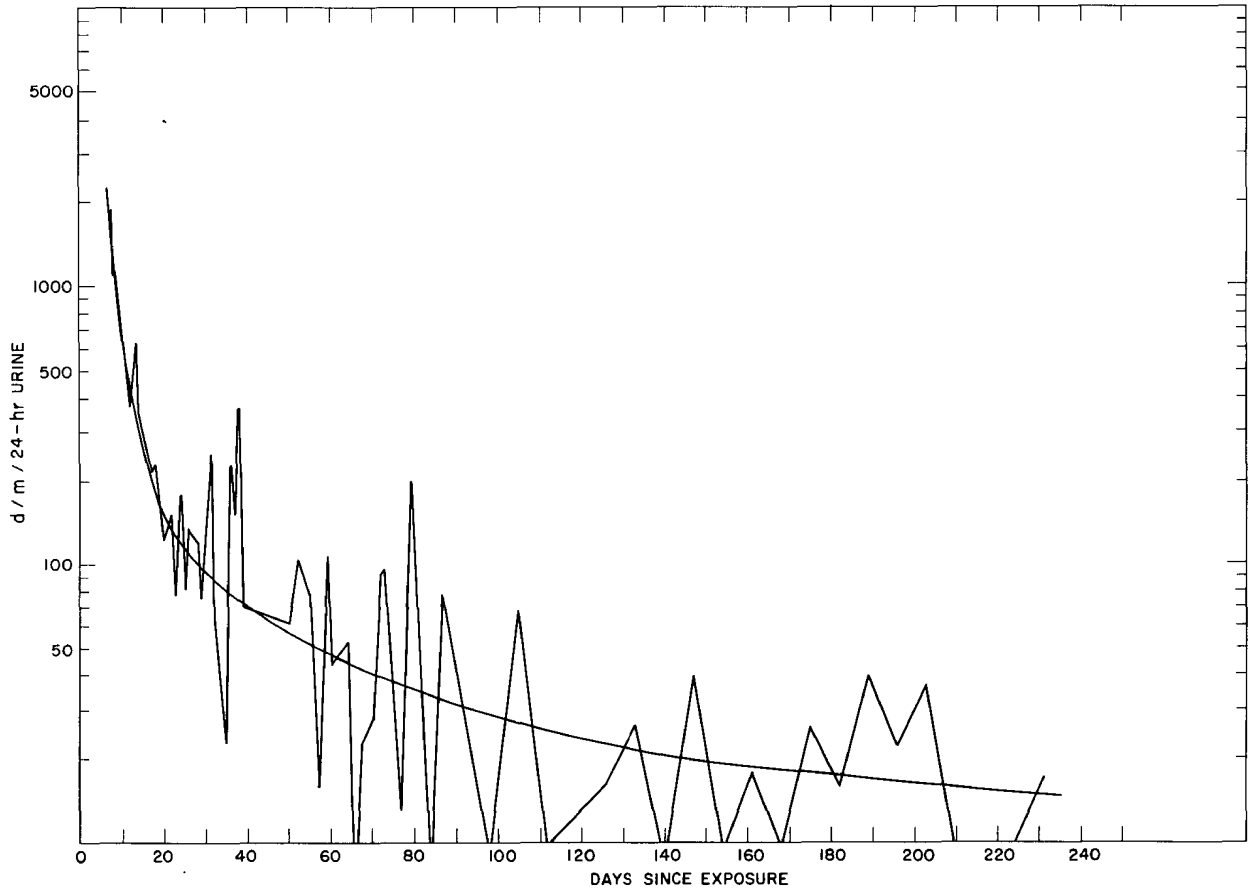

Figure 10. Excretion after exposure to uranyl nitrite spill. The smooth curve was fitted visually.

APPENDIX
ELECTRODEPOSITION PROCEDURE

A. EQUIPMENT

Brewer automatic pipette machine
Carboy, 22-liter
Electroplating cells
Coiled platinum electrodes, brass electrodes, chimneys (4-oz bottle with bottom removed), and rubber gaskets are the essential parts of the cells. The power supply produces a variable direct current up to 40 v at 3 amp.
Glassine bags
Measuring stand for determining incoming volumes
Nuclear Measurements Corporation Model PC-2 proportional counters, modified to control from a master control panel.
Silver discs, 1¾ in. diameter, 0.002 in. thick
Silver disc cutter
Vials, 20-ml plastic

B. PREPARATION AND STANDARDIZATION OF REAGENT SOLUTIONS

1. Ferric Ammonium Oxalate Solution (Electrolyte solution)

a. Compounds

Compounds	Quantity
$(NH_4)_2\ C_2O_4 \cdot H_2O$, reagent gr. crystals	453.6 g
$FeNH_4(SO_4)_2 \cdot 12H_2O$, reagent gr. crystals	3.4 g
Distilled H_2O	16,000 ml

b. The reagents are added to 4000 ml of distilled water in a 22-liter carboy. The solution is mixed thoroughly and then diluted to 16 liters. Subsequently, the solution is stirred until completely dissolved.

2. Tribasic Sodium Phosphate Solution (Synthetic Urine)

a. Compounds

Compounds	Quantity
$Na_3PO_4 \cdot 12H_2O$, reagent grade	50 g
Distilled H_2O	ca 2000 ml

b. The 50 g of reagent grade sodium phosphate is dissolved in 1500 ml of distilled water and then diluted to a volume of 2000 ml.

3. Low Level Enriched Uranium Spike Solution

a. Stock Uranium Spike Solution

1. Compounds

Compounds	Quantity
U_3O_8, enriched	0.0060 g
HNO_3, concentrated	5 ml
Distilled H_2O	ca 2000 ml

2. The accurately weighed 0.0060 g of enriched U_3O_8 is dissolved in 5 ml of concentrated nitric acid and then diluted to 2000 ml with distilled water. The pH of the solution must be less than 3.0, since uranium has a tendency to precipitate at a higher pH.

b. Working Uranium Spike Solution

1. Compounds

Compounds	Quantity
Stock U spike solution	2 ml
Distilled H_2O	998 ml

2. The 2 ml of stock uranium spike solution is diluted to 1000 ml with distilled water. This solution will count 10 to 12 counts per one-half hour per ml.

c. Standardization of Working Uranium Spike Solution

1. The solution is carefully standardized as follows: (a) Several one-ml portions of the solution are transferred with a pipette onto clean stainless steel planchets which have been previously counted to determine the background. (b) The planchets are placed under lights until the solution has evaporated to dryness. (c) Subsequently, the planchets are counted in each of three proportional counters for 30-min periods to obtain the true count value of the spike.

C. PROCEDURE

1. Cleaning of Equipment

a. All equipment that comes into immediate contact with the sample must be cleaned and stored in a manner that will minimize contamination.

b. The glass chimneys are cleaned with a solution of Tide detergent and water, then the cleaned chimneys are stored under distilled water until they are used.

c. The rubber gaskets are scrubbed with Ajax cleanser and are also stored under water until ready to use. The silver plating discs are cleaned prior to use with Ajax cleanser and stored under distilled water. The silver discs are not re-used.

d. Further, to prevent contamination of the electroplated silver discs, the electroplating cell is disassembled with the plated portion of the disc always kept downward, and, when blotting the electroplated disc dry, clean unexposed Kleenex is used.

2. Measuring and Recording

a. The volume of the urine is measured on the measuring stand; the time interval is calculated; and both results along with a sample number are recorded on an IBM card.

3. Plating

a. The electroplating cell is assembled by placing the silver disc and rubber gasket into the brass cathode and then screwing the glass chimney into position. After assembly the cell is filled with distilled water and checked for leaks.

b. A 20-ml aliquant of the urine sample is transferred to the assembled cell, and the top containing the anode is adjusted in place. Using the Brewer automatic pipette 20 ml of electrolyte solution is transferred through the hole in the top of the completely assembled cell and then distilled water is added up to the etched ring on the chimney. The electrical connections are made with the electrodes, and the power supply switches are turned on.

c. The rheostats are set to deliver 2.5 amp until the solution either foams vigorously or reaches a temperature of $\approx$95°C. When the samples have reached a temperature of $\approx$95°C, steam will be discharged from the hole in the top of the cells, and the action caused by convection current will be vigorous. At this point, about 5 min have elapsed, and the current is reduced to 1.5 amp and held steady until the plating time is completed. The total plating time is 50 min.

d. After electrolysis is completed, the power is turned off and the electrical leads are disconnected from the cell. The solution in the cell is quickly emptied into the sink, and the cell is rinsed thoroughly with distilled water. Subsequently, the cell is disassembled so that the plated surface of the silver disc receives little or no exposure to the atmosphere while in an upright position. The plate is blotted dry with Kleenex.

e. The disc is placed in a properly marked glassine bag that contains the sample number, badge number, and date of sample.

f. A second aliquant from each sample is handled in the same manner except that it is electroplated on a different run. A run is defined as that group of aliquants which are electroplated at the same time. If volume permits, a third aliquant from the sample is spiked with a known number of alpha counts (10) and analyzed on a third or separate run. Counts on the spiked portion minus the counts on the unspiked portion of the sample divided by

Table 1A

Agreement of Counts on the Same Plate

Smaller count	Upper limit for larger count	Smaller count	Upper limit for larger count
1	7	16	30
2	9	17	31
3	11	18	32
4	12	19	34
5	14	20	35
6	15	21	36
7	17	22	38
8	18	23	39
9	20	24	40
10	21	25	41
11	23	26	43
12	24	27	44
13	26	28	45
14	27	29	46
15	28	30	48

If the smaller count exceeds 30, the range between them should be less than

$$2.77\sqrt{\frac{\text{Smaller} + \text{Larger}}{2}}.$$

Table 2A

Agreement of Plates From the Same Sample

Average net count	Upper limit for larger count rate	Average net count	Upper limit for larger count rate
0	5	16	32
1	6	17	34
2	7	18	36
3	9	19	37
4	11	20	39
5	12	21	41
6	14	22	43
7	16	23	44
8	18	24	46
9	19	25	48
10	21	26	50
11	23	27	52
12	25	28	53
13	27	29	55
14	28	30	57
15	30		

If the average of two plates exceeds 30, the range between them should be less than 1.57 times the average.

the theoretical or known amount gives the fraction recovered. The average fraction recovered or the average plating efficiency for the week is used in the final mrem calculation.

g. Three aliquants of synthetic urine (unspiked) are analyzed with each run of samples as spot checks for contamination introduced during the procedure and to provide the data from which the blank correction value is determined.

4. Counting

a. The plated disc is counted for 30 min in each of two counters. The blank value is subtracted from each one-half-hour count and both are recorded on the IBM card.

b. If the two counts on a plate differ by more than the values given in Table 1A, a third count is made. If the net counts on the two unspiked plates of a sample differ by more than the values given in Table 2A, two more plates are prepared and counted if the sample volume permits.

D. CALCULATION

1. Corrected Average Blank

a. The average blank used in the calculations is corrected as follows:

$$\text{Corrected blank} = (TB-CB)\times\frac{40}{90}+CB$$

where TB = average total blank for one week and CB = average counter background for one week.

b. The synthetic urine solution used for blanks has a uranium recovery factor of 90%, while the urine has an average recovery of 40%. The factor TB – CB should be equivalent to the amount of contamination present in the synthetic urine. By multiplying this factor by 40/90, the contamination in the synthetic urine is proportioned to the raw urine basis. Then by simply adding the counter background, the corrected blank is obtained. The average net count is calculated by subtracting the corrected blank from the total count of the unknown sample.

c. If the average net count for any sample exceeds 10 counts per half hour, the Health Physics Department is notified immediately.

2. Milliroentgen Equivalent Man/Day

a. Equation for mrem/day:

$$\text{mrem/day} = \frac{\text{av net count}}{30\text{ min}}\times\frac{2\text{ dis}}{\text{count}}\times\frac{\text{volume}}{\text{ml}}$$

$$\times\frac{24\text{ hr/day}}{\text{Time Interval}}\times\frac{100\%}{\text{P.Eff.}}\times 0.5814.$$

b. All constants in the above equation can be combined to give the following simple form:

$$\text{mrem/day} = \frac{\text{av net count}\times\text{volume}}{\text{P.Eff.}\times\text{Time Interval}}\times 4.6512.$$

A Uranium Refinery and Metal Plant Urine Program and Data

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Abstract

This paper describes the urinary uranium program at the Fernald, Ohio, Feed Materials Production Center and its development over a six-year period from August 1952 to the present. An IBM program on trial since January 1958 has provided information used in the interpretation of urinalysis results. Results, correlated data, and interpretations are given. It is concluded that a good urinalysis program with careful documentation of the data is essential to the evaluation of occupational uranium exposure and its effects.

INTRODUCTION

Our urinalysis program was begun in August 1952. In the last five months of that year we analyzed 245 urine samples. From 1953 through 1957 we collected, analyzed, and recorded results from urine samples in steadily increasing numbers, until by the end of 1957 we had analyzed some 40,000 samples. The 40,000 results have served their usefulness. They were collected for control purposes. We do not believe we can use these data for other than their intended purpose.

In January 1958 we began a program of routine urine sampling of all our employees. At the same time we started using IBM equipment for recording and tabulating the urine sample information and results. We believe that we have since learned a great deal about the interpretation of results, and we expect to continue our routine sampling program.

Although the new system has yielded information which might help us to interpret old results, we are of the opinion that little would be gained by going back to the old data. We have taken sections of old data and attempted to fit them to more recent findings. However, in so doing we are nearly always confronted with a lack of information necessary for a proper interpretation. While summaries of data are given in this paper for samples from 1952 to the present, most of the information pertains to our present system. It is from the 1958 data, $\approx$4000 results, that most of the conclusions are drawn.

METHODS

General

At present there are $\approx$ 2400 employees, of whom about 1600 work in process areas. From this group we expect to obtain routine urine samples at $\approx$ 3-month intervals. The remaining 800 employees work in nonprocess areas. This group will be sampled semiannually on a routine basis. Many will be required to give additional samples for various reasons, bringing the total number to 12,000 to 15,000 samples a year.

Types of Samples

Types of urine samples collected are designated, by reason for sampling, as follows:

1. Pre-employment
2. Annual
3. Routine
4. Incident
5. Special
6. Termination
7. Re-employment

Place and Time of Collection

All samples are single voidings collected in the Medical Department in the Health and Safety Building on the plant site. With the exception of pre-employs, re-employs, the first of an incident series, and some special samples, all are collected at the start of the shift. For obvious reasons, however, we cannot confine most of our sampling to the start of the work week.

Routine Samples

Routine samples are collected by prearranged schedule at 3-month intervals for all process area workers and at 6-month intervals for nonprocess area workers. With more information as a guide it is expected that we will be able to cut back on the number of samples, particularly those from nonprocess area employees.

Incident Samples

Incident samples are collected following an accidental exposure to a suspected high concentration of air-borne uranium such as might occur with a uranium fire or accidental release of material to the air. The procedure for submission of incident urine samples is as follows: When the incident is over each involved individual is to void his urine. This first voiding is not collected for analysis. After a suitable time interval, perhaps 2 to 3 hr, the individual is requested to report to the Medical Department to leave a sample. Four additional samples are collected, one each at the start of the next four working days.

Special Samples

Special samples may be collected for any one of several reasons. Two examples are a special study of a particular job or a recheck of individuals showing high results.

Recheck of High Results

All persons submitting annual or routine samples with a uranium concentration >0.025 mg/l are rechecked until the concentration falls below this level or until it is evident that they are excreting above this level. This can usually be determined after one or two recheck samples, from a knowledge of the person's work and the expected excretion level.

Forms

Paperwork is kept to a minimum by using a single form made out in duplicate throughout the various steps of the procedure. This form serves as a notice to the individual to report for sampling, as a laboratory data form, and as the means for transferring information necessary for IBM recording and tabulating.

For most samples such as annuals, routines, incident samples (other than the first), and special samples (other than rechecks) the form is prepared in advance by the department in which the person is employed. Forms for pre-employees, employees first reporting on an incident, and those for recheck samples are prepared by the Medical Department. This department receives the duplicate copy of forms initiated by other departments. These are used to complete the recheck forms.

Notice to Report

Notices are distributed by the initiating department to its salaried employees. Hourly employees' notices are placed with the time cards at the main entrance to the plant on the shift prior to the designated time for reporting to the Medical Department.

Submission of Samples

The Medical Department detains the person reporting to submit a sample only for the time required to stamp his notification form with a sample number corresponding to the number on the bottle he receives and to obtain his sample. Each person, as he leaves, deposits the bottle containing his sample and his notification slip in a receptacle provided for this purpose.

Treatment and Analysis of Samples

Both the bottles and the slips are then sent to the laboratory, where the samples are analyzed and the results entered on the forms. Since delays do occur in the laboratory, all samples are first acidified with a few drops of HCl to keep the uranium in solution. Uranium determination is by the standard photofluorimetric technique.

IBM Tabulation and Reports

Completed forms are sent from the laboratory to the Medical Department, where they are checked for high results and then forwarded to the IBM Department for recording and tabulating. IBM reports of urine results are made only to the Health and Safety Division. In addition to a regular weekly report, in which results for the particular week are grouped by plant and by job number within the plant, a quarterly report is furnished. This report groups all sample results for the quarter by plant, job number within the plant, individual within the job classification, and date of sample if the individual has submitted more than one sample during the 3-month period. This type of report is a valuable aid to the evaluation

Table 1

Numbers of Urine Sample Results in Different Concentration Ranges

Year	Total No. of samples collected	Concentration range: 0 to 0.050 mg/l No.	%	0.051 to 0.100 mg/l No.	%	>0.1 mg/l No.	%
1952	245	210	85.7	15	6.1	20	8.3
1953	2528	2163	85.5	267	10.6	98	3.9
1954	8375	6934	82.8	1060	12.7	381	4.5
1955	9773	7333	75.0	1521	15.6	919	9.4
1956	9543	7546	79.1	1652	17.3	345	3.6
1957*	2169	1966	90.7	148	6.8	55	2.5
1958**	4114	3887	94.5	193	4.7	34	0.8

*October through December only.
**January through August only.

of exposures for various jobs and individuals within a job.

Investigation of High Results

We have always used urine sample results as an indication of the air contamination problem within the plant or on a job. A high frequency of urine sample results above 0.050 mg/l within a particular job or plant has always required industrial hygiene investigation of the work to determine the cause. Field investigations have often led us to a source of exposure of which we were not aware at the time. However, the investigation did not always enable us to determine the cause of exposure. In such cases one could always blame the high result on the laboratory or on sample contamination. We realize that laboratory errors and sample contamination do occur, and we continually guard against them. Nevertheless our attitude is one of high confidence in our laboratory results, and we assume that the reason for high results can be found in the man or in his work if a diligent search is made. Improvements made in our urinalysis program, particularly the use of IBM correlated data to aid interpretation, have virtually eliminated the need for extensive field investigation of urine results.

FINDINGS

Considerable variation is found in urine sample results for an individual from one voiding to another and for a job from one individual to another. Without specific knowledge about the person, his habits, and his particular exposures, any interpretation of a result of a single sample may be unreliable. Yet we find that when a sufficient number of samples are analyzed, good correlation exists between the urinary uranium concentrations and air dust exposures. We find that an average uranium concentration in urine of 0.030 mg/l, in a preshift sample, corresponds to an air exposure of 70 d/m/m^3 or about 50 μg/m^3 of normal uranium.

Samples collected during and immediately after the hours of exposure show somewhat higher concentrations than preshift samples. Individual postshift results may range from a decrease to a 200% increase compared to the preshift results, but in general will average about 50% higher than preshift concentrations.

In evaluating our incident urine sample results it was found that the time of highest excretion following the incident exposure varied in individual cases from a few hours to several days after the exposure with more high results occurring on the second or third day after exposure than at other times. This was found with exposure to both a soluble and a *so-called* insoluble uranium compound.

DATA AND DISCUSSION

Summary of All Data

For comparison, data for the years that the urinalysis program has been in effect are given in

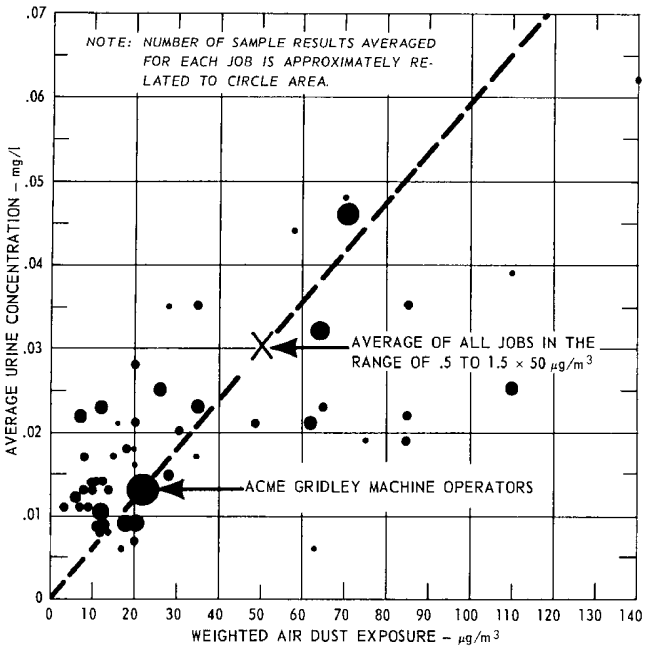

Figure 1. Urine concentrations versus air dust exposures.

Table 2

Summary of Urinary Uranium Results From Job 148
All samples are of preshift, non-incident type.
Results from individuals submitting four or more samples are included.

Person	No. of samples	Urine results, mg/l		
		Low	High	Average
A	7	0.000	0.044	0.019
B	8	.005	.042	.020
C	6	.000	.018	.008
D	8	.001	.028	.012
E	6	.000	.012	.007
F	7	.003	.022	.009
G	6	.006	.022	.010
H	8	.004	.047	.013
I	4	.007	.024	.015
J	6	.002	.019	.010
K	8	.003	.035	.017
L	4	.008	.017	.013
M	8	.000	.022	.013
			Average of group	0.013

Air exposure for group (weighted)=22 $\mu g/m^3$.

$$\frac{22}{0.013} = \frac{50}{0.0295}$$

$\therefore$ 50 $\mu g/m^3$ in air is equivalent to 0.030 mg/l in urine.

Table 1. Because of the significance attached to concentrations of 0.050 mg/l, the former practice was to group data in the concentration ranges given in the table. This practice was discontinued beginning in 1957. However, some of the results for that year have been summarized in this manner and are included in the table.

There is little benefit in further breakdown of the concentration ranges in Table 1. Many of the results >0.1 mg/l were in the 0.2 to 0.5-mg/l range. Most of these higher results occurred with exposure to soluble uranium compounds. In 1956, the first full year of operation of the UF_6 Reduction Plant, there were ≈100 sample results above 0.5 mg/l, of which about 12 were > 1.0 mg/l. Although these results were considerably above a desirable maximum level, there was neither sustained urinary uranium concentration > 0.50 mg/l nor evidence of any resulting personal injury.

Correlation of Air Dust Exposure and Urinary Uranium

IBM tabulated results were used for an exposure-excretion correlation study. Only preshift, annual, routine, and special urine results were used. The average urine result was calculated for each job. These averages were then plotted against weighted air-borne material exposure for the various jobs as shown in Figure 1.

Although the individual points – each representing the average of four or more samples – are widely scattered, a definite trend is seen. The line representing the average has not been statistically located. In fixing this line, more weight was given to those points for which there were larger numbers of samples.

We place the greatest reliance in the point for Acme Gridley operators. There were 120 urine sample results averaged for this job. From this it may be concluded that, if the urinary excretion is a linear function of the exposure, the urinary excretion is ≈0.030 mg/l when the weighted air exposure for 40 hr a week is 50 $\mu g/m^3$.

The 0.030-mg/l concentration for 50 $\mu g/m^3$ is further substantiated by taking the average of the urine concentrations for all jobs in the range from 0.5 to 1.5 times the 50-$\mu g/m^3$ exposure level and relating this average to our "MAC" exposure. In this range we have more confidence in our air exposure evaluations than we have in either lower or higher ranges. Several reasons can be given for this statement, two of which are: fewer air

Table 3

Jobs in 25 to 75-$\mu g/m^3$ Exposure Range

No. of jobs	Weighted air dust exposure, $\mu g/m^3$		No. of urine samples	Average urine concentration, mg/l	
	Range	Average		Found	Expected*
4	26 – 31	28	42	0.023	0.017
4	35 – 49	38	47	.026	.023
4	58 – 64	63	52	.026	.038
5	65 – 75	70	63	.042	.042

*Calculated values assuming 50 $\mu g/m^3$ air dust exposure equivalent to 0.030 mg/l in urine.

Table 4

Numbers of Urine Sample Results in Different Concentration Ranges for Production Jobs With Air Exposures Estimated To Be Above and Below MAC and for Administrative Jobs

Weighted air dust exposure	No. of samples	Concentration range					
		0.026 to 0.050 mg/l		0.051 to 0.100 mg/l		>0.100 mg/l	
		No.	%	No.	%	No.	%
>70 d/m/m^3	318	161	51	50	16	8	2.5
<70 d/m/m^3	762	109	13	12	1.6	0	0
Admin. area	267	13	4.9	2	0.7	0	0

samples are collected for jobs and locations for which the air concentrations are low; whereas higher exposure estimates are usually biased by the use of respirators, which are required when the air concentration has been found high.

The data pertaining to the Acme Gridley operators and to jobs in the range from 25 to 75 $\mu g/m^3$ are given in Tables 2 and 3. The variance between individuals within the job, Table 2, is typical of our urine results. Variance of the same order is seen in comparison of one job with another. In Table 3 the jobs in the 25 to 75-$\mu g/m^3$ range were grouped in smaller concentration ranges.

Interpretation of Frequency of High Results

On our IBM reports each result above 0.025 mg/l is flagged for attention by an asterisk. Since the IBM reports list the results by job number, it is easy to find the jobs corresponding to these higher urine results. Without knowledge of the air dust exposure one can usually predict higher exposure jobs by the number of urine results above 0.025 mg/l. A review of the tabulations indicated that a urine result above 0.050 mg/l (preshift, non-incident samples) was seldom found unless the weighted exposure was estimated to be more than our MAC of 70 d/m/m^3.

Table 4 is a summary of data for 52 production job classifications, of which 18 were jobs with air exposures estimated to be in excess of our MAC. Only those jobs for which there were 9 or more urine samples are included in this comparison study. Also included, however, are results from employees in our administration area, whose exposures are assumed to be much below MAC.

Ten of the 12 samples > 0.050 mg/l in the middle group were submitted by persons in four job classifications for which an exposure in excess of the calculated value is suspected. Of a total of 77 samples from these four jobs, 33 gave results (43% of the total) >0.025 mg/l. This leads us to suspect that their exposures are at least bordering on our air MAC.

One of the two results >0.050 mg/l in the administration area group is for a Production Records man. These people spend considerable

Table 5

Average Urine Concentrations During and After Desludging Operations on Salt Bath Furnace

	Day of sample						
	1	2	3	4	5	6	7
GROUP A: 50 MEN EQUIPPED WITH AIR-FED HOODS							
Air dust, ×MAC	235.0	5.0	2.0	–	–	<1.0	<1.0
Beginning-of-shift sample, mg/l	0.018	0.029	0.022	–	–	0.018	0.023
End-of-shift sample, mg/l	0.119	0.047	0.038	–	–	0.030	0.023
GROUP B: 14 MEN EQUIPPED WITH DUST-TYPE RESPIRATORS							
Air dust, ×MAC	28.0	5.0	2.0	–	–	<1.0	<1.0
Beginning-of-shift sample, mg/l	0.018	0.034	0.016	–	–	0.014	0.018
End-of-shift sample, mg/l	0.038	0.037	0.035	–	–	0.027	0.024

Work began on Wednesday, day 1, and was completed on Friday. Saturday and Sunday were days off. Normal operations were resumed on Monday.

time in the production area. Urine results for the job indicate that some of their group are being exposed to uranium. The other higher result was from a sample submitted by a porter who had not been assigned to the production area.

Acute Exposures

Most of the preceding information pertains to chronic exposure. Some of our data pertaining to acute exposure have been reported.[1,2] We have not added appreciably to our information since these reports were made. In our recent incidents, air dust and urine samples have not indicated that the unusual occurrence was a source of high exposure. It is possible that the exposure level during area cleanup operations was higher than it had been at the time of the incident.

There was one planned high exposure job for which workers were required to wear respiratory protection. A summary of information pertaining to this job is given in Table 5. Tables 6 and 7 list data pertinent to two incidents. The first was a release of 4½ lb UF_6. Urine results for 12 men suspected of receiving exposure in the accident are given. The second incident was a fire in the duct work of the ventilation system for uranium machining operations. After this accident 116 men submitted samples according to our incident urine procedure. Most of these men showed no significant increase in their urinary uranium level. However, 16 men had at least one result > 0.050 mg/l. All the results for these 16 men are given in Table 7.

Table 6

U Concentrations in mg/l in Urine of Individuals After Pilot Plant Hex Release

Person	Time interval after incident				
	2 hr	1 day	2 days	3 days	4 days
A	0.049	0.033	0.025	0.013	0.074*
B	.128**	.058*	.037	.025	.000
C	.021	.016	.066*	.237**	.031
D	.034	.082*	—	.021	.021
E	.003	.025	.074*	.019	.011
F	.074*	.033	.119**	.032	.007
G	.018	.049	.026	.029	.029
H	.022	.032	.037	.033	.011
I	.022	.015	.037	.025	—
J	.023	.008	.237**	.074*	.006
K	.018	.029	.033	—	.006
L	.029	.033	.066*	.012	.028

*Results in 0.050 to 0.10-mg/l range.
**Results >0.1 mg/l.
Persons A and B were thought to have received the greatest exposure.

In the hex release, exposure was definitely to soluble uranium. The exposures in the furnace

Table 7

U Concentrations in mg/l in Urine of Individuals After Plant 6 Machining Area Fire

Person	Time interval after incident						
	2 to 4 hr	1 day	2 days	3 days	4 days	5 days	6 days
A	0.047	0.056*	0.063*	0.061*	0.051*	–	–
B	.005	.027	–	.185**	.024	0.036	0.016
C	.014	.025	.077*	.076*	.022	–	–
D	.028	.036	.040	.039	.049	.052*	–
E	.012	.038	–	.086*	.020	.012	.026
F	.014	.037	.057*	–	.028	–	–
G	.022	.032	.026	.071*	.022	–	.017
H	.044	–	–	–	.042	–	.134**
I	.017	.027	–	.078*	.024	.055*	–
J	.010	–	–	.010	.010	.018	.165**
K	.007	.004	–	–	.007	.009	.093*
L	–	–	.013	.029	.017	.082*	.016
M	.041	–	–	.071*	–	.021	.007
N	.042	.010	–	.076*	.017	.016	.011
O	.013	062*	–	–	.034	.021	.030
P	.006	.052*	–	–	–	–	–

*Results in 0.050 to 0.10-mg/l range.
**Results >0.1 mg/l.

desludging work and in the uranium fire might be regarded as "insoluble" uranium exposure. One would certainly suspect that the air-borne particles were mostly uranium oxides, either UO_2 or U_3O_8. However, the furnace sludge consisting of lithium and potassium carbonates and containing about 30% uranium by weight is streaked with a yellow salt, possibly uranium trioxide or uranium carbonate.

Except for higher uranium concentrations in individual cases, the excretion patterns in the three studies are like those encountered with chronic exposure. The high concentration for group A, Table 5, at the end of the first working day is believed to be more typical of soluble than of insoluble uranium exposure. Yet by comparison in the UF_6 release only one person was at his highest level soon after exposure. We have no explanation for the fact that the groups as a whole (Tables 6 and 7) showed higher uranium concentrations in their urine on the second and third days after exposure, unless there was additional higher-than-expected exposure on the first or second day following the incident. The fact that there were similar occurrences in urine results of UF_6 Reduction Plant operators leads us to search for other explanations.

SUMMARY AND CONCLUSIONS

In spite of individual differences which make interpretation of urine results more complex, good correlation exists between the average uranium concentration in urine and the air dust exposure. An average urinary uranium concentration of 0.030 mg/l in preshift urine voidings corresponds to an air dust exposure averaging 50 $\mu g/m^3$ of normal uranium for 40 hr a week. The relationship of other urine to air dust exposure concentrations is assumed to be linear. However, a strict interpretation based on this finding should not be placed on a single urine result. It is concluded that job and individual exposures can be approximately determined by urinalysis allowing for individual differences. A standard deviation from the norm has not been determined.

If only a few urine results are available for an individual or a job, a high degree of confidence can be placed in this formula: If the uranium concentration in urine is usually <0.025 mg/l and never >0.050 mg/l ,the air dust exposure is not >50 $\mu g/m^3$.

Urinalysis is recommended, not as a substitute for, but as an aid to the industrial hygiene survey program. Much more must be learned about both

before one can be substituted for the other. Additional information is needed about the particle size of the exposure dusts and the relationship of particle size to deposition and retention of uranium in humans. We need to know more about the solubility of various uranium salts in body fluids.

Use of the term "urine MAC" has been purposely avoided in this paper. Formerly our organization referred to a uranium concentration in urine of 0.050 mg/l as the maximum allowable concentration. In so doing, we were arbitrarily establishing a relationship whereby one MAC in air equals one MAC in urine equals one MAC in the body. From our tissue and urine analysis findings it appears that the air, urine, and body concentrations are related, but our present knowledge is not sufficient to estimate a body concentration from either air or urine results. Evidence points to a probable lower body concentration from exposure to 50 or 75 μg U/m^3 air than has been suspected.

We no longer use the term "urine MAC" but refer to a concentration of uranium in urine $>$0.025 mg/l as a recheck concentration. Our experience is that the implication often derived from "MAC" is that any concentration higher than MAC constitutes injury. While we do not suggest the adoption of our practice, we do caution against indiscriminate use of terms which should be carefully defined.

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The Hanford Uranium Bio-assay Program

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INTRODUCTION

The Hanford bio-assay group was founded early in 1946 for detection and measurement of radioisotopes deposited in project workers. Plutonium was of primary interest, but concurrently development work was also initiated on procedures for detection and measurement of uranium and fission products.

Early attempts were made to concentrate uranium in urine samples by ether extraction and by electroplating; however, the results were erratic and not at all successful. Early in 1948 the fluorometeric method, fusing of uranium with sodium fluoride and measuring the fluorescence when exposed to ultraviolet light, was adopted and developed to a measurement sensitivity of 5 to 10 micrograms of uranium per liter of urine. The fluorometric method has since been used continuously, with some refinements of sensitivity to 3.8 μg/l.

The basic philosophy at Hanford regarding uranium contamination control is that deposition of uranium in the human body must be kept at a minimum. Since biological data for humans are scanty, and actual long-term effects of uranium deposition are not well known, all reasonable precautions should be taken to prevent or minimize exposure of personnel. The bio-assay program is the prime measure of the over-all effectiveness of efforts to safeguard our employees.

HANFORD URANIUM PROCESSING AREAS

At present there are two uranium processing facilities at Hanford, one for purification of irradiated uranium and another for preparation of reactor fuel elements.

The only significant contamination and exposure problem from uranium exists in the uranium purification building, where the final step in the separation process is calcination of uranyl nitrate hexahydrate solution to uranium trioxide (UO_3) and packaging for shipment off-site. Equipment and process flow refinements have been made continuously since start-up of the plant in early 1952. An essentially enclosed continuous-flow process now in use has eliminated most of the high air contamination problems in routine processing. However, air contamination normally occurs during maintenance work or occasional equipment malfunction.

Storage and concentration of uranyl nitrate hexahydrate prior to calcination has not presented any radiological hazard of concern. Air contamination levels occasionally range as high as 1×10^{-11} μC U/cc, but normally they are 1 to 3×10^{-12} μC U/cc, which is the same level as that found in the operating areas. In the cells where the continuous calciners are located, air concentrations range from 5×10^{-12} to 1×10^{-11} μC U/cc. Maintenance shutdowns for equipment repair may cause air concentrations of 1×10^{-9} μC U/cc or greater. Usually at these levels a yellow dust can be seen in the atmosphere. Protective clothing including respiratory protection is required to protect personnel adequately under these conditions. The outer clothing becomes contaminated with UO_3 dust after 10 or 15 min in these air concentrations, but, if the protective equipment is properly worn and the openings taped, skin contamination is kept at a minimum. During the summer weather when temperatures are 90° to 100°, perspiration soaking through the layers of clothing occasionally allows contamination to be carried back to the skin; however, no influence on internal deposition has been detected. These same contamination problems exist to a greater degree with the process vacuum system used to transport UO_3 powder, after leaving the calciners, to the hammer mill and final load-out station. During routine maintenance necessary to clean or change filters, the protective clothing of personnel may become covered with a film of UO_3 powder. The use of fresh air or chemox type respiratory protection in place of filtration type masks for maintenance work has

Table 1

Urinary Excretion of Uranium, Case 1

Date	Time	Time after exposure, hr	Sample volume, cc	U excreted, μg	Concentration, μg/l
12-17	0150	0			
	0455	3	400	335	838
	0625	4.5	200	170	850
	0800	6	600	402	670
	1300	11	530	166	313
	2105	19	510	132	259
12-18	0930	31.5	460	437	950
	1400 to				
12-19	0600	43	1600	12.5	8
	1335	58.5	100	1.6	16
	1830 to				
12-20	0530	70	1260	14.9	11.8
	0800	78	110	0.7	6.5
	1015	80	115	0.5	4.4
	1445	84.5	120	1.2	10
	1800 to				
12-21	0700	94	1165	9.9	8.5
	0900	103	—	1.0	—
	1145	106	—	1.0	—
	1445	109	—	1.0	—
	even. to				
12-22	morn.	—	—	1.0	—
	even. to				
12-23	morn.	—	700	1.7	2.4
	0920	151	200	0.6	3.0
	1310	155	190	0.8	4.2
	1445	157	210	0.5	2.4
12-25	1845	209	210	2.0	9.5
	2045	211	220	0.9	4.1
12-27	1050	249	220	0.7	3.2

noticeably reduced the level and number of high bio-assay samples.

Hanford reactor fuels preparation is a highly specialized operation of canning fuel elements for use in the reactors. Tasks, such as rod extrusion and straightening, machining, and operation of a melt plant for metal scrap recovery, that created a significant contamination problem were discontinued in early 1953. As these operations were curtailed and major improvements were made in the ventilating systems, the percent of the total number of urine samples containing >15 μg/l was reduced almost by a factor of three. Air concentrations very rarely reach 3×10^{-11} μC U/cc, the level at which respiratory protection is used, during the operation of the present fuel preparation facilities. Generally it is less than the detectable limit of 1×10^{-12} μC U/cc, and ranges to 2×10^{-12} μC U/cc. Slightly higher air concentrations, ranging to 4×10^{-12} μC U/cc, are encountered in the recovery operation of rejected fuel elements.

SAMPLING METHODS

Routine bio-assay sampling of personnel working in uranium processing facilities is accomplished by collection of "before" exposure and "after" exposure samples. "Before" exposure sampling is done at the beginning of an employee's work week, and "after" exposure sampling at the end of the work week. Adoption of this type of sampling was based on University of Rochester[1] suggestions for distinguishing between skeletal accumulation and recent intakes of readily soluble material. Their studies with animals and limited human data indicate that $\approx$ 75% of the

initial intake of soluble material such as uranyl nitrate hexahydrate is excreted in the urine during the first 24 hr.

Frequency for routine "before" and "after" exposure sampling at Hanford is on a weekly or monthly basis. Supplemental sampling of the "before" and "after" type is scheduled whenever particularly high level contamination work is anticipated to assist in pinpointing the time of possible exposure. Sample volumes obtained from the routine bio-assay program usually range from 100 to 250 ml, and the results are expressed as urinary concentration.

ACUTE EXPOSURE CASE STUDIES

The first case involved an employee who was completely immersed in a uranyl nitrate hexahydrate solution by falling through a large flange opening of a tank on which the temporary cover was not securely fastened. The tank contained 56% uranyl nitrate hexahydrate solution (0.23 *M* free nitric acid) to a depth of 10 ft. The men working nearby were able to pull him out immediately, so that the immersion time was probably less than one minute. His clothing was removed immediately, and he then rushed to a safety shower $\approx$200 yards away. After the shower $\approx$500 counts/min as indicated by portable GM meters remained on the skin areas. A general contamination level of $\approx$4000 counts/min was found on his wet clothing. Further decontamination at the area first aid station reduced the skin contamination level to 100 counts/min or less. Within 2 hr the man was taken to the hospital for observation by company physicians, and their findings indicated very minor chemical conjunctivitis and minor total-body skin irritation. The external radiation dose for the brief immersion time was considered to be one mrad or less. It was estimated that less than 7 min elaspsed between the time the man fell into the tank and completion of his shower, when most of the uranium solution was removed from the skin surface. Three to 4 hr later all traces of uranium that were detectable with portable survey meters had been removed by scrubbing.

Bio-assay sampling was initiated immediately following the exposure, and complete collections were made for several days. Table 1 is a résumé of bio-assay sampling results, and Figure 1 is a plot of uranium excretion versus time. In evaluating this case it was assumed that the urinary excretion of uranium should be very similar to that following intravenous injection of uranyl nitrate hexahydrate.[1,3] The excretion pattern was followed closely during the first 31 hr following exposure, and the cumulative excreted dose was very similar to that expected for uranyl nitrate hexahydrate. The excretion rate continued to decrease rapidly until our detection limit was reached within a few days following exposure. Based on these observations, the estimate of initial body deposition was $\approx$2000 μg with about 120 μg deposited in the bone.

The second case involved an acute intake of UO_3 primarily by inhalation and possibly also by absorption and ingestion. This employee wore a defective assault mask while changing a filter bag connected with the UO_3 powder unloading system. When the mask was removed upon completion of the work, UO_3 powder was visible around his nose, mouth, and chin. Portable alpha survey meters indicated levels to 10,000 dis/min per probe area of $\approx$7 in^2. A shower removed the detectable surface contamination, and oral and nasal irrigations were performed at the area first aid station. Visible traces of yellow UO_3 powder were evident in the initial irrigation water. The first irrigation water removed $\approx$2200 μg uranium and the second $\approx$230 μg.

Extensive bio-assay sampling was done in this case, as shown in Table 2. The excretion rate for inhaled UO_3 powder appears to be similar to that expected for inhaled uranyl nitrate hexahydrate.[13] The mode of intake in this case was as-

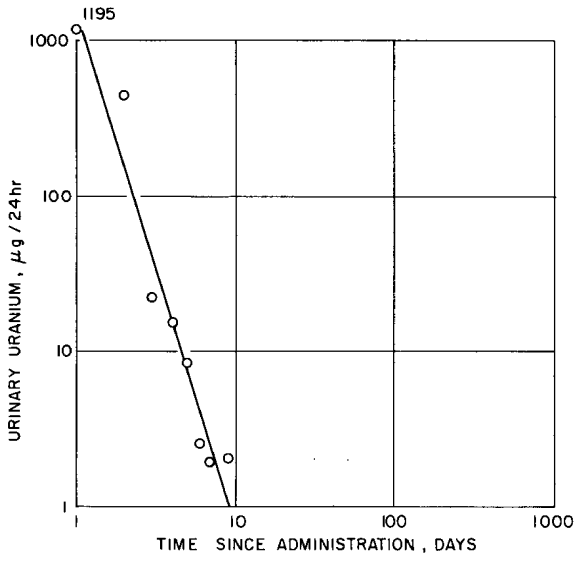

Figure 1. Urinary excretion of uranium, Case 1.

Table 2

Urinary Excretion of Uranium, Case 2

Date	Time	Time after exposure, hr	Sample volume, cc	U excreted, μg	Concentration, μg/l
5-26	2330	0			
5-27	0230	3.0	160	155	956
	1000	10.5	150	152	1015
	1330	14	154	95	631
	1730	18	150	56	373
	2015	20.7	225	77	344
	2200	22.5	168	176	1046
	2355	24.5	170	57	335
5-28	0900	33.5	370	68	183
	1400	38.5	270	320	1185
	1620	41	125	138	1100
	2000	44.5	130	48	381
	2230	47	128	128	1008
	2400	48.5	125	13.5	99
5-29	0730	56.5	390	33	84
	1200	—	220	18	83
	1420	—	150	11	71
	1800	—	125	10.5	85
	2000	—	120	8.5	70
	2200	—	123	9	72
	2400	—	125	8.2	67
5-30	0800	80.5	250	14	55
	1130	—	150	13	86
	1500	—	130	8.6	66
	1800	—	135	11.2	87
	2100	—	160	6.6	41
	2330	—	150	5.5	36
5-31	0800	104.5	340	16	48
	1000	—	120	7.6	63
	1330	—	320	19.5	61
	1730	—	310	19.5	63
	2030	—	330	22	66
6-1	0330	124	116	5.5	48
	0600	—	355	21	60
	1000	—	120	6.5	55
	1300	—	108	4.5	43
	1900	—	260	12	45
	2130	—	200	7	36
6-2	0600	150.5	300	18	60
	0810	—	220	9.5	43
	1015	—	216	11	50
	1330	—	225	9	40
	1930	—	225	9	40
	2230	—	300	14.5	48
6-3	1000	178.5	230	11	47
	1215	—	228	10	45
	1530	—	220	11	49
	2030	—	360	13	35
6-4	0730	200	460	18	38
	0930	—	275	9.5	34
	1500	—	—	4	16
6-6			1400	57	40

Table 2

Urinary Excretion of Uranium, Case 2

Date	Time	Time after exposure, hr	Sample volume, cc	U excreted, μg	Concentration, μg/l
6-12			1300	16.7	12.8
6-13			900	12	13.4
6-14			900	11	12.1
6-15			1300	12.9	9.9
6-16			1000	12.5	12.5
6-17			900	9.5	10.4
6-18			1600	13.7	8.6
6-22			1600	25.5	16
7-23			1100	7.7	7
7-27			1500	7.6	5.1
8-5			1500	4.3	2.9
8-12			1400	9.9	7.1
8-20			600	14.7	24.6
9-17			1200	2.6	2.2
9-24			1600	3.7	2.3
10-1			1600	1.9	1.2
10-8			800	2.2	2.8
10-15			1600	2.2	1.4
10-22			1100	1.9	1.7

sumed to be primarily by inhalation, and the initial lung burden was estimated on the basis of uranyl nitrate hexahydrate inhalation. From the data collected during the 10 days following exposure, the initial lung burden was estimated at 5000 μg. It is interesting to note that, for the entire period of sampling, the excretion rate appears to follow a power function. (See Figure 2.) Data on urinary excretion of uranium by humans covering this length of time are very limited, thus the validity of the relationship is in doubt.

In the third case of uranium exposure, intake was by absorption and inhalation resulting from burning uranium chips. Experiments were being performed to determine the feasibility of drilling uranium metal at elevated temperatures. The turnings were caught in a metal basket and quenched with a melting salt, then dumped into a bucket. Some of the chips were seen to be glowing, and a cover was placed on the bucket. A reaction occurred, showering the surrounding area with several pounds of burning uranium chips. The individual involved received first and second degree burns on the legs and ankles. His exposure to the contaminated atmosphere immediately following the explosion was less than 4 min without respiratory protection. The air concentration during this period was unknown. Low level uranium contamination was detected with portable GM survey instruments in the burned areas of the legs and ankles. On the following day, when the bandages were changed, no contamination could be detected.

Bio-assay urine sampling following this exposure is summarized in Table 3 and plotted in Fig-

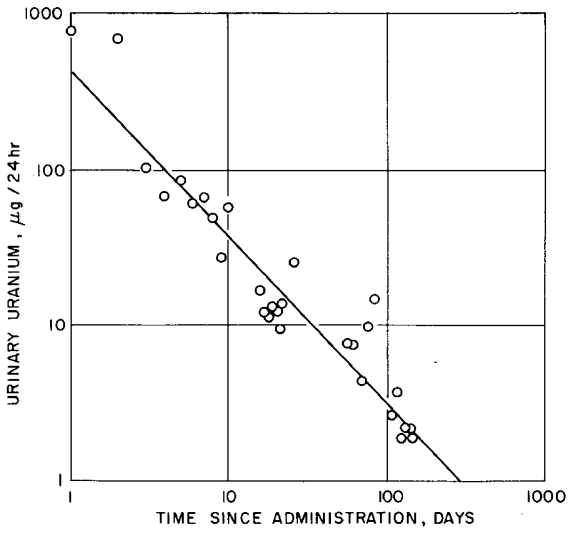

Figure 2. Urinary excretion of uranium, Case 2.

Table 3

Urinary Excretion of Uranium, Case 3

Date	Time	Time after exposure, hr	Sample volume, cc	U excreted, μg	Concentration, μg/l
12-9	1455	0			
	1825	3.5	215	171	791
	2000	5	120	2	17.5
	2200	7	250	40.7	163
12-10	0800	17	110	23.1	210
	1000	19	162	6.8	43
	1340	22.5	110	11.1	101
	1700	26	124	8.8	72.5
	2000	29	152	8.4	55.5
12-11	0800	41	390	12.9	33
12-13	2200	103	—	1.6	15.9
12-14	0915	114.5	—	0.8	8.1
	1305	118	—	1.1	10.7
	1700	122	220	0.5	4.9
	1830	123.5	370	0.2	2.2
	2100	126	110	0.3	3.7
12-15	0700	136	265	0.5	5.3
12-16	1000	163	—	0.2	4.1
	1445	168	—	0.3	5.1
12-28	1330	432	—	—	1.7

ure 3. The mode of intake was assumed to be primarily by absorption through the burned skin areas into the bloodstream and to be analogous to an intake by intravenous injection. During the first 24 hr following exposure an estimated 300 μg uranium was excreted. (An estimate is used because it was not determined whether all voidings were collected.) The excretion rate continued to decrease until it became negligible 10 days later. Of uranium in the bloodstream,[1,3] ≈75% of the injected dose is generally assumed to be excreted during the first 24 hr; therefore, the initial deposition in this case was estimated to be 400 μg. The urinary uranium concentration was plotted in μg/l rather than μg/24 hr because of the uncertainty of total urine collection.

CHRONIC EXPOSURE

The chronic exposure problem has been greatly reduced during the past few years by improvements in air contamination control and process changes. Table 4 gives an example of a year's experience in the UO_3 plant. In almost every case where results were >15 μg/l, it was possible to determine the specific time of exposure, and subsequent samples showed <15 μg/l. A review of sampling results shows the normal level of "before" exposure samples to be in the range of 5 to 10 μg/l for operating personnel and 5 to 15 μg/l for maintenance personnel. "After" exposure samples are generally about 5 μg/l higher.

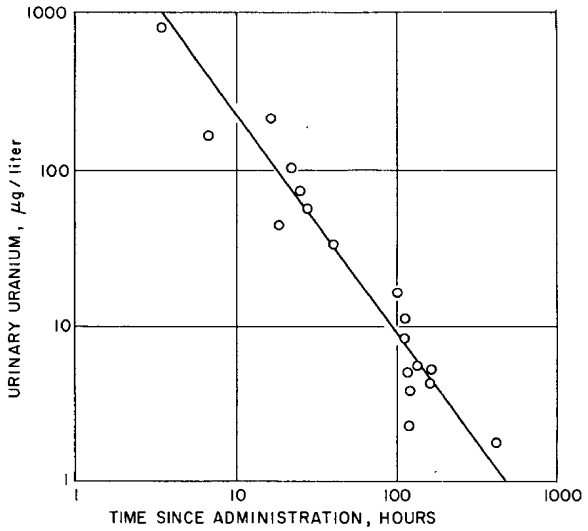

Figure 3. Urinary excretion of uranium, Case 3.

In the fuels preparation facilities, air concentrations are lower than in the UO_3 plant, this being evident in the generally lower excretion rates observed. Fuel element recovery personnel urinary excretions in "after" exposure samples range from less than detection limit to 10 μg/l, and "before" exposure samples from less than detection limit to 5 μg/l. For other operations the "after" exposure samples usually show <10 μg/l, and "before" exposure samples are usually at background level.

To keep a tight control on chronic exposure, Radiation Protection personnel investigate all bio-assay sample results of 15 μg/l or greater. A concentration of 25 μg/l in "before" exposure samples has been established as the warning level, which, if sustained for several weeks, would indicate a significant body burden of uranium. In such a case the individual would be removed from further uranium exposure to prevent additional deposition and to facilitate estimation of the body burden.

Table 4

Uranium Bio-assay Sampling in the UO_3 Plant

		No. in given range		
	Total	15 to 50 μg/l	50 to 100 μg/l	>100 μg/l
No. of employees sampled	176	77	37	20
No. of samples collected	2800	548	78	31*

*Highest result was 2800 μg/l following a known exposure; 5 hr later the excretion rate dropped to 240 μg/l, and 30 hr after exposure to <15 μg/l.

ANALYTICAL METHODS

Determination of uranium concentration in urine samples is by comparison of the fluorescence of Na_2CO_3-NaF flux biscuits containing the urine sample with that of biscuits containing the sample spiked with a known amount of uranium and that of blanks. Thus, each determination requires six biscuits, two of each. Sodium citrate buffer is added to the urine samples to prevent precipitation while they are awaiting processing. The analytical procedure is described in the Appendix.

SUMMARY

Hanford uranium bio-assay sampling methods and evaluation of body burdens are based primarily on studies made at the University of Rochester. "Before" and "after" exposure samples are collected on a weekly or monthly basis according to the potential for exposure. In addition, supplemental "before" and "after" samples are collected whenever the potential for exposure appears significant, in order to assist in establishing the possible time of intake.

Analysis for uranium in urine samples is done by the fluorometric method with a detection limit established at 3.8 μg/l.

Results of the Hanford bio-assay program during the past several years indicate that no significant chronic internal deposition problem exists in the uranium processing facilities.

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APPENDIX
DETERMINATION OF URANIUM IN URINE

The uranium content of a urine sample is determined by comparing the fluorescence of a Na_2CO_3-NaF flux biscuit containing a urine sample aliquot with that of a biscuit containing a sample aliquot spiked with a known amount of uranium and corrrecting for blank values.

1. Prior to making the determination, the platinum dishes should be cleaned and checked for contamination. Dissolve used flux from dishes with 8 *N* nitric acid. Decant and liberally wash dishes with water. Boil dishes for 15 min in 8 *N* nitric acid, decant, wash dishes liberally with water, and dry under heat lamp. If still contaminated, repeat boiling with 8 *N* nitric acid, rinsing with water, and drying.

2. For each determination, take six clean dishes. Add 100-lambda aliquot of urine sample to first four dishes, and add 50 lambda uranium spike solution containing $\approx$10,000 μg U/l to two of these under heat lamp. The other two dishes are blanks.

3. Add 0.6 g NaF-Na_2CO_3 (90-10% by weight) to each of the six dishes. (The dishes are so constructed that 0.6 g forms a biscuit flush with top of dish when fused.)

4. Load the six dishes onto a platinum tray and rapidly load the tray into a furnace at 1000°C. Remove tray from furnace after exactly 3 min.

5. Remove dishes from tray with platinum tongs and allow to cool for 3 min.

6. Load dishes into the fluorometer sample wheel (with tongs), and reset sensitivity of fluorometer.

7. Read the fluorescence of each biscuit and record results.

8. Calculation: Average separately the two blank values, the two sample values, and the two spiked sample values. Then

$$\mu g\ U/l = \frac{\text{av sample reading} - \text{av blank reading}}{\text{av spiked sample reading} - \text{av sample reading}} \times 0.5 \times \text{spike concentration.}$$

The factor 0.5 is included because 50 lambda spike was used per 100 lambda sample. The spike concentration is in units of μg U/l.

Uranium Urinalysis Data at Los Alamos Scientific Laboratory*

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SAMPLING METHODS

The urine sampling program for normal uranium at the Los Alamos Scientific Laboratory was inaugurated in the spring of 1950. In 1952, we began radiometric analyses for uranium on personnel exposed to enriched uranium, and since then we have attempted to determine the optimum collection times. The most extensive work was done on the comparison of Saturday evening versus Monday morning samples collected from normal uranium workers working a 48-hr week. Our data were most inconsistent and gave no trend that enabled us to say that one time gave more information than the other. At one period in the early days of the program, we had machinists working with normal uranium who consistently excreted an average of 50 μg uranium per liter. We took spot samples from these employees on the evening before they left on vacations varying from 7 to 21 days, and again on the morning of their return to work. Of the 16 men studied, 5 showed higher results on return to work, 8 lower, and 3 zero. These last three men had excretion values of 10 μg/l or less prior to vacation. In view of these experiences, we have arbitrarily chosen to collect Friday evening spot samples from our normal uranium workers. A sample taken at the end of the work week should show a detectable concentration if there has been an acute or cumulative exposure.

Daily spot samples were collected from two workers exposed to enriched uranium during a week with a heavy work load. The daily variations of the spot samples are shown in Figure 1. At present a simulated 24-hr sample, which consists of two consecutive morning and two consecutive evening voidings, is collected near the end of the work week for radiometric uranium analyses. The larger volume increases the sensitivity of the analyses and also reflects the exposure for a two-day period.

Our sampling frequency has varied with conditions through this period of eight years. We essentially have two areas of normal uranium exposure: the foundry, fabrication, powder-metallurgy, and research laboratories have consistently had low excretion rates, while our machine shop has shown higher exposure levels. Until 1956, we collected weekly samples from all personnel, and daily specimens during peak work loads from the machinists, for both normal and enriched uranium

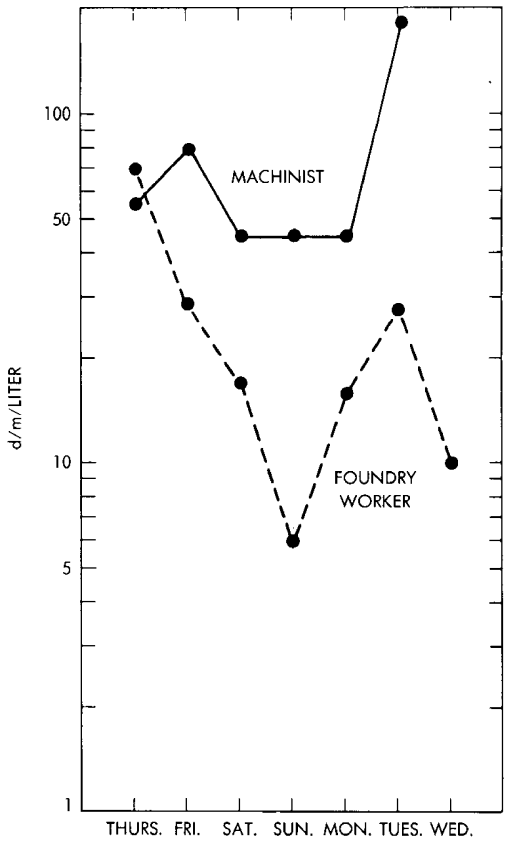

Figure 1. Daily variations in U^{235} excretion in spot samples from two workers.

*This work was done under the auspices of the U.S. Atomic Energy Commission.

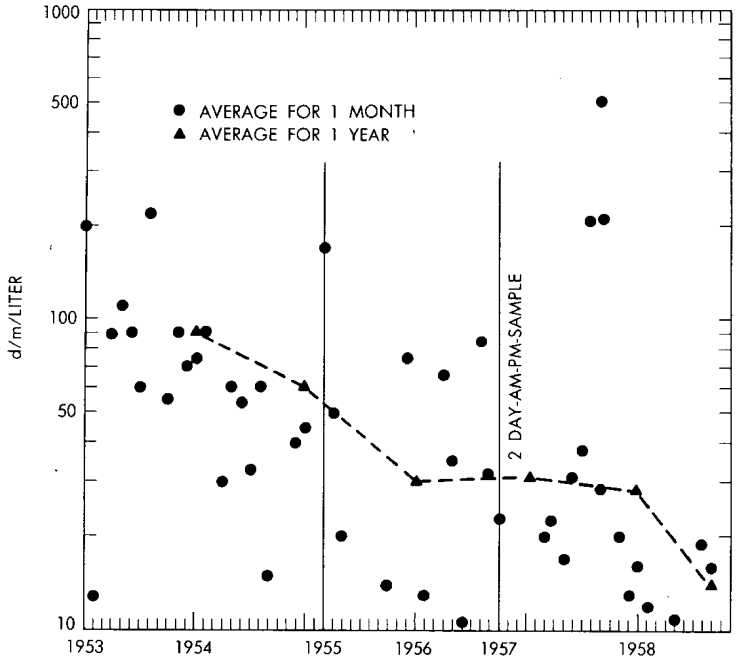

Figure 2. Typical excretion pattern for U^{235}.

Table 1

Normal Uranium Excretion of Workers by Metallurgical Process
(The numbers given are the percents of sample results in the given ranges)

	Range, μg U/l urine			
	<5	5 to 25	25 to 50	>50
Foundry, 1950 to 1958	56	36	8	0.2
Powder metallurgy, 1954 to 1958	77	23	0	0
Fabrication, 1950 to 1954	74	24	2	0.3
Craftsmen, 1951 to 1958	81	19	0	0
Janitor, 1954 to 1958	90	10	0	0
Research, 1956 to 1958	100	0	0	0

exposures. In 1955, local exhaust ventilation was installed in the machine shops, and the uranium excretion dropped markedly. In 1956, we changed to routine monthly samples for all personnel working with uranium. A similar decrease in excretion levels for enriched uranium workers is shown in Figure 2. Since no changes in ventilation have been made in these shops, the decrease is probably due to employee education, the respirator program, better housekeeping, the larger sample volumes used, and improved analytical techniques. In view of these consistently low levels, the enriched uranium workers have been put on the routine monthly sampling program. In case of an accident involving either contaminated wounds or inhalation, we collect a 24-hr sample as soon as practical after the accident.

ANALYTICAL METHODS

Urine samples from personnel exposed to normal and depleted uranium are examined fluorophotometrically by the method of Price, Ferretti, and Schwartz.[1] The method is based on the intense yellow-green fluorescence produced by traces of uranium fused in sodium fluoride and is sensitive

KAPL Experiences in Uranium Health Programs

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Abstract

The facilities for machining uranium at the Knolls Atomic Power Laboratory are discussed. The health physics aspects of the uranium health program are summarized, including air cleaning systems, calculations of maximum permissible urinary excretion levels, bio-assay analyses, and typical bio-assay results.

INTRODUCTION

Since KAPL was awarded its first AEC contract, technical studies have been conducted which have included uranium machining and metallurgical development. KAPL has been concerned in recent years with the development, design, construction, and installation of reactors for underwater and surface craft propulsion. The uranium processing required for this special development work includes standard machine shop operations, i.e., lathe operations, milling, grinding, cutting, and drilling of uranium (normal and enriched) and uranium alloys. In addition to the shop operations there are metallurgical operations such as rolling, swaging, drawing, extruding, melting, plating, and firing of uranium and its alloys and powder metallurgy.

HEALTH PHYSICS ASPECTS OF MACHINING URANIUM

Uranium machine and metallurgy shop areas at KAPL, which present potential health problems, are controlled-contamination zones, and the standard regulations regarding regulated zones are followed. The KAPL health physics procedure has always been to use complete employee protection for each new operation, including constant air sampling (see Figure 1) and respiratory protection, until the operation is classified as radiologically safe. After a procedure has been designated as radiologically safe, intermittent general atmosphere and routine exhaust sampling is begun. The standard machine shop operations are carried on with exhaust control and coolant (Figure 2), and grinding and cutting operations are done in hoods (Figure 3). Uranium is machined wet, even in the form of uranium alloys; if dry machining is necessary, it is done in an inert gaseous atmosphere. Powder metal requires special handling in either dry boxes (Figure 4) or "Hanford-type" hoods (Figure 5).

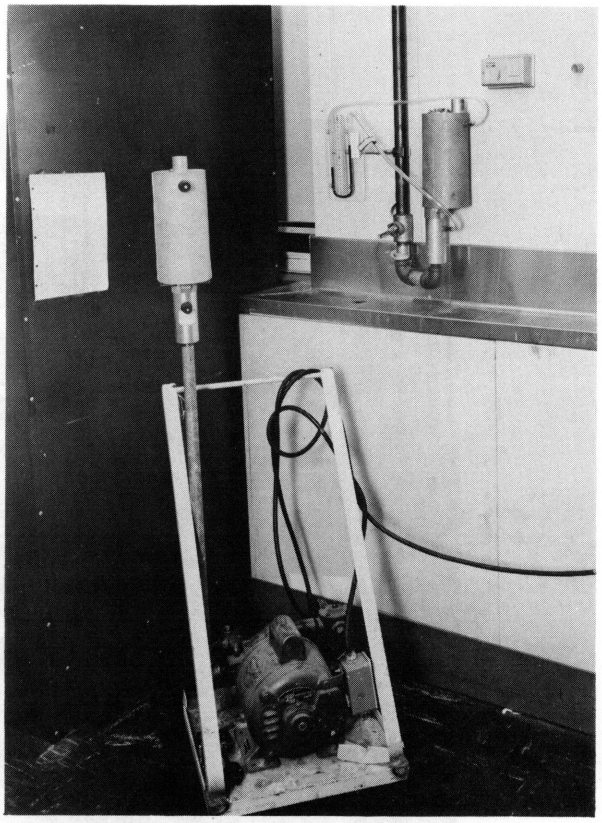

Figure 1. Air samplers.

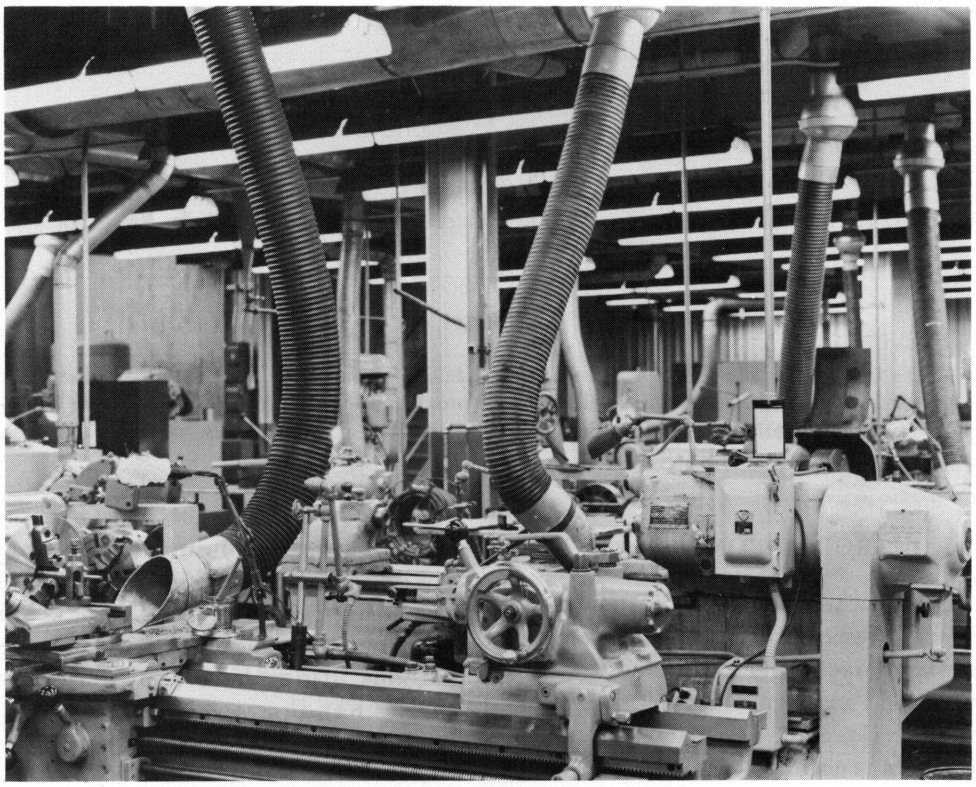

Figure 2. Lathe operation with exhaust control and coolant.

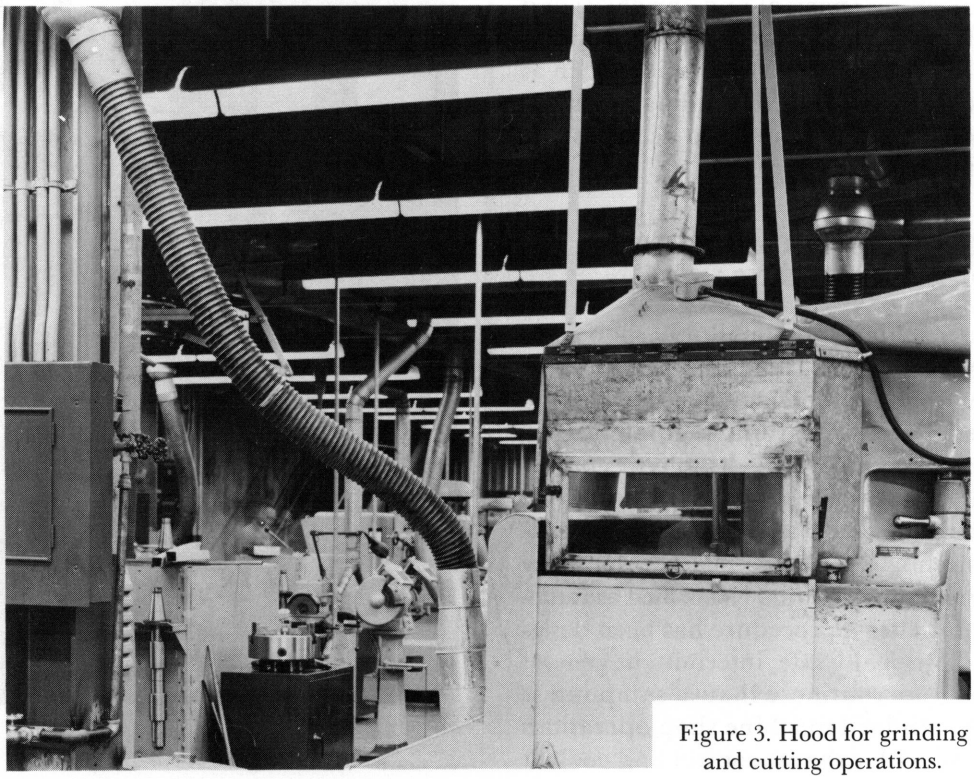

Figure 3. Hood for grinding and cutting operations.

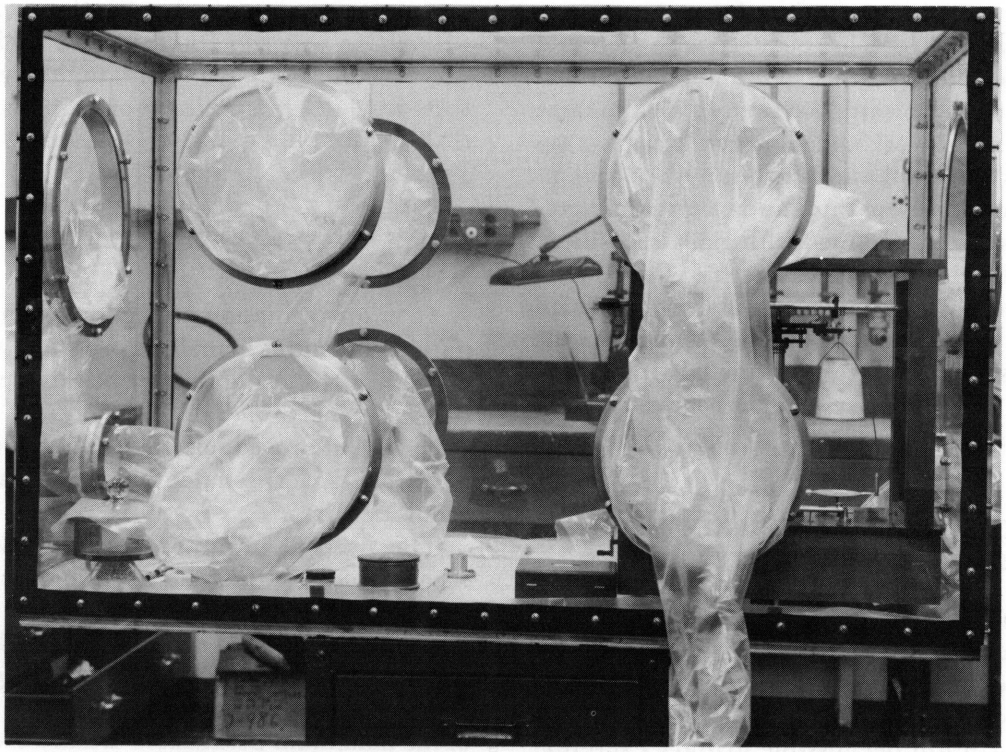

Figure 4. Dry box for powder metal.

Figure 5. "Hanford-type" hood.

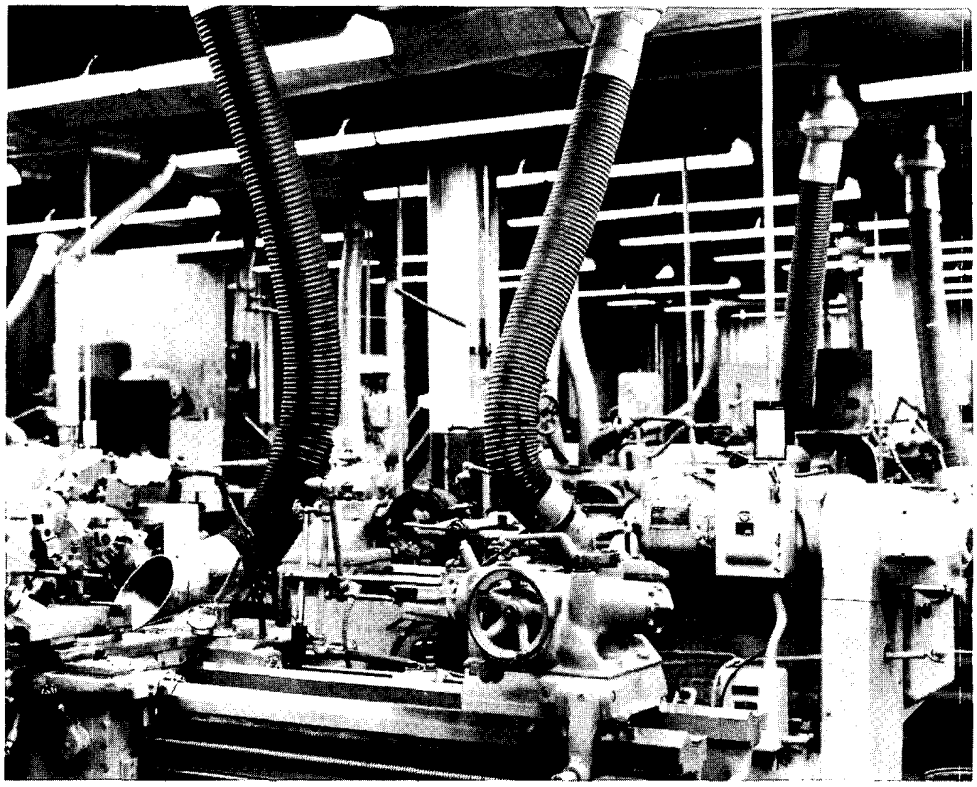

Figure 2. Lathe operation with exhaust control and coolant.

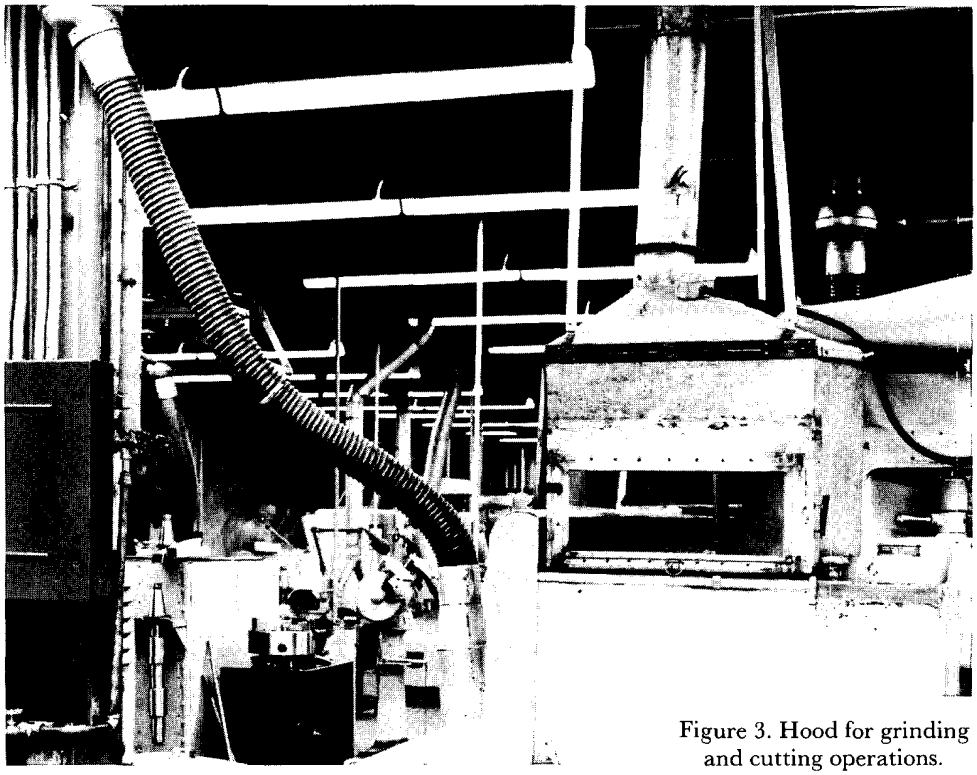

Figure 3. Hood for grinding and cutting operations.

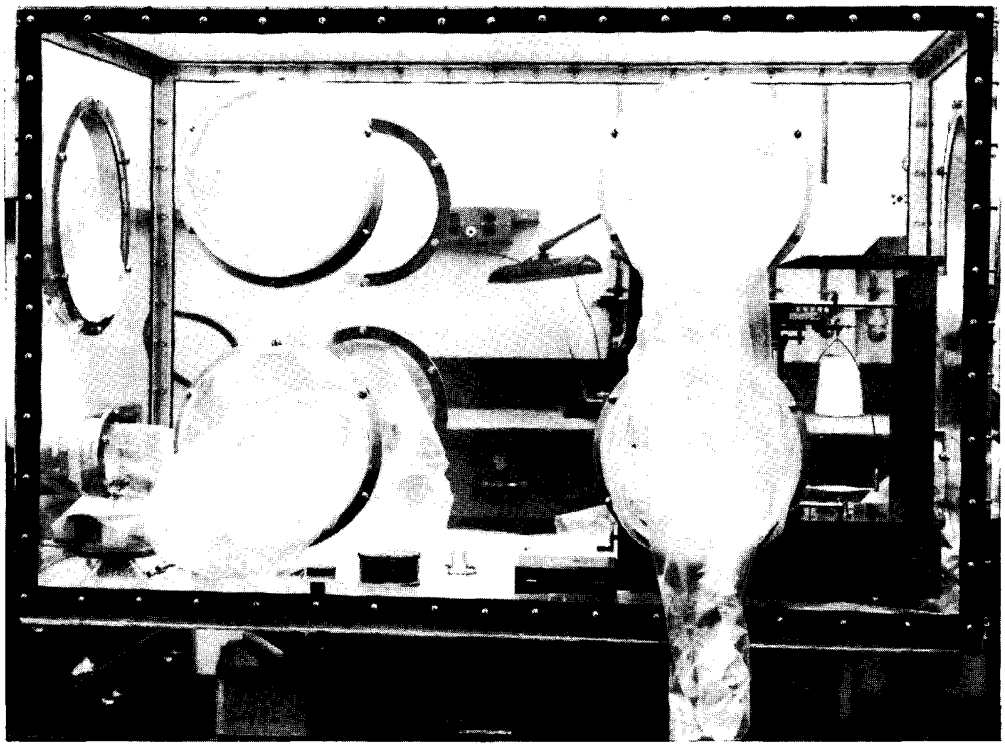

Figure 4. Dry box for powder metal.

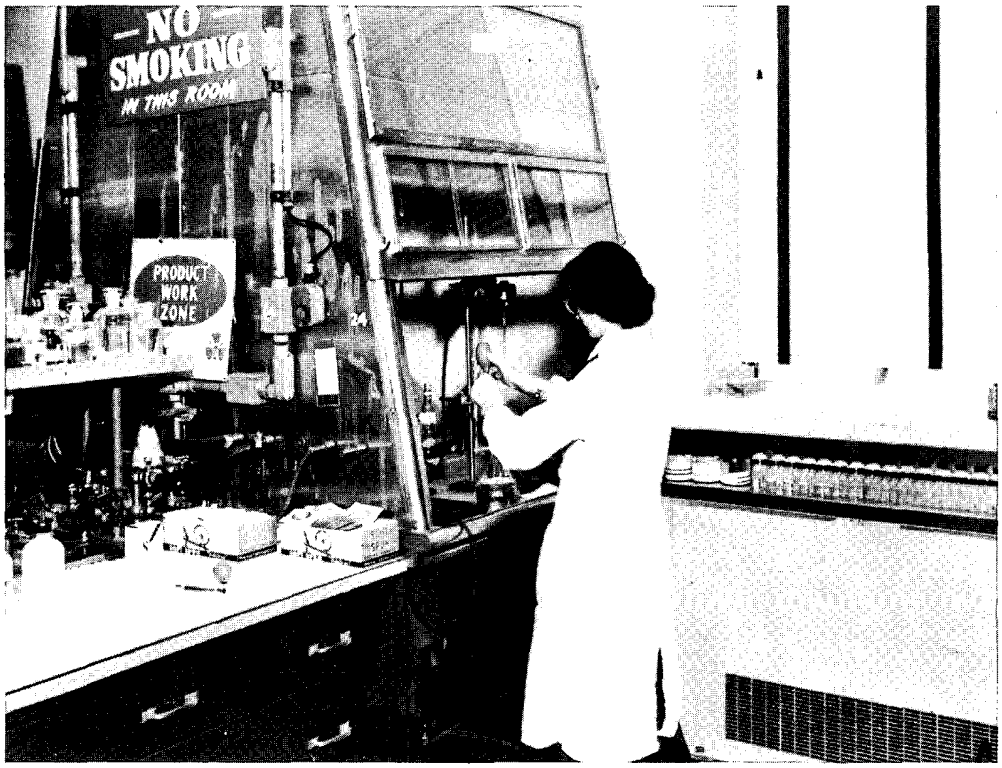

Figure 5. "Hanford-type" hood.

The air cleaning system[1] in the uranium processing areas at KAPL has changed with increased knowledge and experience in uranium processing. The original air cleaning system in the machine shop and metal working laboratory areas was designed for uranium and beryllium work and included a cyclone separator and electrostatic precipitator system combined with glass wool filters. In metallurgical laboratory areas, where metal powder work was being done, glass wool and high efficiency CWS filters were used. At present in the machine shop and metal working laboratories an adequate air cleaning system has been established using glass wool filters.

CALCULATION OF MAXIMUM PERMISSIBLE CONCENTRATIONS

Uranium presents either a chemical toxicity or radiological hazard, dependent primarily on the solubility of the compound and its mode of passage into the body. The KAPL Health Physics Department maximum permissible urinary excretion rates for uranium originally were calculated from data presented by the University of Rochester[2] and the NCRP subcommittee on internal dose.[3] The interpretation of our routine uranium bioassay program was simplified because of the absence of any soluble uranium or mixtures of soluble and insoluble uranium in the machine and metallurgy shops. Since our personnel had contact only with insoluble uranium, it was not necessary to obtain specimens at definite times in relation to specific work activities. The maximum urinary excretion for insoluble natural isotopic uranium was calculated to be 29 μg U/24-hr specimen. (This calculation was done as follows:

$$13\ \mu g \times 1000\ g \times 0.007 \times \tfrac{1}{2} \times \tfrac{2}{3} = \approx 29\ \mu g/day$$

where 13 μg = permissible amount of U per g of lung; 1000 g = weight of lung; 0.007 = fraction of lung deposit removed per day, based on a half-time of 100 days; $\frac{1}{2}$ is the fraction of the amount removed from the lung that is absorbed in body fluids; and $\frac{2}{3}$ is the fraction of the amount absorbed in body fluids that is excreted in the urine.)

Since the hazard arising from deposition of insoluble uranium compounds in the lung is a radiological one, the allowable concentration of enriched uranium in the lung should be inversely proportional to the activities of the natural and enriched mixtures; at KAPL the approximate proportion was 1 to 100. The allowable urinary excretion for enriched uranium, therefore, was 0.29 μg U/24-hr specimen or (150 dis/min/μg) (0.29 μg) = 44 dis/min/24-hr specimen.

BIO-ASSAY ANALYSES

To ensure radiation protection to employees, a natural uranium urinalysis program was established in 1949. A fluorimeter based essentially on the University of Rochester model was constructed, but proved unstable. KAPL is now using a Galvanek-Morrison fluorimeter with an average accuracy of $\pm 2\%$.

The natural uranium analysis involves the measurement of the fluorescent light emitted by uranium in fused NaF. Aliquots of 100 λ of urine were used with the University of Rochester fluorimeter with a sensitivity of 3.0 ± 2.5 μg U/24-hr specimen. Since this was greater than one tenth of the maximum permissible urinary excretion for insoluble uranium (29 μg U/24-hr specimen), the method was improved by use of a 100-ml aliquot, which increased the sensitivity of the analyses to <1.0 μg U/24-hr specimen.

Late in 1951, because of an increase in enriched uranium work at KAPL, an enriched uranium urinalysis program was established. The method involves the extraction of uranium from the inorganic salts remaining from an ashed 24-hr urine specimen with diethyl ether, aluminum nitrate being used as the salting out agent. The sensitivity of the analysis is 0.5 dis/min/24-hr specimen.

RECALCULATION OF THE MAXIMUM PERMISSIBLE CONCENTRATIONS

In March 1953 the Subcommittee on the Permissible Internal Dose summarized[4] the experimental and theoretical studies that had been completed in the atomic energy field in regard to standards for the maximum permissible amounts of radioisotopes in the human body. In 1955 the International Commission on Radiological Protection[5] issued a glossary of radiation safety standards. A summary of calculations based on the recommendations of these committees is given in Table 1.

Table 1

Summary of the Maximum Permissible Amounts of Uranium in the Body

Material and solubility	T_e, days	q, μC (a)	q, μC (b)	U excretion per 24-hr specimen corresponding to maximum permissible body burden: μC/day	
Sol. enriched U in kidney	30	0.04	0.08	6.0×10^{-5} or 1.2×10^{-4}	1.33×10^{2} dis/min
Insol. enriched U in lungs	120*	0.009	0.019	5.2×10^{-5} or 1.1×10^{-4}	1.15×10^{2} dis/min
Sol. natural U in kidney	30	0.04	0.08	6.0×10^{-5} or 1.2×10^{-4}	89 μg
Insol. natural U in lungs	120*	0.009	0.019	5.2×10^{-5} or 1.1×10^{-4}	77 μg

*KAPL experimental data indicate that this value is closer to 35 days.
(a) NBS *Handbook 52*, Equation G4.
(b) *Suppl. No. 6, ICRP Recommendations*, Equation C1.

Table 2

Typical KAPL Urinary Uranium Results*

Year	No. of urinalyses	No.** of uranium urinalyses	No. >1/10 MPL	No. <1/10 MPL but significant
1950	491	102	–	1
1951	1436	160	2	9
1952	1655	285	2	2
1953	2080	340	–	–
1954	2374	869	–	–
1955	1362	496	4	6
1956	824	377	–	4
1957	1526	500	1	1
1958†	793	380	–	–

*Natural uranium urinalysis begun 2/23/50; enriched uranium urinalysis begun 10/22/51.
**Number includes specimens submitted by outside contractors.
†Data to June 30, 1958.

TYPICAL KAPL BIO-ASSAY RESULTS

A compilation of KAPL bio-assay results is given in Table 2. The term "greater than one-tenth the maximum permissible" is derived from the maximum permissible lifetime radioactivity excretion rate given in Table 1 and has values of 8 μg U/24-hr specimen for insoluble natural uranium and 12 dis/min/24-hr specimen for insoluble enriched uranium. The term physically significant level refers to the minimum practical lower limit of reliable analytical and counting detectability of uranium in urine; its values are 3 μg/24-hr specimen for natural uranium and 5 dis/min/24-hr specimen for enriched uranium.

TYPICAL SUBCONTRACTOR BIO-ASSAY RESULTS

In 1953 the Schenectady Operations Office of the Atomic Energy Commission requested that KAPL provide bio-assay services to a fuel fabrication subcontractor. The facilities of the subcontractor were built for work with limited amounts of uranium, and significant air-borne concentrations were being encountered. The bio-assay data indicated a significant inhalation problem. During one monthly report period, 50% of the 16 specimens analyzed showed biologically significant concentrations, including two specimens with urinary uranium levels indicating a body burden greater than the maximum permissible. The

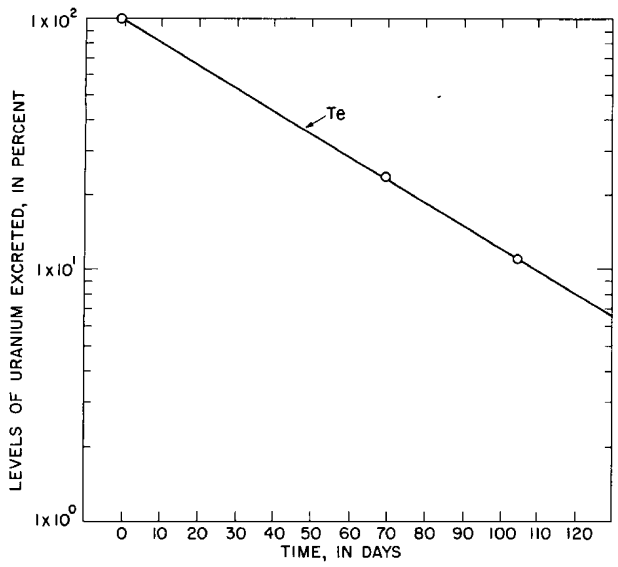

Figure 6. Average urinary uranium results obtained from KAPL subcontractor personnel.

material was insoluble enriched uranium. Figure 6 is a summary of the bio-assay data from several of the subcontractor personnel; this curve indicates an apparent effective half-time of 35 days.

DISCUSSION OF VARIABLES INVOLVED IN THE DETERMINATION OF INTERNAL DOSE

Many factors must be considered in the determination of the internal dose of uranium. The main items are (1) the manner of reception, i.e., inhalation, ingestion, incision, or absorption, and (2) the physical properties of the uranium, i.e., particle size and its effect on solubility, retention in the body, i.e., biological half-time, physical half-life, energy of the radioactive material, and deposition in the critical organ.

At KAPL each individual having a potential exposure to air-borne uranium is sampled for bio-assay to detect an internal dose. The frequency of analysis is based on the amount of contact he has with uranium. The classification of personnel for bio-assay is the responsibility of management, under Health Physics personnel direction.

For both acute and chronic exposure to uranium the placement for bio-assay is based on the following plan:

Group No.	Classification by number of exposures*	Frequency of Bio-assay: Natural uranium	Frequency of Bio-assay: Enriched uranium
0	Acute	Immediate	Immediate
1	20	Twice/year	Four times/year
2	10–20	Twice/year	Twice/year
3	<10	Once/year	Once/year

*Exposure is an inhalation, ingestion, or incision experience to uranium per 13-week period.

CONCLUSIONS

The KAPL uranium health program has been in existence since 1947. During this period no individual has had a significant exposure to air-borne particulate uranium. This is a result of proper design of facilities, control of exposure, and other radiological safety measures.

Prior to 1950 when the present site of the Laboratory was occupied, uranium work was carried on in temporary quarters where there was possible air-borne contamination. Personnel over-exposures were prevented, however, by proper air sampling and survey procedures and the utilization of respiratory protection when necessary to attain radiologically safe conditions.

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2. W.F. Neuman, *Urinary Uranium as a Measure of Exposure Hazard*, UR-82, July 14, 1949.
3. NCRP Subcommittee on Internal Dose.
4. National Bureau of Standards, *Handbook 52, Maximum Permissible Amounts of Radioisotopes in the Human Body*, 1953.
5. Recommendations of the International Committee on Radiological Protection, Dec. 1, 1954.

Interspecies Correlations

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Oak Ridge National Laboratory, Oak Ridge, Tennessee

The title of this paper is ambitious, but I shall present a point of view rather than arrive at definite conclusions. During the past several years I have had the privilege of working closely with Dr. Morgan and other committee members on the revision of handbooks for both the NCRP and the ICRP. At the moment I cannot say what will be in the final versions, as two of the numbers are still under discussion; however, I do not anticipate a change in permissible internal dose.

I think we all realize that man is our subject of interest, and when we study mice, rats, and dogs it is with the idea of applying the knowledge gained to man, although this may not be explicitly stated. Often this is not carried out consistently and carefully, and I respect the reluctance to reach wider conclusions than the data allow; nevertheless, our point of interest is still man. Working on these committees has made me very uncomfortably aware of this situation.

The point of view I wish to present is that we must consider the whole series of observations and experiments and not isolated ones. At this symposium many speakers have described actual human data accumulated either accidentally or by design. In every case the many uncertainties in the information have been emphasized. This information is of the greatest value, but it must be remembered that there were no controls, that human data usually cannot be calculated accurately, and that the human subjects are a very heterogeneous population. On the other hand, in the case of animal experiments, although the population is homogeneous and of a well-defined strain, controls are available, circumstances can be regulated, and known doses are carefully administered, one is uncomfortably aware that the rat is not a man and a man is not a rat, or even a dog, and again one is skeptical in applying these results. It seems to me that we must work from both ends of the spectrum of the experience we have had. I doubt that in the near future we will be able to obtain human data sufficiently accurate by itself to establish the rate safe for long-term use. The ingenuity and devotion brought to this task are a cause for hope; nevertheless, as things stand now, we must resort to animal data for some of the more important parameters needed for setting maximum limits.

One of the many ways of calculating the MPC is shown below, kidney toxicity being the criterion used.

$$\frac{(\mathrm{MPC})_a \times 2 \times 10^7 \times f_a}{[(\ln 2)/T] \times 300} = C;$$

$$(\mathrm{MPC})_a = \frac{C \times 10^{-5}}{T f_a}.$$

$(\mathrm{MPC})_a$ is in μC/cc; the next factor is air intake in cc/day for continuous exposure; and f_a is the fraction of the uranium inhaled which reaches the critical organ, in this case the kidney (there is some disagreement about the value of this); T is exposure time in days; and C is the critical concentration in the kidney in μC/day. The effect of particle size is not taken into account in this formula, because, although extensive work is being done on it both at Oak Ridge and at Rochester, the results to date are not definite enough for inclusion. The numerator of the first fraction is, therefore, the amount of uranium going into the kidney per day, and the denominator includes the factor 300 days, the usual figure used for the half-time of uranium in the kidney. There is also disagreement about this figure, and the formula may be changed to take this into account. This formula can also be used to calculate the kidney burden at any given time if a factor for the weight of the kidneys is included. If the critical concentration under the circumstances of interest is decided, then the above formula can be used to calculate $(\mathrm{MPC})_a$. It may be seen that the formula has three parameters: the level in the kidney assumed to be nontoxic, the fraction of the inhaled uranium going to the kid-

Table 1*

Kidney Toxicity of $UO_2(NO_3)_2 \cdot 6H_2O$
(Exposures were for 1 year to the levels indicated.)

	4.2 mg/m³		0.5 mg/m³		0.3 mg/m³	
	μg U/g kidney	Fraction affected	μg U/g kidney	Fraction affected	μg U/g kidney	Fraction affected
Dogs	1.7	5/5	1.0	21/25	0.5	0/17
Rats	5.6	14/37 21/37	1.6	5/146	1.4	2/103
Rabbits	1.4	3/4	0.9	4/6		
Guinea pigs	0.5	4/10	0.1	0/30		

*Data from Voegtlin and Hodge.[1]

ney, and the half-time in the kidney. None of these has been definitely determined as yet.

I would like to discuss some data from standard sources[1] on the relationship of uranium level in the kidney to toxicity. Table 1 lists values found at the end of a year of exposure.

The dogs in the first group showed an average of 1.7 μg U/g kidney, and the observation was mild to moderate renal tubular atrophy. The fraction 5/5 indicates that 5 dogs were subjected to the exposure indicated and that this symptom was noted in all 5 cases. In the second group the average concentration in the kidney was 1 μg/g, and only 21 of 25 dogs were affected; the symptoms were renal tubular injury and mild tubular changes terminally. Even in dogs with no symptoms during the year, on sacrifice some changes were indicated. At the lowest exposure level there was no sign of uranium poisoning in 13 dogs studied terminally, or in 4 serially sacrificed. One might say that for dogs the turning point appears to be at about 1 μg U/g kidney. At this point mild changes at least are observed.

However, it can be seen that rats react somewhat differently from dogs. They apparently take up more uranium, but it seems that their kidneys can stand more too. In the first group the average kidney concentration was 5.6 μg/g, which seems quite high compared to that of the dogs, and the assessment was very mild renal tubular changes in 14 out of 37 and moderate changes in 21 out of 37. Of a total of 150 animals used, 100 showed uranium poisoning; I do not know exactly what the symptoms were. In the next group the average kidney uranium concentration was 1.6 μg/g, and there was mild tubular degeneration with regeneration in 5 out of 146 and mild chronic interstitial nephritis. At the lowest exposure level slight renal tubular changes were seen in only 2 out of 103, and both occurred during the first two weeks of the experiment. Thus, although the rat seems to accumulate uranium to a greater extent in the kidney, there is some indication that this animal may tolerate the somewhat higher concentrations.

In the case of the rabbits and guinea pigs, I will not discuss the lowest exposure level. In the first group of rabbits, with a kidney concentration averaging 1.4 μg/g, the assessment was moderate tubular change in 3 out of 4. In the second group, with an average of 0.9 μg U/g kidney, mild renal tubular changes were found in 4 out of 6 with mild chronic interstitial nephritis. Apparently at this level there are very mild changes, and above this level moderate changes. The guinea pigs seem to accumulate the least. Even at the highest exposure level, the average kidney concentration was 0.5 μg/g, and the assessment was mild chronic interstitial nephritis in 4 out of 10. The second group had an average kidney concentration of 0.1 μg/g, and showed mild chronic interstitial nephritis with no sign of uranium poisoning.

It seems to me that the data compared here indicate a rather remarkable similarity in the kidney uranium levels at which mild kidney changes begin to occur in these animals. For man, unfortunately, I know of no accurate assessment of this level. Bassett[2] made some studies, but without a direct estimate of the exact concentration, and indica-

tions are that the general range was far above the animal figures given here. Some of the Boston patients had levels considerably above this, but every one of them showed considerable kidney damage. Thus, the two bits of human data available do not help settle the question of where this level is in man and how much it differs from that in animals.

I will not attempt to evaluate the animal data presented above. Unfortunately no indication was given of the actual spread in concentrations around the averages given. It would be interesting to know whether or not the animals with the greatest changes were the ones with the highest kidney levels.

We come now to the question of the half-time of uranium in the kidney, some estimates of which are listed below.

Mice	4-4.5 days
Rats	6-6.5 days
Man	10-16 days 10-12 days

The estimate for mice is taken from the work of Kieseleski. The rat figure I calculated myself from data in the Tannenbaum monograph, as this author did not mention an actual number; therefore, it may be in error. The estimate of 10 to 12 days for man is from Dr. Butterworth, who has discussed its source at this symposium. The figure of 10 to 16 days for man comes from very indirect mathematical analyses of excretion data on Boston patients and other human data of that kind, largely by Mr. Bernard and Mr. Fish.[3] Mr. Bernard originally calculated about 16 days, and Mr. Fish calculated similar figures of 12 to 15 days in various cases. Unfortunately, after this figure of 16 days had been arrived at on the basis of the Boston patients, another patient was studied whose data indicated a much longer half-time; whether this was due to the difference between individuals, to a high exposure level, or to just what, we do not know. We cannot say that we have definitive human data, but rather indications.

The figures discussed above for kidney concentration and half-time in the kidney could be used in some formula to calculate a possible $(MPC)_a$ for comparison with the ones previously suggested; but no choice can be made among them until further careful experimentation has been carried out. The various figures quoted are not really comparable. They are subject to large margins of error. Particularly in the case of the human data, there is a very large variation between individuals, and the number studied so far is small. We need to obtain reliable data on uranium retention in the kidney in the group of people we are interested in, and on the range of its variation, and on the fractions of these people falling into the different parts of the range. We need similar data on animals. In saying this, I am not unappreciative of the efforts of the experimentalists and of the health physicists. The human data so far collected are important and useful. I only wish to emphasize that we should consider all the data together, rather than make calculations from each individual set. It is unlikely that we will ever have sufficient human data, and animal data will have to be used in conjunction with it to determine the most reliable permissible exposure limits.

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Discussion of Session II Papers

Chairman, W.B. HARRIS

HAZEN: I am a consultant for some uranium mills in the West, the staffs of which are very much concerned about the problem of uranium hazards. I wanted to ask about the use of 70 $d/m/m^3$ as a limit and also 50 $\mu g/m^3$. Are these terms interchangeable?

HARRIS: Yes; 50 μg natural, normal uranium will yield 70 disintegrations per minute.

QUESTION: Nothing was said about whether the uranium in bone is eliminated in time.

HARRIS: I don't think we know.

SNYDER: Uranium does accumulate in the bone, and it has a longer half-time than in the kidney. In the bone we have a permissible limit of 0.3 rem/week, or, if we compare with radium, about 0.6 rem/week, and this standard has been accepted without any real experimental demonstration; it is a figure selected on a rather arbitrary basis by comparison with old radium figures. To my knowledge, accumulation slightly above this level has been demonstrated in the bone. The best experimental values for bone and for kidney indicate a lower concentration in kidney, so that the MPC for chemical toxicity turns out to be lower than the limit for radiation level; that is why we say the kidney is the critical organ.

HARRIS: In general, the material deposited in the lung gives a larger radiation dose to the lung than that deposited in the bone gives to the bone; therefore, it is generally assumed that it is lung dose which governs the radioactive toxicity. I am not sure that this is a fact.

HAZEN: In the West a good many of our water supplies contain more radioactivity than the law allows, yet people seem fairly healthy there. Has there been any study made of whether the people drinking such water are less healthy than people, say, in New York?

HARRIS: To date no extensive studies have been made. Unfortunately, the study of radiation toxicity requires a fairly large population group, and the number of people in the plateau exposed to these relatively elevated radium concentrations is comparatively small. Any effects would be very difficult to prove or disprove because of the smallness of the population.

QUESTION: Argonne National Laboratory is conducting an epidemiological investigation in the State of Illinois, where there is also a considerable amount of water containing 3 to 4 times the maximum permissible radium concentration. They are comparing the people drinking this water with the Chicago population, whose water comes from the lake and is very low in radium.

HOLADAY: I believe that in 1949 or 1950 an attempt was made to determine the level of naturally occurring alpha-active material in the bone in widely separated areas of the country, such as Denver, Salt Lake City, Birmingham, etc., to see whether any differences would be found. It is very difficult to obtain bone samples, and even more difficult to develop a method of alpha counting at the extremely low levels found, about 1 or 2 counts/hr. I don't know the present status of this study, but probably eventually some data will be obtained. The low levels would seem to indicate that not much radium had been absorbed from drinking water. There are some relatively high radium areas in the country; in the East some water contains up to 60 to 80×10^{-12} g radium per liter, and people have been drinking it.

QUESTION: The concentrations of heavy metals are usually expressed by industrial hygienists as milligrams per cubic meter of air. However, some talk in terms of microcuries, some in micrograms, some per liter, some per cubic meter; others talk about disintegrations per minute. Of course, these units can all be converted to each other, but it would be easier for the listeners to compare the results if all were uniformly reported. I would like to know why the speakers used different methods of reporting figures.

HARRIS: In *Handbook 52* the units are microcuries.

QUESTION: Why doesn't everyone follow it?

HARRIS: Because there is some difficulty in understanding or in directly converting microcuries of a material which exists in several isotopic forms.

A curie is generally considered to be of a particular element, and when there are several isotopes together, it is difficult to express the concentration in these terms.

Question: But today we were talking about uranium.

HARRIS: Natural uranium consists of three isotopes. Although it can be expressed in curies (as was done in the ICRP report, where a curie was specifically defined as being a curie of the mixture), this becomes cumbersome when the curie must be defined for each individual case.

QUESTION: Is there any reason for reporting per cubic meter, per liter, and per cc?

HARRIS: I think everyone could defend his own system.

QUESTION: In the paper by Meyer and Hyatt the statement was made that in urinalysis the use of a larger volume, i.e., a simulated 24-hr sample, increased the sensitivity of the method. I wonder if Mr. Meyer would enlarge on that.

MEYER: I should not have said it increases the sensitivity; it gives a more consistent result. What does increase sensitivity is the use of a 100-cc rather than a 20-cc sample, because then, to convert to liters, the multiplication factor is reduced to 10 from 50. The biggest error comes in counting; and this error is increased by a large conversion factor. Use of 24-hr samples seems to reduce the fluctuation. Figure 1 showed that daily spot samples collected every day for one week from two men fluctuated considerably. We now collect big samples and get more consistent results.

SESSION III

Correlation of Urine Data and Medical Findings With Environmental Exposure to Uranium Compounds

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INTRODUCTION

This discussion is based on analyses of air and urine samples taken in a uranium refinery in 1950 and 1951. The plant had been in operation since 1942, and exposures to air-borne uranium compounds in most operating areas were as high as or higher than in any other AEC plant.

Data from two plants of the refinery will be discussed in detail. In Plant 1, UO_2 was hydrofluorinated to UF_4, and in Plant 2, UF_4 was fluorinated to UF_6. The exposure in Plant 1 was largely to UO_2 and UF_4, which are considered insoluble uranium compounds; while in Plant 2, the exposure was mostly to UF_6 and its hydrolysis product UO_2F_2, which are considered soluble uranium compounds. Both plants operated 24 hr/day, with four shifts of operating personnel. The men worked 8-hr shifts for 5 days and were off for 48 hr, returning to a later shift.

A urine sampling program was initiated in January 1950. Spot samples were taken in pairs, one at the end of the work week, and the other 48 hr later, just before the men returned to work. Samples were taken every week, but usually not from the same group. An average of about one pair of samples per man per month was collected. The urine sampling program continued until late in 1951 when the two plants were permanently closed. A comprehensive air sampling program had been initiated in 1949 and continued until the plants were closed, so that for 1950 and 1951 air and urine data are available for the same group of men. Since both general air and breathing zone air samples were taken, time-weighted average air dust exposures could be calculated for each man in the plants. All air samples were collected on 1-in. circles of Whatman #41 filter paper at a flow rate of 20 l/min and analyzed by alpha scintillation counting. Samples too high for accurate alpha counting were analyzed fluorimetrically. The Health and Safety Laboratory has a sufficient number of sample records to permit the calculation of an average exposure for each group of operators by month for most months in 1950 and 1951. Figure 1 is a representative worksheet, indicating the procedure used to calculate the time-weighted average exposures. During this period, $\approx$3000 air samples were collected in Plants 1 and 2, and $\approx$1000 pairs of urine samples were collected from the operating personnel.

Sources of High Exposures

In Plant 1 most of the exposures resulted from handling dry UO_2 and UF_4 and showed up in breathing zone air samples. Accordingly, an individual's exposure would be expected to vary from the average for the group, depending on his work habits.

In Plant 2 most of the high exposures were due to general air contamination, since the plant was essentially one large open area, and contamination released in one part spread rapidly throughout. One major source of general air contamination was the cleaning of plugged UF_6 lines. These lines were ¼-in. copper tubing and often leaked slightly, especially at the joints. Humidity in the UF_6 stream hydrolyzed the UF_6 to UO_2F_2, which built up inside the tubes and often plugged them. To clean the lines, the operators disconnected them and dislodged the caked UO_2F_2 with a rod, releasing large amounts of UO_2F_2 and UF_6 into the room. The problem of plugging was especially serious during humid days, since high humidity increased the hydrolysis rate. The concentration of UO_2F_2 in the air of Plant 2 was frequently high enough to reduce visibility. General air samples were as high as 90,000 $\mu g/m^3$, and the average of 68 samples taken from mid-September to mid-October 1949 was 5300 $\mu g/m^3$. The time-weighted average air dust exposures calculated for Plant 2

NY FORM 144
(11-4-52)

January 1951

JOB ANALYSIS SHEET

OPERATOR: Plant 1 Loader 2 MEN/SHIFT: 3 SHIFTS/DAY: 8 MEN/DAY

OPERATION OR OPERATING AREA	TIME PER OPERA. (MIN)	OPERA. PER SHIFT	TIME PER SHIFT (MIN) (T)	NO. OF SAMPLES	CONCENTRATION, $\mu g/m^3$ LOW	HIGH	(C) AV.	AV. CONC.* TIMES TOTAL TIME (T×C)
Breaking, removing caps off 8 tubes and vacuuming	2.5	5	12.5	3	800	9,650	4,570	59,000
Unloading 4 trays UF_4 and dumping into trough	1.3	42	54.5	15	2,200	13,200	5,600	305,000
Loading 4 trays UO_2 and placing into tubes	2.6	42	109	9	300	8,600	4,240	462,000
Preparing and sealing 8 tubes	2.4	5	12	3	915	5,150	2,650	32,000
Weighing 1 drum UF_4 to correct weight and sealing	1.5	10	15	4	1,000	2,500	1,820	27,000
Changing drum under crusher	1	10	10	1			720	7,000
Cleanup of area	10	3	30	3	1,300	9,700	4,370	131,000
Loading room area			132	6			250	33,000
Lunch room 1-2			90	6			41	4,000
Locker room 1-2			15	12			125	2,000
			ΣT 480					Σ(T×C) 1,062,000

$\frac{\Sigma(T \times C)}{\Sigma(T)} = 2200$ $\mu g/m^3 = 44$ TIMES THE MAXIMUM ALLOWABLE CONCENTRATION

Adjusted to two significant figures.

Figure 1. Average exposure calculation sheet.

personnel were based on a large number of air samples and are good representations of the average exposures, but, considering the wide variation in concentration with time and location, are not always representative of the exposure of any individual. Thus anyone who spent any time in Plant 2, including supervisors, maintenance men, laboratory technicians, shipping and receiving men, etc, could have received one or more massive exposures.

JOB DESCRIPTIONS FOR POPULATIONS STUDIED IN DETAIL

Exposure to "Insoluble" Uranium Compounds

Plant 1 Loaders loaded UO_2 in trays and loaded the trays into tubes, which they sealed and removed to a storage area, where they were picked up by the Reactor Furnace Operators. The Loaders removed the tubes after the reaction and dumped out the UF_4, which they also weighed and sealed into drums. The remainder of their work day was taken up with cleanup time, lunch, and breaks.

Plant 1 Reactor Furnace Operators placed the tubes of UO_2 into the reactor furnaces, connected HF lines to the furnaces, removed the lines after completion of the reaction, and removed the hot tubes of UF_4 to the storage area. The remainder of the work day was spent in the furnace area, and at lunch and breaks.

Exposure to "Soluble" Uranium Compounds

Plant 2 Operators tended the reaction cells which fluorinated UF_4 to UF_6. They did little more than check gauges and observe the progress of the reaction, except when they had to unplug clogged UF_6 lines. Their exposure was almost entirely to "soluble" uranium.

Plant 2 Redistillation Operators purified the bottled UF_6 collected from the reaction furnaces by redistilling it. They connected and disconnected UF_6 lines and unplugged them when they clogged. Their exposure was almost entirely to "soluble" uranium.

Exposure to Both "Soluble" and "Insoluble" Uranium Compounds

Plant 2 Loaders filled trays with green salt, inserted the trays into the reaction cells, sealed the cells, and connected the HF lines. After the reaction was complete they removed the lines, opened the cells, and removed the trays. They also cleaned out the ice traps and cleaned up spilled green salt. They were exposed to both "insoluble" UF_4 when handling the green salt and to "soluble" uranium compounds from the general Plant 2 air.

MEDICAL PROGRAM AND FINDINGS

All employees received pre-employment physical examinations and repeated laboratory examinations from the time of their employment. Prior to 1950 no cases of renal injury had been found; there had been an occasional case of abnormal urinary findings, but these had not been followed up. However, in the early months of 1950, one worker in Plant 1 and five in Plant 2 showed abnormal urines, and a follow-up study was initiated on these men. The medical tests, described in detail by Eisenbud and Quigley,[1] indicated no diminution in renal function. The men were removed from further contact with uranium when the urine abnormalities were first encountered, and, in all cases in which follow-up was possible, urinary findings returned to normal within a matter of months. The job descriptions and dust exposure of these men are summarized in Table 1 and are discussed in detail in the next section.

For Plant 1 personnel exposed to "insoluble" uranium compounds, no evidence was found of renal injury, nor have any lung pathology or blood changes been observed. Two men from Plant 1 died of nonoccupational causes, and the autopsy findings were reported by Eisenbud and Quigley.[1] The measured uranium concentrations in the two men's lungs were 0.06% and 0.41% of the concentrations predicted from the plant air sampling data and the inhalation experiments on dogs conducted at the University of Rochester.

Unpublished autopsy data from other sources[2] also indicate much lower concentrations of uranium in the lung than would be expected. The data are for men with exposures of several years duration in uranium refining and machining operations, and, although detailed exposure histories are not available, there is no doubt that the exposures were real. The lung concentrations for these people were only slightly above the levels for unexposed people in the same geographic areas.

These fragmentary data indicate the need for further studies. (1) Increased efforts should be

Table 1

Job Description and Dust Exposure for Six Men With Abnormal Urine Findings

Case	Job description	January-June 1950 calculated av exposure to "soluble" U, $\mu g/m^3$	Months between start of employment and first urine abnormality
K	Loader, Plant 2	3500	9
M	Loader, Plant 2	3500	19
R	Loader, Plant 2	3500	13
C	Operator, Plant 2	300	22
P	Shipping and Receiving, Plant 2	100	16
H	Operator, Plant 1	<50	91

made to obtain additional information on tissue concentrations of exposed populations, and (2) studies should be made to determine the retention factors, deposition sites, and elimination rates for air-borne uranium particulates in humans, and the effects of particle sizes, shape, solubility, etc., on these factors.

AIR AND URINE DATA FOR GROUPS EXPOSED TO "SOLUBLE" URANIUM COMPOUNDS

Inhaled "soluble" air-borne particulates are eliminated rapidly following exposure. Since "soluble" natural uranium is considered to be primarily a nephro-toxic agent, and since the urinary uranium concentration of an exposed individual reaches a peak very shortly after exposure, after-work or before-weekend urines are generally considered to be indicative of exposure. In the discussion to follow, before-weekend urines are correlated with air exposures. The monthly average air and urine concentrations are summarized in Table 2.

Discussion of Plant 2 Loaders (including Cases K, M, and R, Table 1). During the first half of 1950 the 29 Plant 2 Loaders were exposed to a time-weighted average dust exposure of 5500 $\mu g/m^3$ total uranium, of which 3500 $\mu g/m^3$ was "soluble" uranium. This was the highest exposure to "soluble" uranium of any group in the plant, and the group had been at this level or higher for several years. Respirators were made available to the Plant 2 workers and were generally worn by the Loaders when handling the trays of green salt and ash but rarely at other times. They probably were not used enough of the time to have a material effect on the air exposures to "soluble" uranium compounds.

During the period January to June 1950, these men contributed 100 before-weekend urine samples. Figure 2, which shows the distribution of these samples, indicates that 50% were >900 $\mu g/l$, 25% >2000, and 10% were >4000. The highest sample was 13,200 $\mu g/l$. While none of the 29 men showed any clinical symptoms or diminution of renal function, 3 of the 29 (Cases K, M, and R), a possibly significant fraction, showed abnormalities in their urine which could have been pretoxicity signs.

Discussion of Plant 2 Operators (including Case C, Table 1). In 1950 and 1951, 35 men worked as Plant 2 Operators. These men were exposed to "soluble" uranium and had monthly average exposures ranging from 61 to 470 $\mu g/m^3$ (see Table 2). During this time they contributed 298 before-weekend urine samples (see Figure 2), of which 50% were >260 $\mu g/l$, 25% were >540, 10% were >1000, and the highest sample was 6780 $\mu g/l$. These men showed no clinical symptoms and only one case of urine abnormality (Case C). Considering the variability of working habits and plant conditions in Plant 2, Operator C could have been exposed to considerably more than the average exposure for a significant period of time.

Discussion of Plant 2 Redistillation Operators (no cases). In 1950 and 1951, 10 men worked regularly as uranium hexafluoride Distillation Operators. These men were exposed to "soluble" uranium and had monthly average exposures ranging from 91 to 930 $\mu g/m^3$ (see Table 2).

During this time they contributed 122 before-weekend urine samples (see Figure 2), of which 50% were >400 μg/l, 25% were >850, 10% were >1600, and the highest sample was 6060 μg/l. These men showed no clinical symptoms and no urine abnormalities.

Discussion of Shipping and Receiving Personnel (including Case P, Table 1). Relatively few data were collected on these people. Too few urine samples were collected for meaningful discussion. Air samples indicate that the average dust exposure did not exceed about 100 μg/m³ of "soluble" uranium.

Discussion of Other Personnel Exposed to "Soluble" Uranium (no cases). In addition to the operating personnel already discussed, about 10 supervisory personnel, 15 maintenance men, 10 laboratory men, and 16 security guards were exposed to "soluble" uranium compounds during part of their work day. None of these showed any clinical symptoms or urine abnormalities.

Discussion of Plant 1 Reactor Furnace Operators (including Case H, Table 1). In 1950 and 1951, 14 men worked as Reactor Furnace Operators. They were exposed almost exclusively to "insoluble" uranium, and their monthly average exposures ranged from 80 to 240 μg/m³ (see Table 2) of total uranium. During this time they contributed 85 before-weekend samples (see Figure 3), of which 50% were >40 μg/1, 25% were >75, 10% were >130, and the highest sample was 348 μg/1. These men showed no clinical symptons, and, ex-

Table 2

Monthly Average Dust and Urine Concentrations

	"Soluble" uranium						"Insoluble" uranium					
	Plant 2 Operators			Plant 2 Still. Op.			Plant 1 Loaders			Plant 1 Reactor Fur. Op.		
		Urine, μg/l			Urine, μg/l			Urine, μg/l			Urine, μg/l	
Month	Air, μg/m³	B*	A	Air, μg/m³	B	A	Air, μg/m³	B	A	Air, μg/m³	B	A
1950												
1	440	970	410	620	1100	860	9700	160	68	170	–	–
2	370	840	330	640	830	83	–	110	40	150	49	18
3	–	1200	130	–	1900	140	9700	120	48	–	110	35
4	170	890	79	420	2100	140	9500	140	33	–	130	7
5	–	560	63	–	1500	91	–	200	40	–	71	13
6	200	490	74	320	670	84	8100	150	70	230	67	21
7	140	900	51	170	130	70	4400	210	45	110	65	22
8	–	150	34	260	230	47	3700	50	28	–	33	18
9	–	200	23	–	290	77	–	–	–	–	45	8
10	170	240	54	–	400	45	2600	81	49	140	30	17
11	470	440	23	160	–	–	2500	61	35	140	30	17
12	81	390	37	310	360	45	2800	51	15	180	21	11
1951												
1	88	260	29	500	830	53	2200	40	20	160	40	8
2	180	310	32	940	580	41	2600	42	21	110	30	13
3	300	280	39	160	330	46	3600	49	29	120	12	6
4	130	240	43	120	370	59	3600	49	29	120	12	6
5	–	180	30	240	190	35	1800	54	25	–	53	15
6	140	160	30	90	390	25	1800	54	25	200	75	22
7	–	780	48	–	450	48	1300	–	–	–	50	12
8	100	410	130	250	270	81	–	–	18	–	67	11
9	130	280	33	150	340	58	1300	–	21	88	–	18
10	61	250	41	91	190	39	–	–	–	–	–	–
11	–	230	32	–	260	40	–	–	–	–	–	–
12	160	200	32	–	235	42	–	–	–	–	–	–

*B indicates before-weekend urine and A, after-weekend urine.

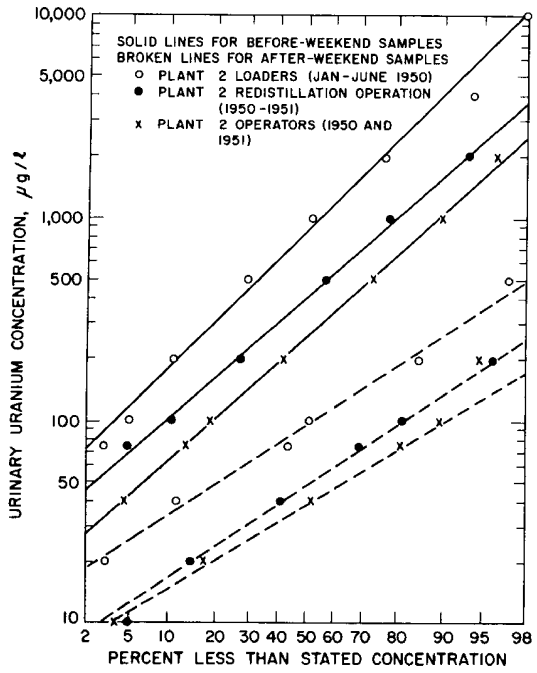

Figure 2. Distribution of before-weekend and after-weekend urinary concentrations for men exposed to "soluble" uranium compounds.

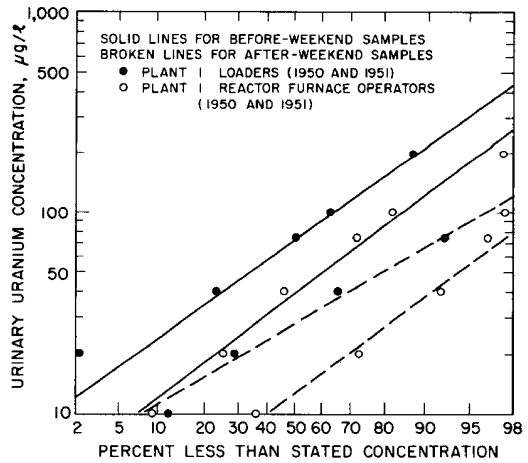

Figure 3. Distribution of before-weekend and after-weekend urinary concentrations for men exposed to "insoluble" uranium compounds.

cept for Case H, no urine abnormalities. It is questionable whether the urine abnormalities of Case H were due to "soluble" uranium exposure, since his exposure was exclusively to UO_2 and UF_4.

Discussion of Plant 2 Urinary Uranium Levels. The urinary uranium concentrations for the Plant 2 operating personnel were considerably higher than any currently accepted levels. There is a wide range of acceptable levels in the uranium industry, with some installations considering any detectable amount too much and others using levels as high as 300 μg/l as an upper limit. It can be seen from Figure 2 that 80% of the Plant 2 Loaders had before-weekend urines >300 μg/l. A review of the literature did not yield urine values of this magnitude for people with chronic exposures to air-borne uranium. It is interesting to compare these urine levels to those found in experiments in which human subjects were injected with "soluble" uranium. Two such experiments have been reported, one on 6 hospital patients (group R) at the University of Rochester[3] and the other on 5 terminal patients (group B) at the Massachusetts General Hospital.[4,5] The dose data, excretion rates, and medical findings for these experiments are summarized in Table 3. Patients 1, 2, and 3 of the Rochester group are omitted because they had low doses and no positive findings. For all the hospital patients the excretion rate increased until it reached a maximum at about 1 hr after injection, remained at about that level until about 5 hr, and then decreased rapidly. From the excretion rate curves of these patients, the rate (in μg/l) at 2 to 4 hr after injection was calculated for comparison with the before-weekend urinary concentrations of the Plant 2 workers (Table 2). Table 3 shows that positive medical findings were reported only for patients 6R, 2B, 4B, and 5B. Patient 6R showed only a trace of albuminuria; patient 2B showed both a positive catalase and a trace of albumin. Patients 4B and 5B, with the highest doses, showed positive findings for urinary catalase, albumin, and casts, and for serum nonprotein nitrogen. Patients 1B, 2B, 3B, and 5B died 2.5, 74, 556, and 139 days following injection, respectively; autopsies showed their kidney pathology to be negative.

AIR AND URINE DATA FOR GROUPS EXPOSED TO "INSOLUBLE" URANIUM COMPOUNDS

In Plant 1 the Reactor Furnace Operators and the Loaders were exposed to UO_2 and UF_4, which are considered to be "insoluble."

"Insoluble" uranium is considered an internal hazard as an alpha emitter, the critical organ being the lung. Bale,[6] of the University of Rochester, has recommended 25 μg uranium per gram tissue as a maximum allowable level for lung dep-

Table 3

Uranium Excretion and Medical Findings Following Injection of Hexavalent Uranium

Patient	U injected		Calculated excretion rate 2 to 4 hr after injection		Medical findings				
					Urine			Serum nonprotein nitrogen	Kidney pathology
	μg	μg/kg	μg/hr	μg/l	Catalase	Albumin	Casts		
1B*	5,000	97	280	4,400	–	–	–	–	–
2B	5,900	130	590	9,300	+	trace	–	–	–
3B	4,300	70	390	6,300	–	–	–	–	–
4B	11,200	170	1,000	16,000	+	+	+	+	no test
5B	15,800	280	1,100	17,000	+	+	+	+	–
4R	1,900	30	200	3,200	–	–	–	–	no test
5R	2,700	42	300	4,800	–	–	–	–	no test
6R	3,900	70	400	6,400	–	trace	–	–	no test

*Other tests on the group B patients included electrocardiograms and liver function, hematologic, and glucose tolerance tests; all were negative.

osition, based on a tissue dose of 300 mrem per week to the lung and an RBE of 10 for alpha-particles. Using this figure, and a half-time of uranium in tissue of 100 days, Neuman,[7] of the University of Rochester, calculated that 60 μg/24 hr would be excreted due to this lung deposition. Since uranium is also stored in other body tissues and bone, he suggested that an excretion level of 100 μg/24 hr (equivalent to about 67 μg/1) would be a safe urine concentration for people exposed to "insoluble" uranium. He suggested that this limit be applied to Monday-morning (after-weekend) urine samples, which would indicate body burden elimination rather than the elimination of recently inhaled "soluble" uranium, which would be present in a before-weekend or Friday sample. Actually, even a Monday-morning sample is not indicative of body burden, as will be brought out below in the discussion of urine sampling, and leads to high estimates when used to estimate body burden.

In this section, after-weekend urinary concentrations are correlated with air dust concentrations. The air dust concentrations are not actual exposures in these cases, since respirators, providing an unknown measure of protection, were usually worn during the dusty operations of loading and unloading trays. Plant 1 Loaders wore respirators during operations representing about 60 to 80% of their calculated exposure, but, since in practice respirators are rarely used to their best advantage, their exposure was probably at least 25% of the calculated average air concentration. The Plant 1 Reactor Furnace Operators did not normally wear respirators.

During 1950 and 1951, 14 Plant 1 Loaders worked in average air concentrations ranging from 1300 to 9700 μg/m³ (Table 2). Between January and June of 1950 the average air concentrations ranged from 8100 to 9700 μg/m³, so that for this period the Loaders' *actual exposures* were in the *milligram* per cubic meter range, even allowing for a 75% reduction by the use of respirators. The distribution of the Loaders' 1950-1951 after-weekend urinary concentrations, shown in Figure 3, indicates that 50% were >28 μg/l, 25% were >46, 10% were >70, and the highest sample was 108 μg/l. During the same period 14 Reactor Furnace Operators working in air concentrations ranging from 80 to 240 μg/m³ (Table 2) contributed 87 after-weekend urine samples (Figure 3), of which 50% were >12 μg/l, 10% were >40, and the highest sample was 60 μg/l.

Figure 4 is a plot of after-weekend urinary concentrations versus monthly time-weighted average air dust concentrations. The calculated regression line is shown. For men working in average air concentrations of from 1250 to 4400 μg/m³, only one urine sample in 56 was >67 μg/l, while for men working in air concentrations of 8100 to 9700 μg/m³, 8 samples in 33 were >67 μg/l. Urine samples that really indicated body burden, such

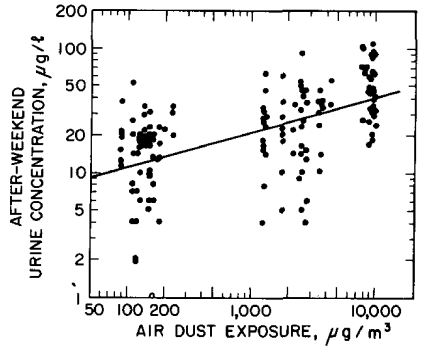

Figure 4. Air dust exposure to "insoluble" uranium compounds versus after-weekend urinary concentrations.

as samples taken a week or more after the last exposure, would have been lower than the after-weekend values plotted in Figure 4.

URANIUM EXCRETION RATE FOLLOWING EXPOSURE

The before-weekend and after-weekend samples discussed so far represent only two points in time on a uranium excretion curve. To define the manner in which uranium excretion varies with time after exposure ceases, additional points in time would be required. Such data were not available from the records of Plant 1 and 2, but data of this type for other people exposed to uranium are available. Figure 5 is a plot of urinary uranium excretion versus time for three men who had single massive exposures to uranium. The validity of a straight-line relationship on log-log paper is supported by the injection studies at Rochester[3] and Boston[4,5] discussed above. The excretion curves for these studies were also straight-line plots, starting at about 5 hr after injection and continuing to the lower limit of measurement for the Rochester group, and to expiration for the Boston group. Two observations about the curves in Figure 5 will be useful in the discussion of urinary uranium sampling: (1) the excretion rate continues to drop steadily for about 3 weeks following exposure, and (2) the rate of decline is a function of time, so that a concentration at any subsequent time should be predictable if the excretion rate at day 1 is known.

The second conclusion was tested on the data from Plant 1 and 2 personnel. If the before-weekend sample is considered to show the concentration at day 1, the second sample should correspond to day 3 and should depend on the concentration at day 1. An investigation of the data showed that, as the before-weekend value increased, the relation between before-weekend and after-weekend samples became more constant. Two factors contributed to the variable results among the samples having lower concentrations. One was the analytical precision, which at that time was about ± 10 μg/l for samples <100 μg/l, and the second was the influence of body burden excretion. The variations in length of employment and previous exposure history resulted in a wide variation of body burden excretions. The after-weekend samples included both recently absorbed uranium and body burden excretion. To eliminate this second source of variation in the relation of before-weekend to after-weekend results, the before-weekend results were compared with the difference between before-weekend and after-weekend results. Figures 6 to 9 show that these variables have a constant relation both for exposure to "soluble" uranium compounds (Figures 6 and 7) and for exposure to "insoluble" uranium compounds (Figures 8 and 9), although the absolute amount of uranium in the urine is much lower in the latter case. Figure 10 is a similar plot of values from the populations of Figures 6 and 7 that were too high to fit on those graphs and also of high samples from the Plant 2 Loaders. These values fall on the same line as the lower values. The 48-hr declines for the three single massive exposure cases of Figure 5 are also plotted in

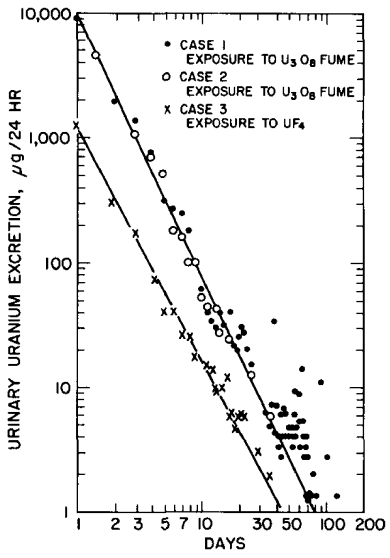

Figure 5. Excretion of uranium in urine after a massive single exposure.

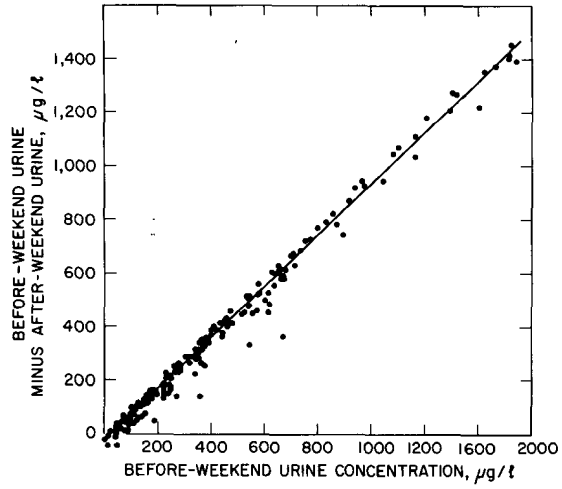

Figure 6. Decline in urinary uranium concentration over a 48-hr weekend leave, Plant 2 Operators.

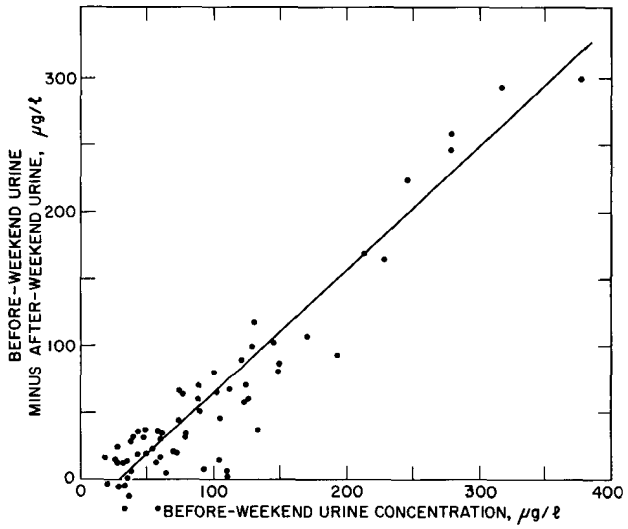

Figure 8. Decline in urinary uranium concentration over a 48-hr weekend leave, Plant 1 Loaders.

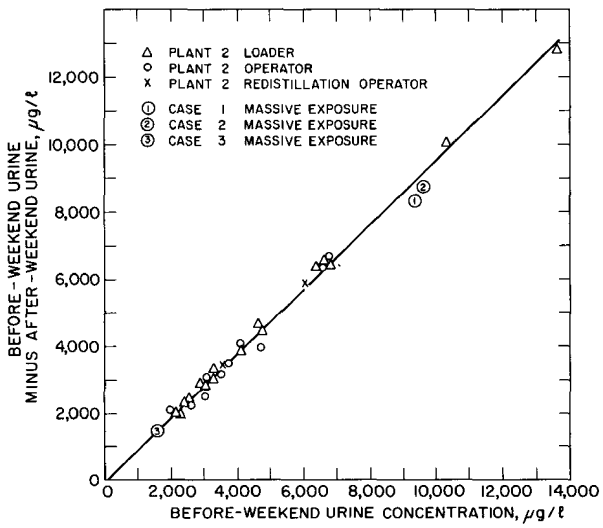

Figure 10. Decline in urinary uranium concentration in 48 hr in three cases having before-weekend concentrations >2000 μg/l and in three cases of massive exposure.

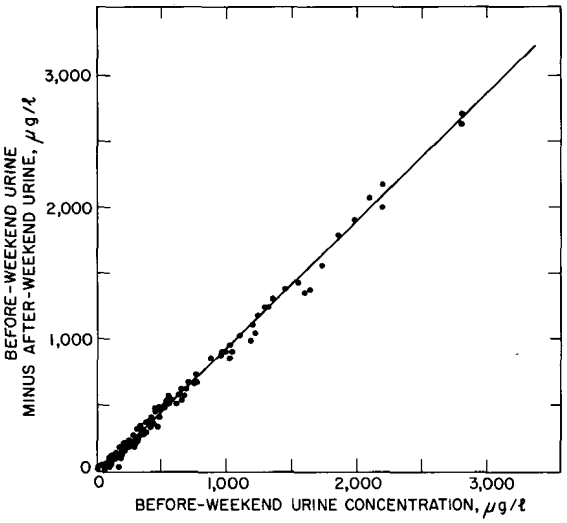

Figure 7. Decline in urinary uranium concentration over a 48-hr weekend leave, Plant 2 Redistillation Operators.

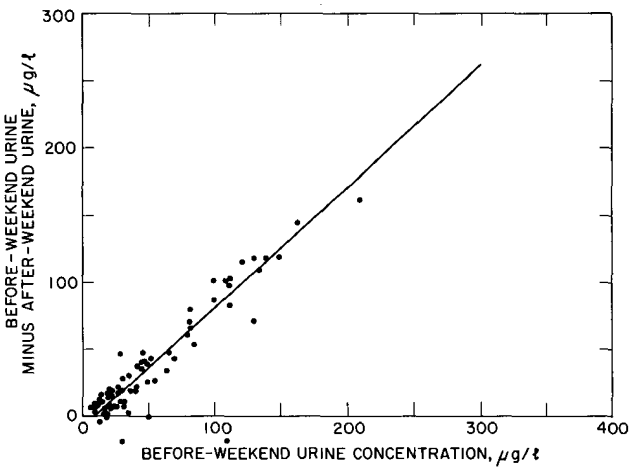

Figure 9. Decline in urinary uranium concentration over a 48-hr weekend leave, Plant 1 Reactor Furnace Operators.

Figure 10 and are in good agreement with the Plant 1 and 2 data.

Apparently this consistent decline in concentration in 48 hr for both groups represents the elimination of soluble uranium. In the case of exposure to "insoluble" compounds, it appears likely that the very small dust particles, with their relatively large surface area, are soluble. This soluble portion of the "insoluble" dust is a small fraction, since the ratio of air to urine concentration is much higher than for exposure to "soluble" compounds. If the decline in urine concentration represents the elimination of soluble uranium, and if the decline continues for several weeks, as suggested by Figure 5, then an after-weekend urine sample overestimates the long-term body burden excretion if there was exposure to soluble uranium in the weeks preceding the sample. Only if the before-weekend and after-weekend samples do not significantly differ, will the after-weekend samples be representative of long-term excretion.

URINE SAMPLING FOR URANIUM AS A MEANS OF CONTROL

In order for urine sampling to be a useful method of control, a urine sample should be indicative either of exposure or of body burden. If a given sample cannot be compared with the established standards and evaluated in relation to those standards, then urine sampling is of no value. With the air and urine data from Plant 1 and 2, an evaluation of urine sampling can be attempted.

Urine Sampling for Men Exposed to "Soluble" Uranium Compounds – Evaluation of Before-Weekend Urine Sampling. Since soluble uranium is eliminated very rapidly by the body, only a urine sample taken during or immediately after exposure could be representative of exposure. Conceivably, for men exposed to a constant atmospheric concentration of "soluble" uranium, urine results might correlate closely with air concentration. For more typical industrial conditions, where exposure varies with the operation performed and with time, the urine concentration at the end of the work day will depend on whether the peak exposure was early in the morning, at midday, or just before quitting time. It could even be influenced by a high exposure earlier in the week. A before-weekend urine concentration of 100 μg/l could have been caused by an exposure

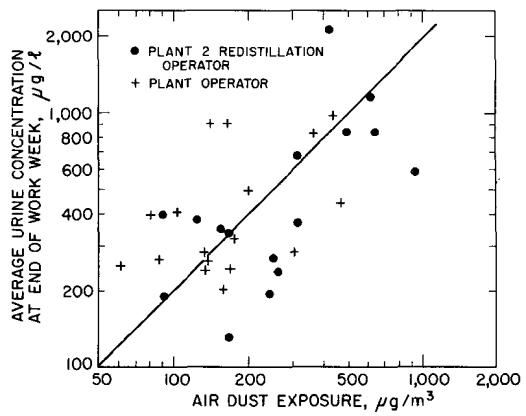

Figure 11. Air dust exposure to "soluble" uranium compounds versus average of before-weekend urinary concentrations.

of 500 μg/m³ two days before, an exposure of 250 μg/m³ the day before, or an exposure of 50 μg/m³ on the day the sample was taken. Figure 11 is a plot of average before-weekend urine results for a number of Plant 2 workers versus average dust exposure. Butterworth and McLean[8] of the United Kingdom Atomic Energy Authority reported average air and urine concentrations of uranium of 14 μg/m³ and 33 μg/l respectively over a 2-month period for a man* exposed only to UF_6 and UO_2F_2. If the line on Figure 11 were extrapolated downward, these data would fall on it. The authors state that similar values have been found for other individuals working in the same group. The air samples were taken daily for the entire 8-hr shift. The urines were collected daily at the end of the work shift, and therefore correspond closely to the before-weekend samples of Plant 2.

It can be seen from Figure 11 that there is considerable scatter in the Plant 2 points. If the 283 individual urine results were plotted instead of average values, the scatter would increase. A regression analysis of these data indicates a positive correlation between individual urine values and the average air dust exposures, i.e., urine values tend to increase as air exposure increases. However, statistical analyses also indicated that the exposure cannot be calculated from a given before-weekend urine sample to better than an order of magnitude. It appears, therefore, that before-weekend urine samples cannot be used to

*The individual died of coronary thrombosis 12 hr after leaving work, and autopsy findings are discussed in the report. See also Butterworth's paper in Session I of this symposium.

obtain a reliable estimate either of average exposure or of body burden.

Urine Sampling for Men Exposed to "Insoluble" Uranium Compounds – Evaluation of After-Weekend Samples. After-weekend urine samples are usually taken in order to estimate long-term steady-state excretion of uranium stored in body tissues, and compared to a level of 100 μg/day (67 μg/l) suggested by Neuman.[7] It has been shown that an after-weekend sample is influenced not only by steady-state excretion, but also by material inhaled within the previous week or two. For example, an after-weekend (Monday-morning) urine value of 25 μg/l could result from any of the following combinations: a steady-state contribution of 5 μg/l plus 20 μg/l resulting from a Friday exposure of 200 μg/m^3; or a steady-state contribution of 15 μg/l plus 10 μg/l resulting from a Friday exposure of 100 μg/m^3; or a steady-state contribution of 25 μg/l if there had been no appreciable exposure within the previous week. To compound the confusion, the relative contributions would have been altogether different if the major exposure had occurred on some day earlier in the week than Friday. Another source of error is the assumption that a spot urine sample is representative. Thus, an after-weekend value of 25 μg/l could be obtained from an individual whose steady-state excretion corresponds to a value somewhat <5 μg/l or >25 μg/l, or anything in between. However, even if it were granted that an after-weekend sample of 67 μg/l represented a maximum permissible lung deposition, it would mean that average air exposures in the milligram per cubic meter range would be allowable, since exposures of that magnitude were required to produce after-weekend urine concentrations >67 μg/l in the Plant 1 workers. It hardly seems reasonable to use urine sampling to detect exposures of that order.

It would appear that the only urine samples which would give a reasonably reliable indication of steady-state excretion would be after-vacation samples.

At first glance, to those familiar with uranium metabolism, the after-weekend urine concentrations in Plant 1 may seem much lower than expected from the exposures received. Although no definitive correlation has ever been established between air exposure to "insoluble" uranium and urine concentration, many installations have proposed correlation factors for specific operations or groups of operations. These limited correlations are often consistent within themselves, but at wide variance with each other. Perhaps the discrepancies arise because the various installations are not comparing the same variables. Most investigators have attempted to correlate exposure to urine concentration without taking into consideration the time of urine sample collection. Some installations collect samples before a shift starts, some at the end of a shift; some collect samples before, or after, or even during a weekend leave. Since it has been demonstrated that a man's urine concentration often declines by an order of magnitude during the course of a weekend, it is not surprising that different correlations are found when different collection times are used. For the same exposures on a Monday to Friday work week, the urines collected on Monday morning, or Monday afternoon, or any other morning or afternoon, would differ from each other, and cannot be compared. Other major variables in any air-urine correlation include the percent retention in the lung of the inhaled air-borne dust and the solubility of the dust in the lung. Unfortunately, the effect of these variables has received scant attention in the past and is poorly understood.

Several prerequisite conditions would have to be met before any correlation could be expected between different sets of uranium air and urine data, including collection of urine samples at the same time interval after exposure (or correction to the same time) and the knowledge that exposure was to compounds with similar physical properties, such as particle size distribution and solubility. If these conditions were satisfied, the data from various installations handling "insoluble" uranium could be compared.

SUMMARY

Air and urine data and medical findings covering a 2-year period have been presented for employees of two plants of a uranium refinery. The Plant 1 personnel were exposed to "insoluble" uranium compounds, the Plant 2 personnel to "soluble" uranium.

Monthly average air dust exposures to "soluble" uranium compounds were as high as 3500 μg/m^3. For 29 men exposed at this level, the median before-weekend urine concentration was 900 μg/l; 25% were >2000 and the highest was 13,200 μg/l. None of these men showed any diminution of renal function, but 3 of the 29 had abnormal urine findings. For men exposed to lower air con-

centrations, no clinical symptoms and only occasional urine abnormalities were found.

Monthly average air dust concentrations of "insoluble" uranium compounds were as high as 9700 $\mu g/m^3$. Although respirators were worn at some operations, they could not have reduced average exposures by more than 75%, so that some exposures were in the milligram per cubic meter range. For 8 men so exposed, 8 of 33 after-weekend urines contained >67 $\mu g/l$, the highest showing 108 $\mu g/l$. For men who worked in average air concentrations up to 4400 $\mu g/m^3$, only 1 urine sample in 118 was >67 $\mu g/l$.

The urinary excretion rate following heavy exposures was found to drop steadily for several weeks, and the rate of excretion at any subsequent time was found to be a function of time and of the excretion rate during the first day following exposure.

No useful correlation could be found between air concentration of "soluble" uranium compounds and before-weekend urine concentration, or between air concentration of "insoluble" uranium compounds and after-weekend urine concentration. In addition, it was shown that an after-weekend urine sample does not provide an accurate indication of uranium body burden.

ACKNOWLEDGMENTS

Grateful acknowledgment is hereby made to W.B. Harris, Chief, Industrial Hygiene Branch, Health and Safety Laboratory, for suggesting this paper and for assistance in its preparation; to Dr. J.A. Quigley, National Lead Company of Ohio, for providing medical data; to P.B. Klevin of the New York Operations Office, who did most of the survey work, for providing background information; and to the personnel of the Analytical Branch of HASL, who performed all the analyses.

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Correlation of Urine Data With Environmental Exposure to Uranium

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The attempt to correlate urinary uranium excretion with the on-the-job uranium exposure received under the usual working conditions has often been difficult because of the many variables inherent in the jobs and the individuals. While a relatively large amount of information pertaining to exposure-excretion relationships is available from laboratory and theoretical studies, the opposite is true with respect to actual job studies, mainly because of the difficulty in maintaining experimental control under actual working conditions.

At the Westinghouse Electric Corporation, Bettis Plant, a manufacturing group engaged in the fabrication of U^{235} fuel alloys appeared to lend itself to the high degree of control necessary for an on-the-job study. In the course of their work, these men were exposed to low concentrations of air-borne uranium. In-plant air sampling and uranium urinalysis over several years have shown that none of these men received exposures to uranium in excess of the recommended maximum permissible concentration, and no job-related health problems had been observed. Since past experience with this group indicated that exposures were minimal and subclinical, it was anticipated that a high degree of sensitivity in sampling and analysis would be necessary to make any subtle relationships more obvious. With this in mind, an investigation of the uranium intake versus excretion relationship was made.

Fifteen men, aged 19 to 53 years, had been exposed primarily to enriched uranium during their 16 to 102 months of employment in the fuel alloy shop at the Bettis Plant. The complete enclosure and separation of the fuel alloy shop from other uranium processing areas helped to insure that any exposures observed were obtained in the study shop only. Movement of personnel, equipment, and materials in and out of the study shop was controlled and limited. The manufacturing process consisted basically of melting, forging, rolling, acid pickling, shearing, surface conditioning, machining, and inspection of enriched uranium fuel alloys.

Uranium intake was based on that fraction of each breathing zone sample consisting of particles $<3\ \mu$ in diameter. It was assumed that particles $>3\ \mu$ in diameter would not be mobilized with sufficient speed to contribute significantly to the urinary uranium concentration. Fractionation of the samples was accomplished by pre-impingement of the particles $>3\ \mu$ in diameter prior to collection of the <3-μ-diameter particles on molecular filters. It was also assumed that 25% of the <3-μ-diameter fraction was re-exhaled or remained suspended in the tidal lung air, 50% was lost by impingement on bronchi or bronchioles, and 25% reached the alveolar spaces. It was further assumed that only ½ of that reaching the alveolar spaces (12½% of the <3-μ fraction) was absorbed by the blood. Normally, these assumptions are made with respect to the total dust sample, and it is quite likely that a dust consisting only of particles $<3\ \mu$ in diameter might behave differently. For the purposes of the comparisons made in this study, these assumptions are not considered binding. It would be relatively simple to modify the results on the basis of a different pattern of dust distribution in the lung.

Motion studies were made on each job to provide a basis for estimating work rates in Btu/hr and, subsequently, respiratory ventilation rates in liters/min. Shop records, worker records, and observer records provided a good breakdown of the time spent by each employee on each job. The exposure time, air-borne uranium concentration, and respiratory ventilation data provided the basis for predicting the weighted average uranium intake.

Each individual was given a dose of citrocarbonate during the luncheon break at noon to insure alkaline infusion of the uranium in the kidney. Daily urine samples were collected from each individual at the end of the work shift (4 hr after

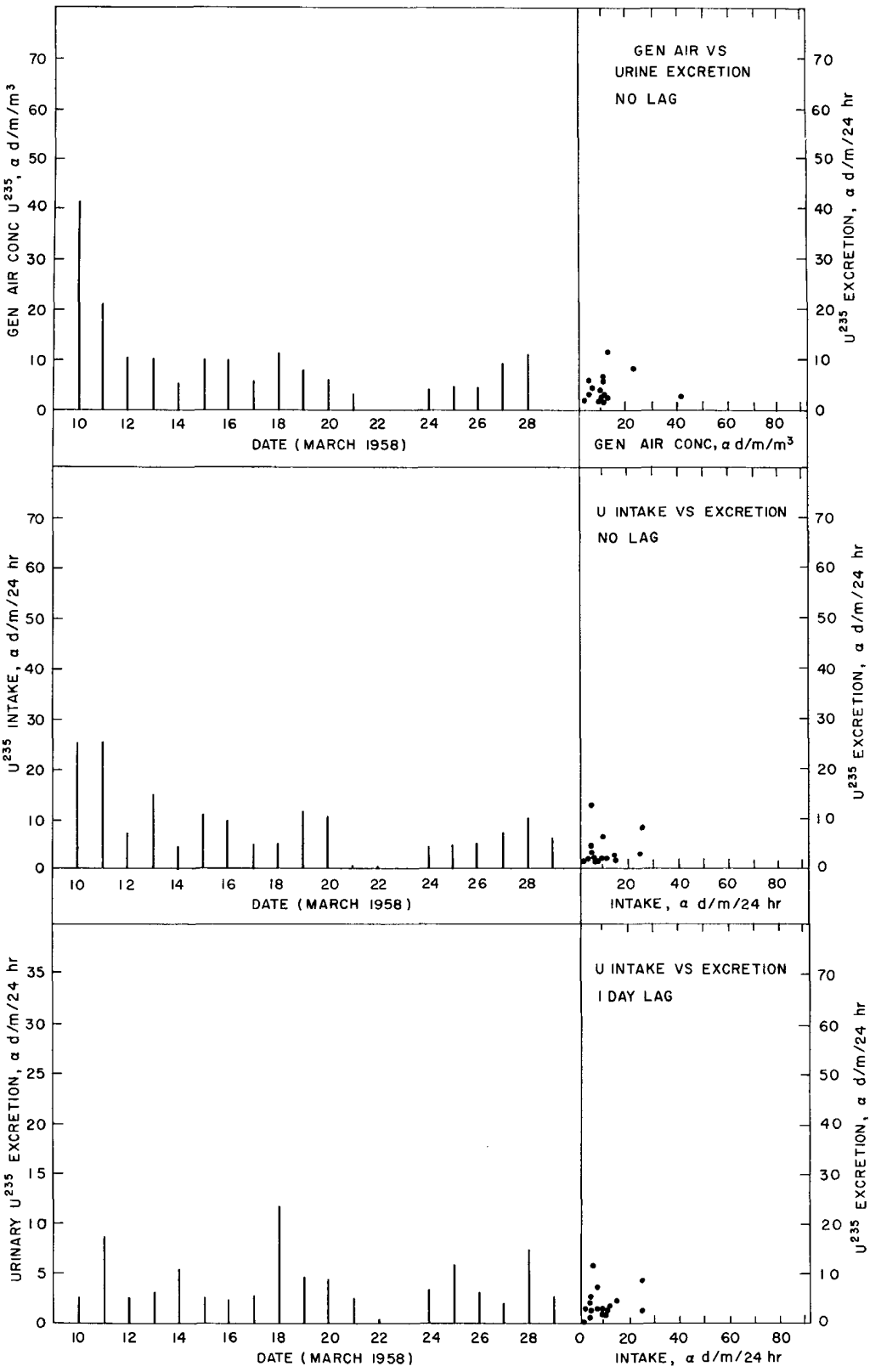

Figure 1. Average group data, charge preparation and couriers, alloy shop, Bettis Plant, March 1958; 2 men.

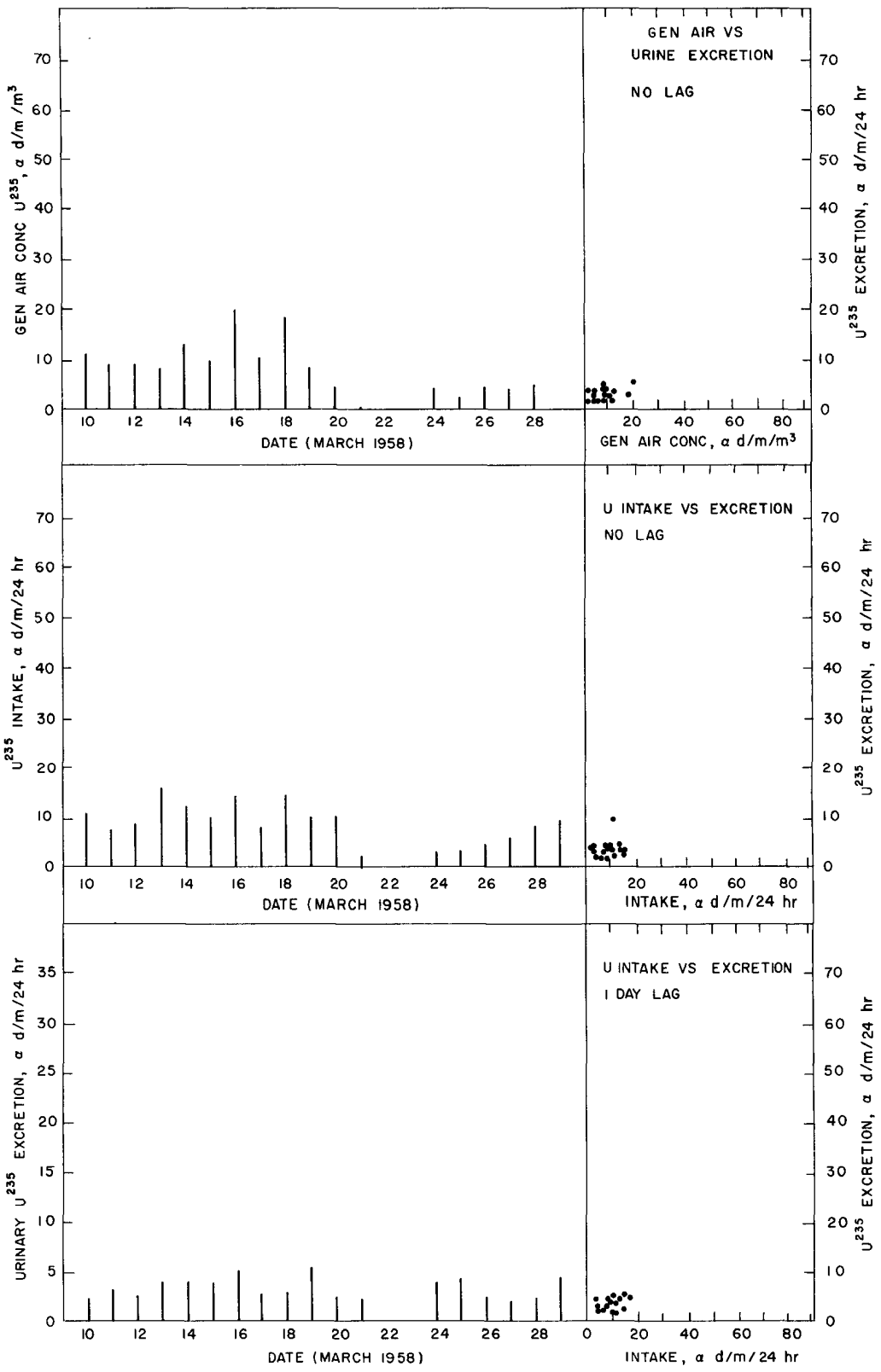

Figure 2. Average group data, melting area, alloy shop, Bettis Plant, March 1958; 2 men.

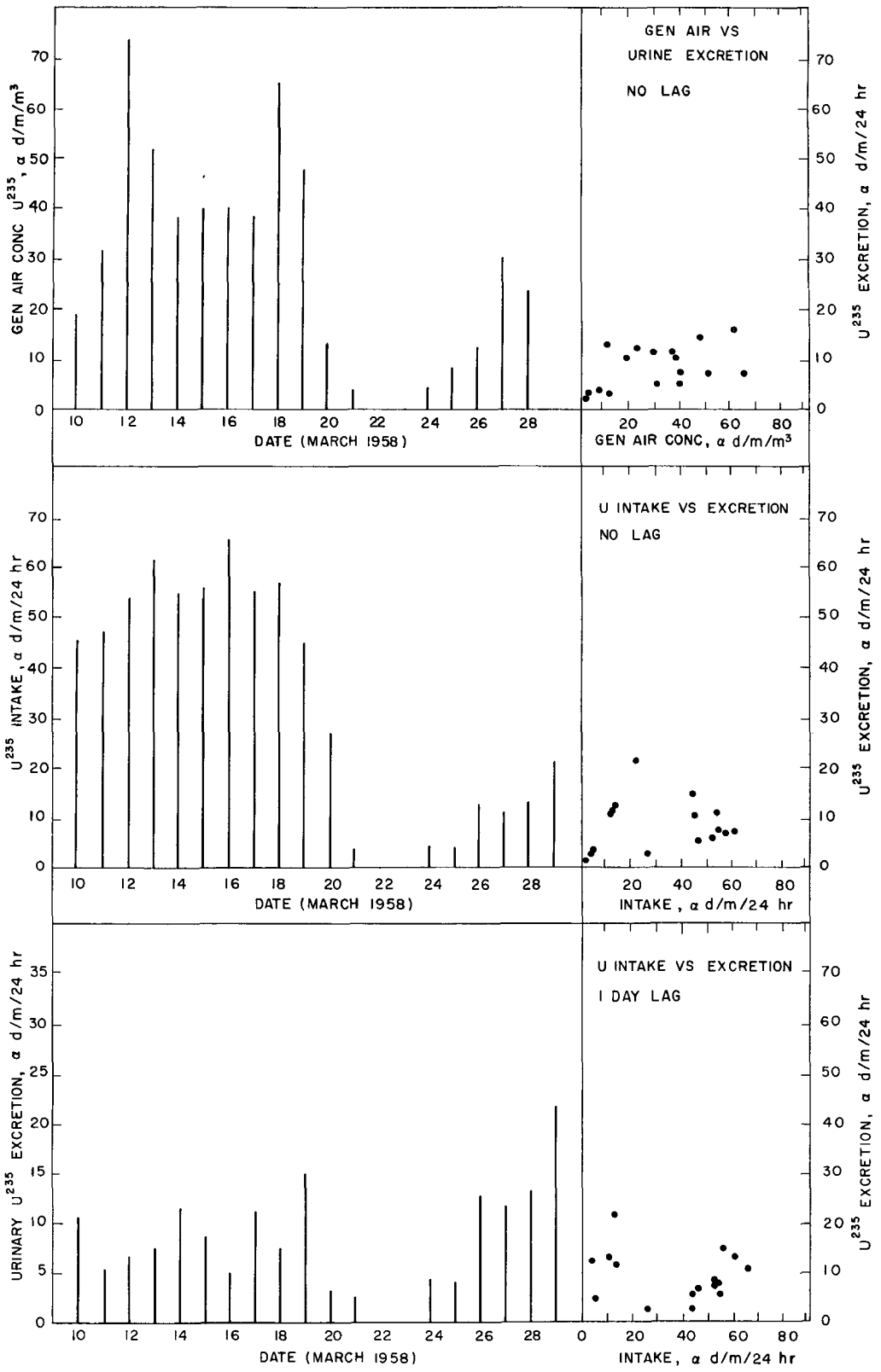

Figure 3. Average group data, metal working area, alloy shop, Bettis Plant, March 1958; 2 men.

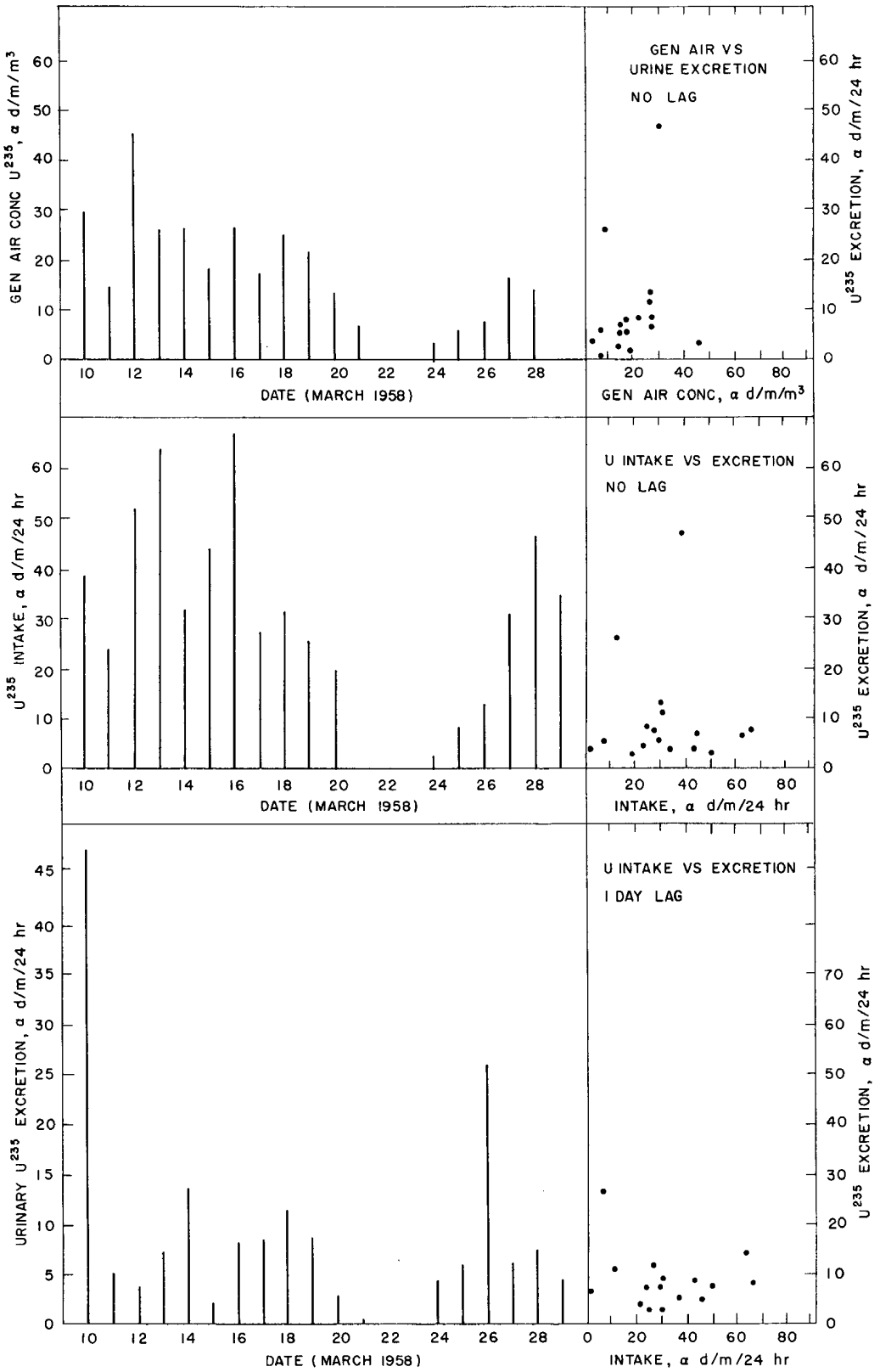

Figure 4. Average group data, ingot conditioning area, alloy shop, Bettis Plant, March 1958; 1 man.

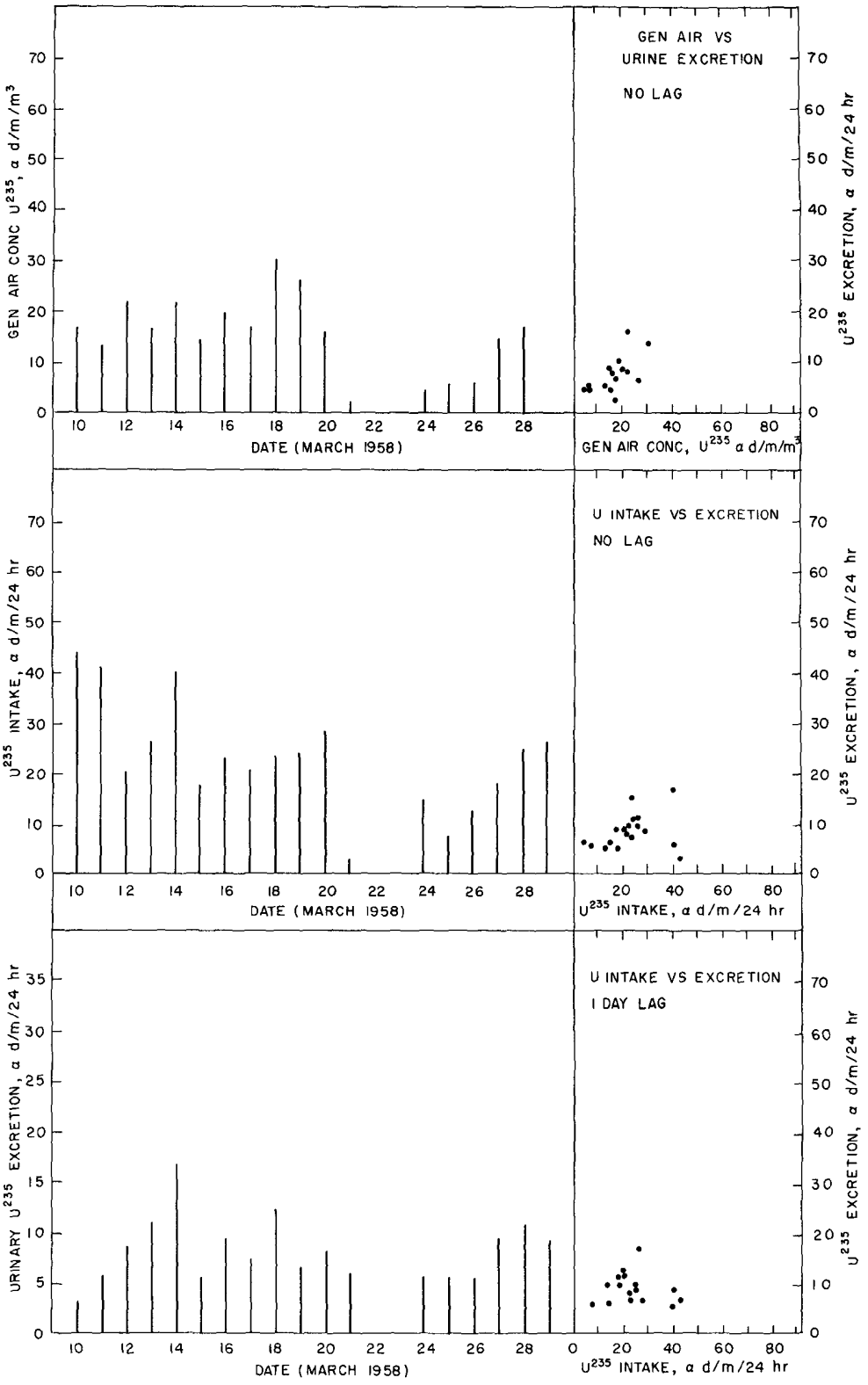

Figure 5. Average group data, special operations, alloy shop, Bettis Plant, March 1958; 3 men.

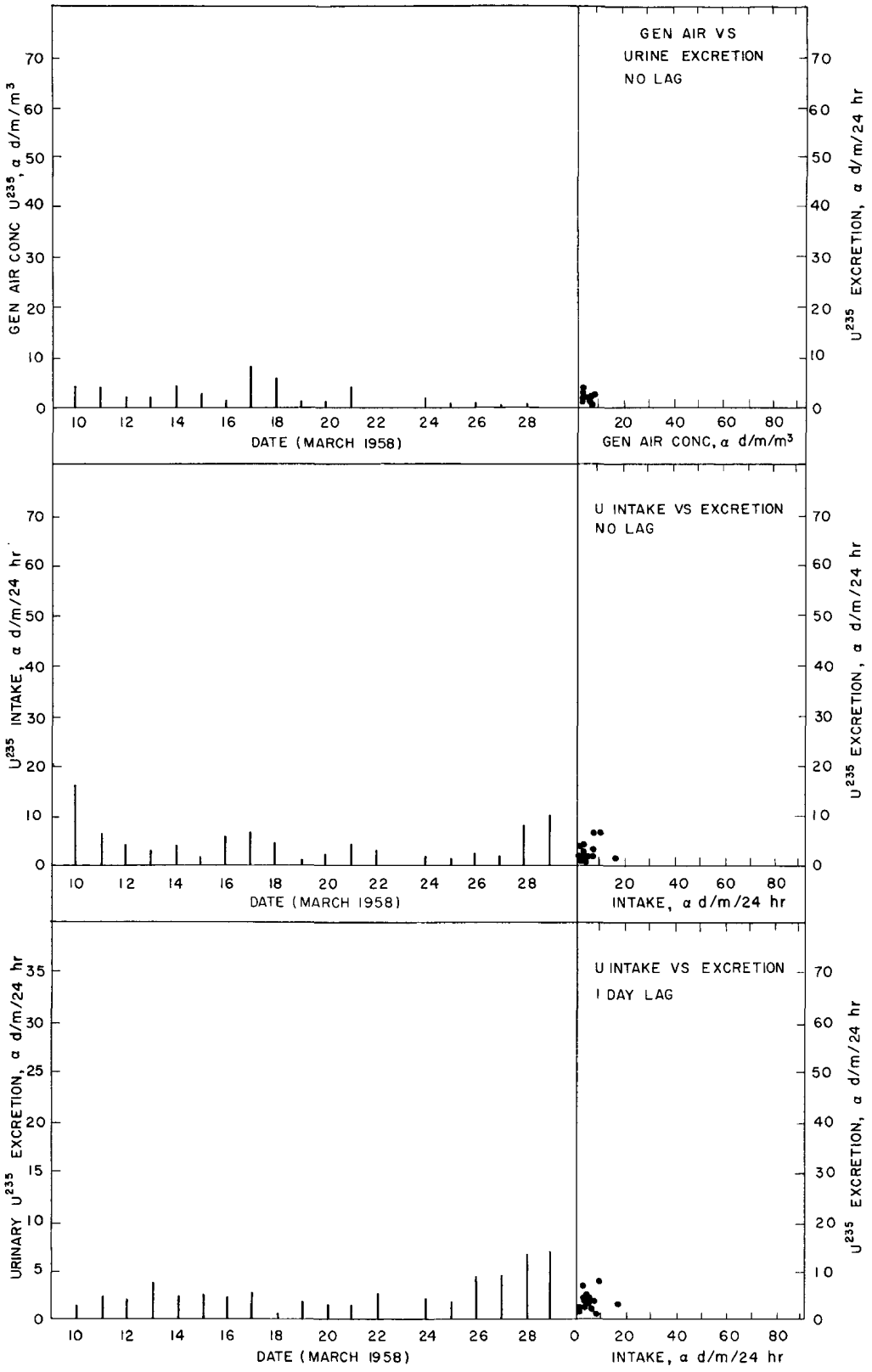

Figure 6. Average group data, machine area, alloy shop, Bettis Plant, March 1958; 2 men.

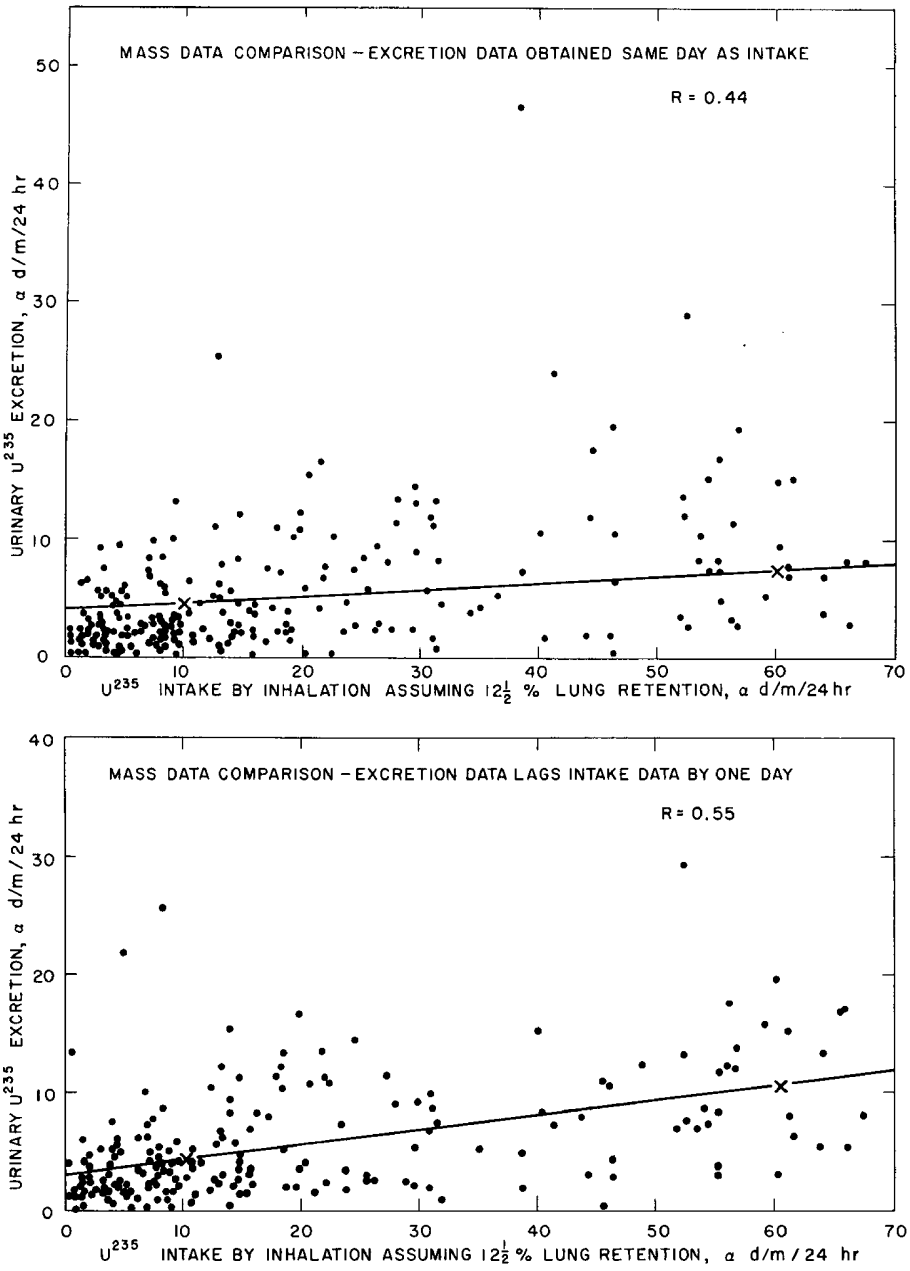

Figure 7. U^{235} intake versus excretion in a 15-man study group, March 10-29, 1958.

taking citrocarbonate). The uranium was extracted by complexing with diethyl ether in the presence of calcium nitrate and ferric nitrate salting agents. The samples were counted for uranium alpha activity in a gas flow proportional counter, and the results were corrected to uranium excreted per 24 hours.

There had been no uranium work in the study area for $\approx$6 weeks prior to the study, hence uranium surface contamination, background uranium air concentrations, and background uranium urine levels had returned to extremely low values. This study was conducted during the 19-day period March 10 through 29, 1958, during which the usual uranium fabrication jobs for this shop were in progress.

Graphs of urinary uranium excretion and uranium intake were used to make comparisons on an individual, group, and mass basis. Attempts were made to correlate the uranium excretion of a par-

ticular day with the uranium intake of the same day and with the uranium intake of the previous day. Additional comparisons of intake versus excretion were made with the intake based on the concentration of uranium as determined by general air samples collected by 13 area monitoring stations. There was no fractionation of these samples according to particle size other than that occurring naturally as a result of the distance of the sampling station from the point of dust generation.

The figures show the relationships among the various data obtained. From a review of these data the following observations can be made.

1. From the individual data shown in Figures 1 to 6 it can be seen that the general patterns of intake and excretion tend to follow the general work pattern, but it is difficult to make a quantitative correlation between the uranium intake and the uranium excretion. Because of the obvious scatter, no attempt was made to dignify the correlation plots by using the line of best fit or by using correlation coefficients. Note that when the uranium intake increased, the urinary uranium excretion increased rapidly. Removal of an individual from exposure resulted in a rapid decrease in urinary uranium levels.

2. The mass data shown in Figure 7 give a better indication of the intake-excretion relationship and also show that a higher correlation is evident if the excretion is compared to the intake of the previous day.

3. Figure 8 shows a comparison of the average concentration of uranium in the general shop air with the average urinary uranium excretion. It is

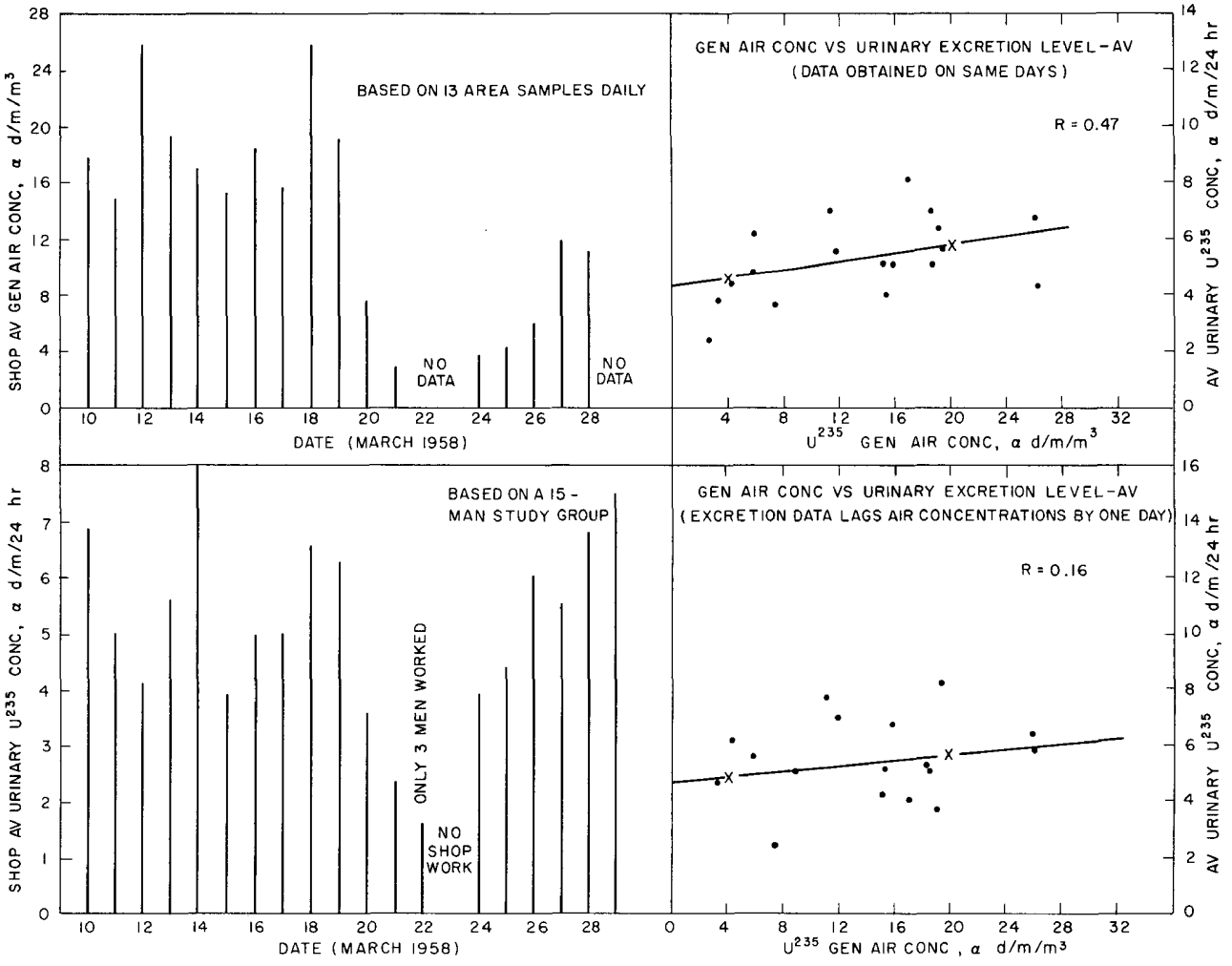

Figure 8. Comparison of average general air U^{235} concentration with average urinary U^{235} excretion in a uranium fuel fabrication shop.

interesting that the highest correlation exists when the urine results are compared to the air samples obtained the same day, i.e., with no lag. This may possibly be due to the fact that the particle size of the dust obtained in the general air samples (with no pre-impingement) was smaller than that obtained in the breathing zone air samples (with pre-impingement). Consequently, it is possible that the dust collected by general air samples was more representative of the particle size which undergoes rapid mobilization in the body than was the dust in the fractionated breathing zone samples. Unfortunately, no particle size information is available on the general air samples.

4. Figure 9 is a plot of individual average uranium intake versus individual average uranium excretion. The correlation coefficient calculated for this plot is comparable to those shown in Figures 7 and 8.

It is realized that conclusions drawn from such a study as this are limited by the difficulties encountered in obtaining complete histories of uranium distribution and retention in the lung and by assuming that short-term breathing zone sampling is indicative of total exposure throughout the day. In spite of these restrictions, the mass data indicate that there is a correlation between uranium intake and urinary uranium excretion which affords a rough basis of predicting intake from excretion data with respect to this study group. Certainly, one could not take a given urine result and calculate the exposure of that individual. However, grouping of results may offer some promise of success. It appears that certain trends are evident even at these low levels of exposure encountered.

Possible future work might involve the selection of a fraction of the air-borne uranium having a smaller particle size (possibly $<1\ \mu$) as a basis for comparison with excreted uranium. The choice of the 3-μ separation used in this study was based on density differences which indicated that a 3-μ ura-

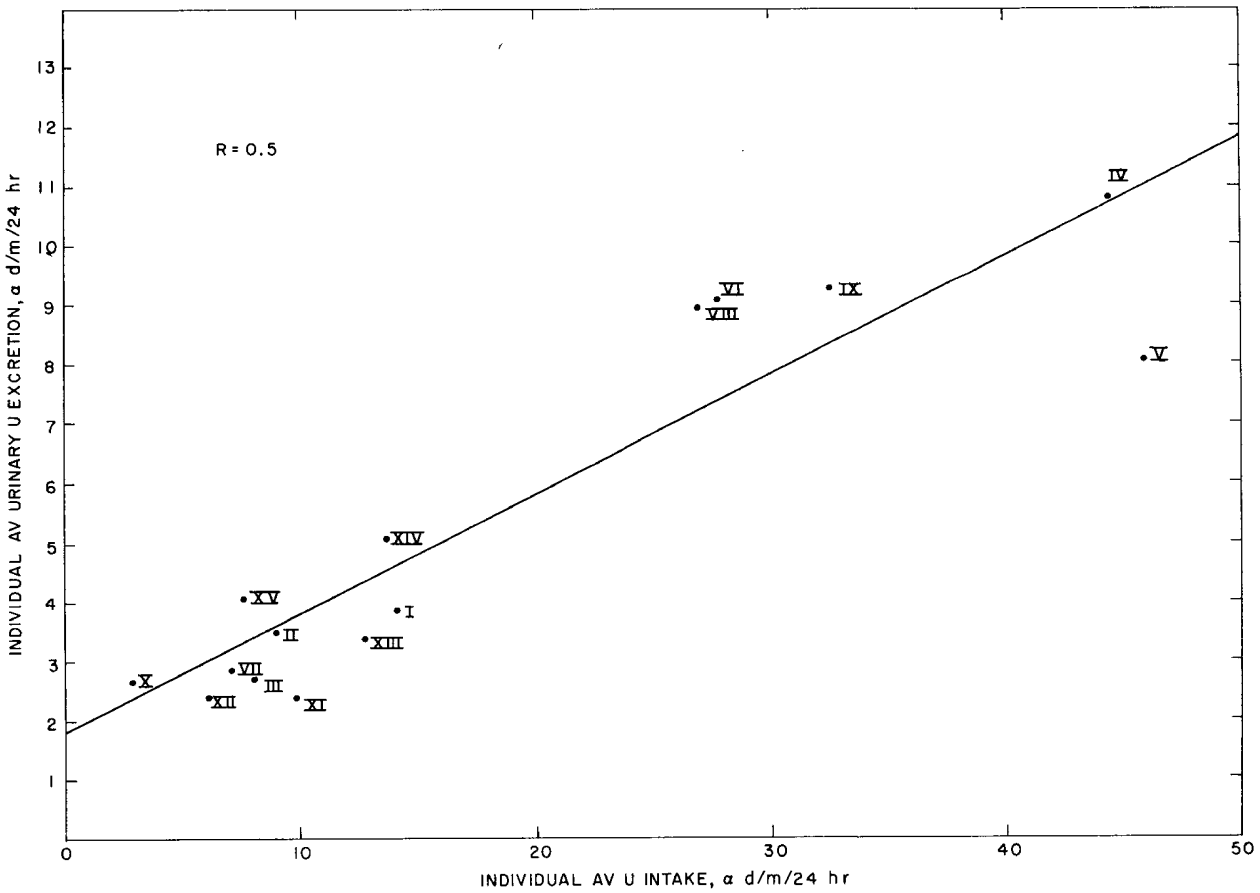

Figure 9. Individual average U^{235} intake versus urinary excretion in a U^{235} fuel alloy fabrication shop, March 1958.

nium particle would behave similarly to a 10-μ atmospheric dust particle in the lung. The differences in correlation with time lag as shown in Figures 7 and 8 may support such action, since the small particles, with a higher surface area to mass ratio, suspected in the general air samples are going into solution and into the blood more rapidly than the larger particles.

It is hoped that additional studies of this type may lead to more meaningful information concerning lung particulate influence on blood and urine concentrations.

Estimation of Body Burden and Internal Dose Based Upon Urinary Uranium

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INTRODUCTION

Whether a uranium exposure involves primarily radiotoxicity or chemical toxicity depends upon both the isotope and the mode of entry into the body, but in either case it is desirable to know the body burden and the internal distribution pattern. The problem of determining the detailed distribution of uranium in the organs is not an easy one, but, although no really satisfactory methods exist, several approaches might be used to obtain rough estimates of the organ burdens.

Measurements of loose surface contamination and of air-borne uranium provide estimates of potential exposure and may be very useful in establishing an upper limit for the body burden in an individual case. *In vivo* gamma counting offers a direct means of estimating gross body burden in many cases. Analyses of body tissues, fluids, and excreta can provide a measure of dose in the individual organs. Unfortunately, biopsy specimens of lung, kidney, or bone tissue can seldom be obtained in sufficient quantities to provide representative samples. There is some indication that the concentration of uranium in the teeth could be used to estimate total bone burden. Since the blood intercommunicates with most deposition sites, blood samples should provide a very useful measurement; however, the sensitivity of detecting uranium in blood must be greatly improved before this approach will be of much value. Fecal analysis alone has very limited usefulness, but may be used in conjunction with other methods to give a better estimate of a particular exposure.

DISTRIBUTION AND EXCRETION FOLLOWING INJECTION

Since urine represents the end point of many metabolic functions, urinalysis has long been used as an index of the proper operation of some internal processes. Considerable data have been accumulated on the relation of uranium excretion rate to body burden. Nine terminal brain tumor patients at the Massachusetts General Hospital in Boston[1] and six other human subjects at the University of Rochester[2] have received intravenous injections of uranyl nitrate.

Figure 1 shows the urinary uranium data reported by Bassett[2] from the Rochester study, in which single intravenous injections of uranyl nitrate were given to human subjects. From the lowest dose of 6.3 μg/kg body weight up to a dose of 42 μg/kg the patterns remain very similar. Patient VI received 70.9 μg/kg. The curves in Figure 2 represent the excretion results on eight terminal brain tumor patients at the Massachusetts General Hospital, the first six of whom were injected intravenously with uranyl nitrate and the last two with uranium tetrachloride. Comparison of the initial excretion rates of the uranyl nitrate subjects shows that, as the gravimetric dose increased, the initial elimination rate decreased. Boston patient VI, who received the largest injected dose (907 μg/kg), showed the greatest deviation in the excretion pattern. In connection with the Boston study, tissue specimens obtained at autopsy were analyzed for uranium content (see Table 1). Although detectable amounts of uranium are deposited in most of the tissues, the blood-bone-kidney system contains about 70% of the total body burden at any particular time.

One of the many possible models that might be used to approximate the time-dependent distribution of uranyl ions in the body is illustrated in Figure 3. It should be emphasized that this particular model is used merely as an approximation to the much more complex true situation. For example, the bone data from the Boston study indicated two elimination rates (half-times of 18 days and 300 days) rather than one as shown here. Nevertheless, this relatively simple model provides a good description for much of the single injection data. In

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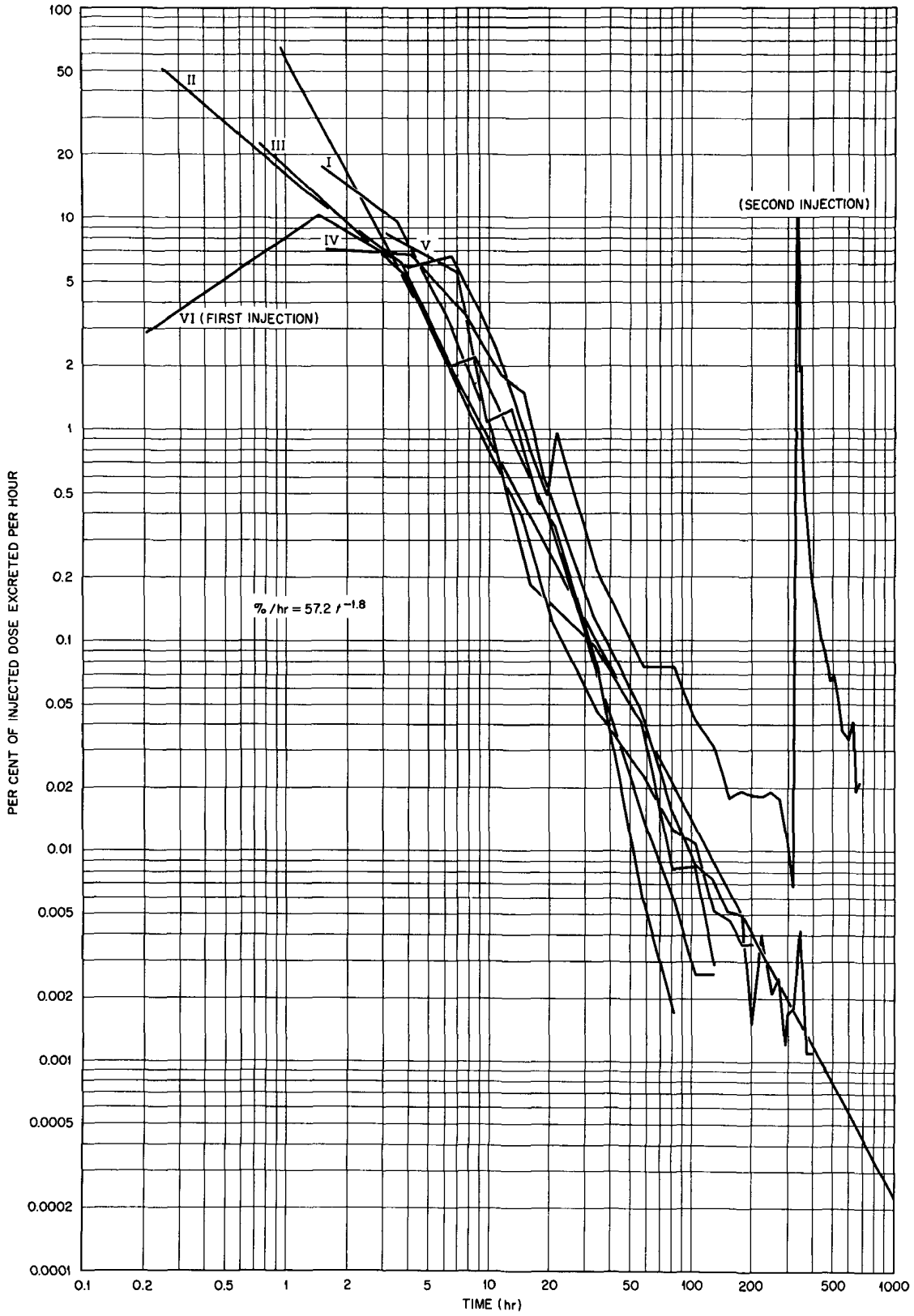

Figure 1. Rate of urinary excretion of six patients following intravenous injection; University of Rochester.[2]

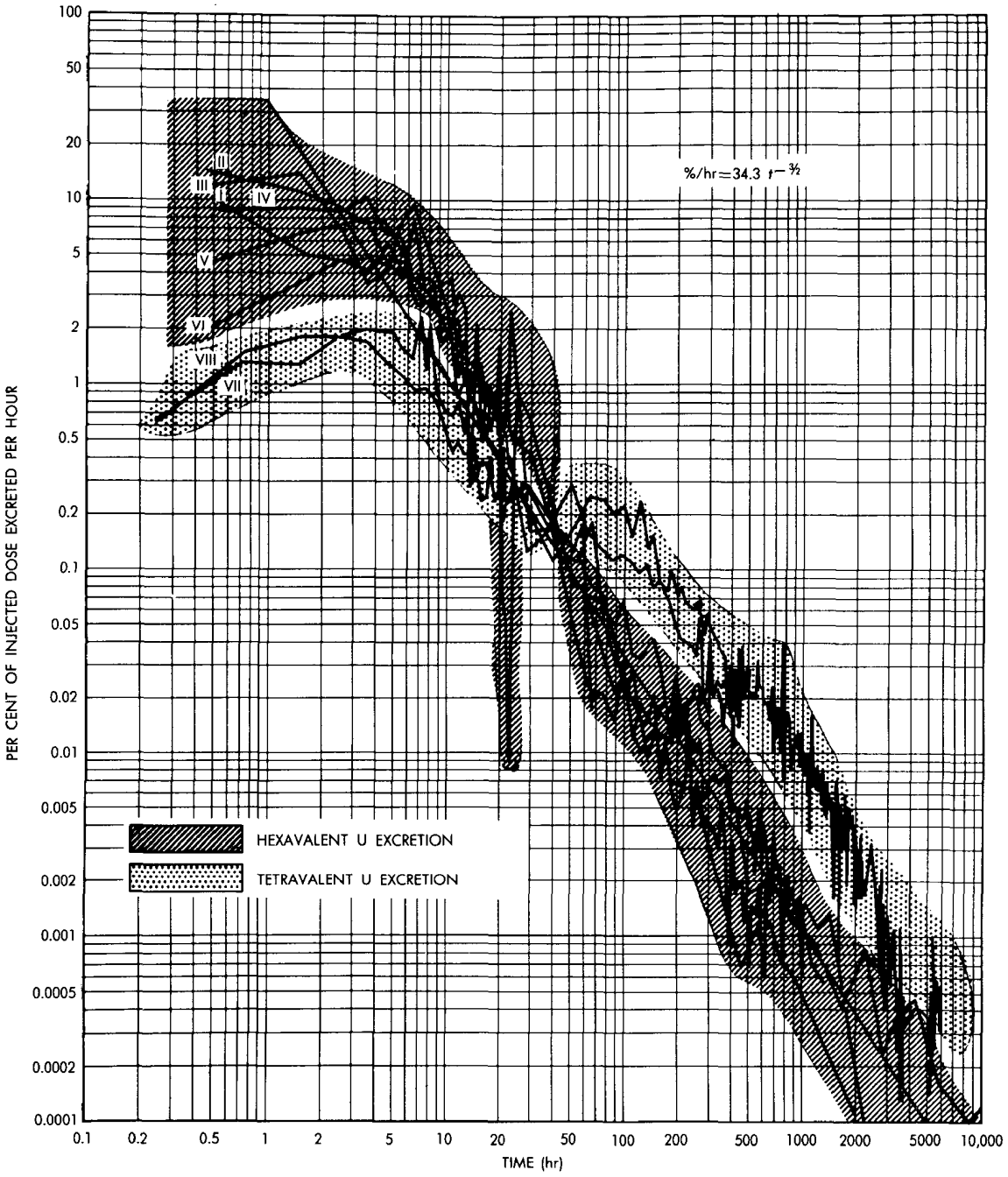

Figure 2. Rate of urinary excretion of eight terminal brain tumor patients following intravenous injection; Massachusetts General Hospital.[1]

order to follow the excretion data from the Boston study, it is necessary to modify the basic model to account for an apparent dose dependence. Since larger doses resulted in a decreased initial percent excretion, it is suggested that a saturable excretion pathway in the simple model might be sufficient to approximate the data.

Electronic components of an analog computer may be used to simulate the exchange of uranyl ion between the compartments of the model. Common mathematical operations such as integration, multiplication by a constant, sign changing, and addition, which may be performed with the computer, are represented symbolically in Figure 3.

Table 1

Percent of Injected Dose Per Standard Man Organ or Tissue for Five Terminal Brain Tumor Patients After Injection of $UO_2(NO_3)_2 \cdot 6H_2O$

Organ or tissue	Grams	Expiration time, days				
		2.5	18	74	139	566
Bone	7,000	10.0	4.9	1.4	0.6	1.3
Kidney	300	16.6	7.2	0.7	1.2	0.4
Muscle	30,000	1.2	2.1	0.9	0.3	0.06
Skin and subcutaneous tissues	6,100	1.8	1.0	0.1	0.06	—
Fat	10,000	0.6	0.6	—	—	0.04
Red marrow	1,500	—	—	0.02	0.03	0.1
Yellow marrow	1,500	—	—	—	—	—
Blood	5,400	1.0	0.2	0.005	0.002	0.004
Lower large intestine	150	—	—	—	—	—
Stomach	250	0.88	0.02	0.003	0.001	0.001
Small intestine	1,100	0.2	0.2	0.03	0.01	0.006
Upper large intestine	135	—	—	—	—	—
Liver	1,700	1.8	1.1	0.2	0.2	0.05
Brain	1,500	—	—	—	—	—
Lungs	1,000	0.5	0.4	0.03	0.02	0.008
Lymphoid tissue	700	—	—	—	—	—
Heart	300	0.06	0.02	0.003	0.006	0.002
Spleen	300	0.6	0.2	0.1	0.02	0.006
Urinary bladder	150	0.03	—	0.002	0.001	0.0003
Pancreas	70	0.7	0.008	0.008	0.0006	0.0004
Salivary glands	50	—	—	—	—	—
Testes	40	—	0.01	0.008	0.002	0.002
Spinal cord	30	—	—	—	—	—
Eyes	30	—	—	—	—	—
Thyroid gland	20	—	0.003	0.0002	0.0001	0.0002
Teeth	20	—	—	—	—	—
Prostate gland	20	—	0.003	0.0004	0.0004	0.0001
Adrenal gland	20	0.02	0.01	0.003	0.001	0.0004
Thymus	10	—	—	—	—	—
Misc. tissues (blood vessels, cartilage, nerves, etc.)	390	0.3	0.2	0.04	0.002	0.002
Urine (% of dose accumulated)		61	63	92	84	98.2
Body content = 100 − % in urine		39	37	8	16	1.8
Total in tissues		35	18	4	3	2

The system of differential equations relating voltage at various points of the circuit to time is the same as the set of equations describing the time patterns of uranium content in the different parts of the model. Saturation may be brought about on the simulator by comparing the rate of elimination with a fixed maximum. If the transport rate exceeds the maximum, the excess is held up in a long-term storage portion of the "kidney" compartment.

Figure 4 illustrates the results of applying the saturable model to the excretion data of Boston patient VI, the one receiving the largest dose and whose data showed the greatest departure from the normal trend. It is significant that the biological parameters used on the computer setup were not derived from this patient's data, but were the average values computed from the Rochester patients' excretion rate curves.

One of the major goals of the model work is represented by the diagram in Figure 5. The inverse model is an attempt to operate upon urine data to obtain a dynamic estimate of body burdens in the several compartments. In principle, the inverse

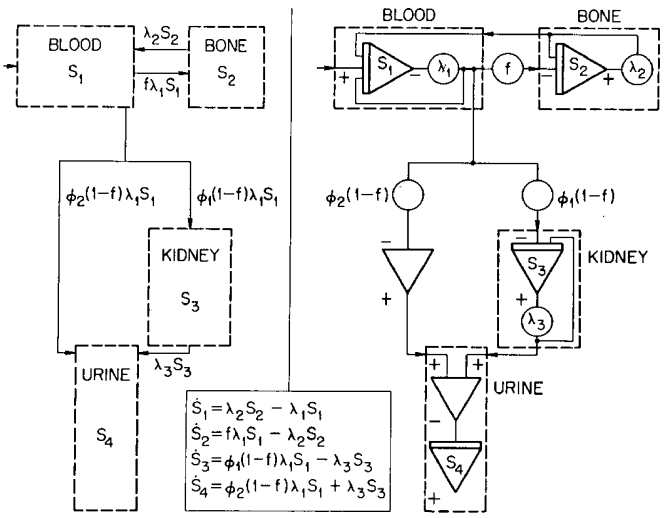

Figure 3. A possible model to approximate time-dependent distribution of uranyl ions in the body.

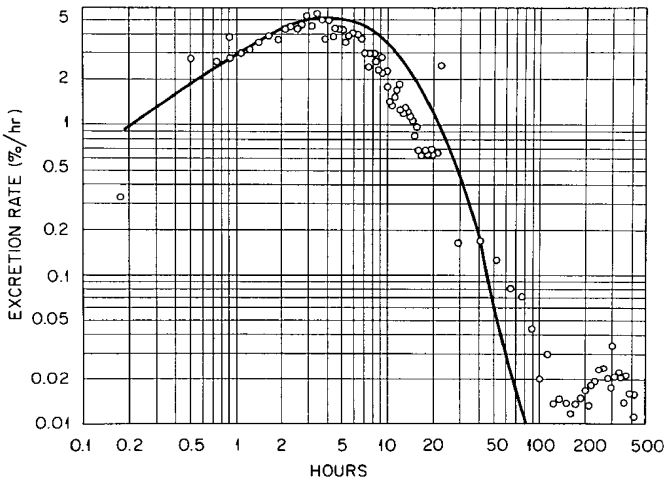

Figure 4. Application of the saturable model to the excretion data from Boston patient VI.

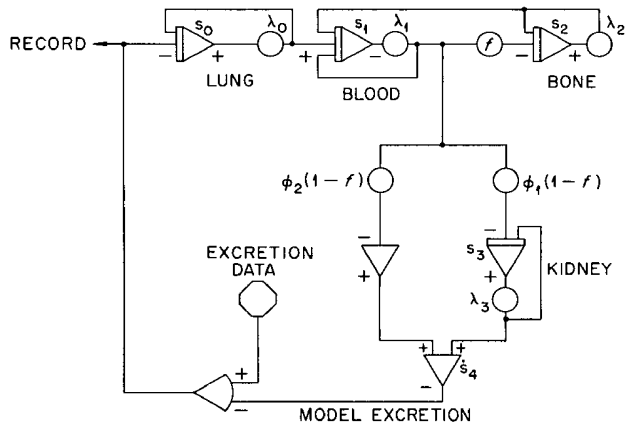

Figure 5. Inverse model.

model may be made as sophisticated as necessary; for example, the model may include two bone compartments and additional compartments for liver and other tissues. However, a great deal of study remains to be done before such an approach will become a tool useful to the individual engaged in health protection field work. Until adequate biological data are available, the inverse model will remain a worthwhile goal and an aid to the research worker.

BODY BURDEN OF A SOLUBLE COMPOUND

In the meantime the approach illustrated by Figure 7 may prove to be useful in estimating body burden in certain types of exposure. The curves were plotted from excretion data from the six Rochester patients and six of the Boston patients. The body burden was obtained by subtracting the integrated excretion from the known injection dose (Figure 6). Body burden at a particular time was combined with the excretion rate at the same time to obtain the points for Figure 7.

This set of curves may be used to estimate the body burden resulting from an exposure similar to an injection. In addition to the more obvious application to exposure involving wound contamination, these curves may be useful in estimating the body burden resulting from acute inhalation of soluble compounds such as uranyl nitrate or uranyl fluoride. After the first two days the excretion curve for such an inhalation becomes very similar to that for an injection, and these curves should yield reasonable estimates, but before that time the body burden might be seriously underestimated by this method. The family of curves has been fitted with the arbitrary function, $q/\dot{\mu}=320\,e^{-280\dot{\mu}}+3\,t^{1.3}$, for $\dot{\mu}$ in milligrams per day and t in days. Curves shown on the graph as solid lines were calculated for the given times by using this function. In practice, one would obtain a urine specimen at some time T after exposure (greater than two days for inhalation of soluble compounds). The time since the last previous void would be noted at ΔT, and the excretion rate $\dot{\mu}$ would be expressed in units of milligrams per day. At early times when the excretion rate is rapidly changing it is important to use the midpoint time, $t=T-\frac{1}{2}\,\Delta T$, rather than the end-point time T. By using the determined values of t and $\dot{\mu}$, a multiplication factor may be obtained from Figure 7 or by substitution in the empirical equation. Body

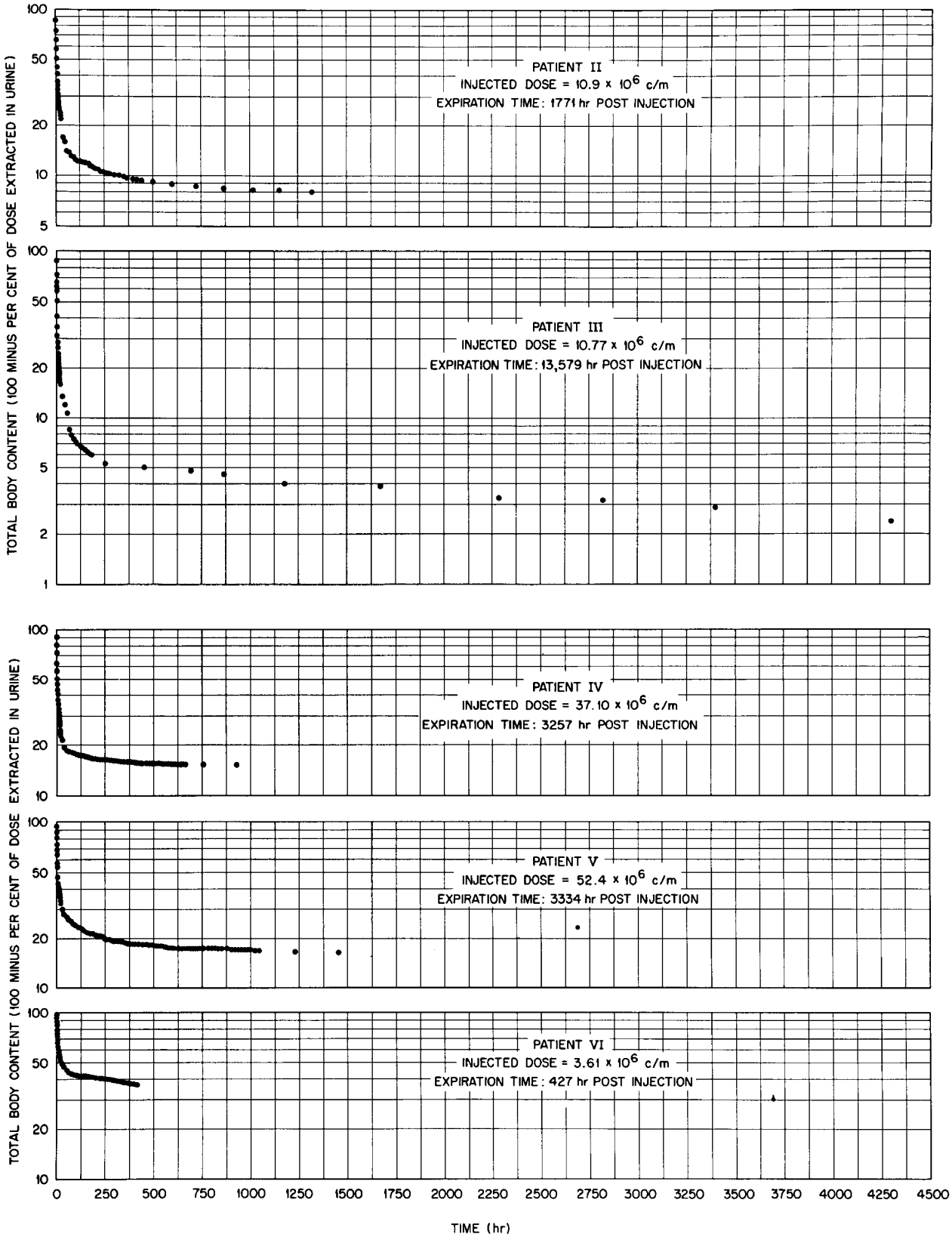
PATIENT II
INJECTED DOSE = 10.9 x 10^6 c/m
EXPIRATION TIME: 1771 hr POST INJECTION
PATIENT III
INJECTED DOSE = 10.77 x 10^6 c/m
EXPIRATION TIME: 13,579 hr POST INJECTION
PATIENT IV
INJECTED DOSE = 37.10 x 10^6 c/m
EXPIRATION TIME: 3257 hr POST INJECTION
PATIENT V
INJECTED DOSE = 52.4 x 10^6 c/m
EXPIRATION TIME: 3334 hr POST INJECTION
PATIENT VI
INJECTED DOSE = 3.61 x 10^6 c/m
EXPIRATION TIME: 427 hr POST INJECTION
TOTAL BODY CONTENT (100 MINUS PER CENT OF DOSE EXTRACTED IN URINE)
TIME (hr)

Figure 6.

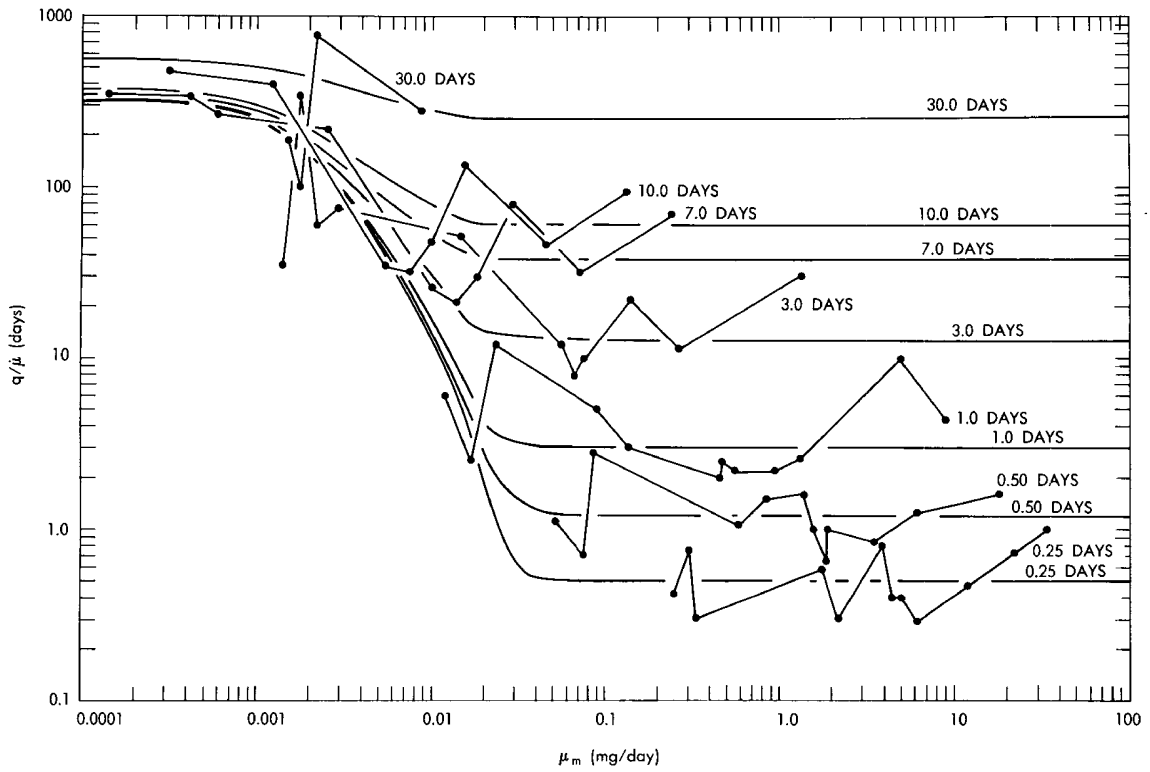

Figure 7. Body burden versus excretion rate at a fixed time following intravenous injection of uranyl nitrate.

burden at time t may then be estimated as the product of this factor and the excretion rate.

AN ACCIDENTAL EXPOSURE TO U_3O_8

Some of the estimation procedures that were applied to data obtained from an accidental inhalation of U_3O_8 are outlined in Table 2. Throughout this paper "lung burden" is arbitrarily taken to mean that portion of the material deposited in the lung which may be transferred to the blood-bone-kidney system and eventually excreted in the urine. Initial lung burden was estimated by several methods. Urine specimens indicated an excretion of 0.5 mg/day at the time the employee was removed from exposure. By assuming that λ_L, the initial transfer fraction from lung to blood, was in the range 0.006 to 0.06 per day (equivalent half-times range from 12 to 120 days) and that 2/3 of the amount reaching the bloodstream during the first day would appear in the urine, the lung burden was expected to be in the range 1.3 to 13 mg.

A 6-ml blood sample was chemically ashed, plated, and counted in an alpha scintillation counter. This determination indicated a uranium concentration of 1 mμg/ml, which would represent about 6 μg in the total blood. Another sample taken the next day indicated about the same concentration; thus, if total blood content is denoted by S_1, then $dS_1/dt \approx 0$. The individual's history indicated no previous significant exposure, hence it was assumed that the amount reaching the blood from tissues other than lung was negligible, and therefore $dS_1/dt \approx \lambda_L S_L - \lambda_1 S_1$. Lung burden, S_L, was estimated then from $S_L \approx S_1\lambda_1/\lambda_L$. A value of 9.9 has been obtained for λ_1 from the human injection studies, and the range on λ_L was used as above. This approach gave an estimate of 1 to 10 mg in the lung.

A mock-up of the operation was run, and air samples were taken in an attempt to characterize the exposure with respect to chemical compound, particle size distribution, and air activity levels. Electron diffraction analysis confirmed the initial assumption that the compound was U_3O_8. The mass median particle size was found to be 1.9 μ by use of a cascade impactor, and the geometeric standard deviation was 1.8. Air activity levels varied greatly with location and with operating tech-

Table 2

Method	Estimated lung burden, mg
Urinalysis (soon after exposure):	
Fraction per day from lung to blood × Lung burden ×2/3= Excretion rate	
$\lambda_L S_L \times 2/3 = dU/dt \rightarrow 0.006\, S_L \times 2/3 = 0.05$ mg/day	
$S_L = 13$ mg (or 1.3 mg if $\lambda_L = 0.06$)	1.3 to 13
Blood sample:	
During first post exposure day $dS_1/dt \approx 0 \approx \lambda_L S_L - \lambda_1 S_1$	
$S_L \approx \lambda_1 S_1/\lambda_L = 9.9 \times 0.006$ mg/0.006=9.9	
$S_L \approx 10$ mg (or 1 mg if $\lambda_L = 0.06$)	1 to 10
Air sampling:	
Maximum air concentration =0.3 mg/m³	
Alveolar retention =20%	
Breathing rate =10 m³/shift	
Duration of exposure =11.5 shifts	
Amount remaining of total dose =90%	
Potential lung burden $=0.3 \times 0.2 \times 10 \times 11.5 \times 0.9 = 6.2$ mg	6.2
In vivo counter:	3
Urinalysis (after 100 days):	
$dU/dt = 0.0433\, e^{-(0.693/20)t} + 0.0067\, e^{-(0.693/300)t}$ mg/day	4.1

niques, but it appeared possible for an individual to be exposed to as much as 0.3 mg/m³ averaged over his daily shift. Assuming a breathing rate of 10 m³ per shift and an alveolar retention of 20%, it would have been possible to accumulate as much as 6 mg in the alveoli over the known period of exposure. In this connection it should be mentioned that two other men were performing the same operation that produced this exposure. At the same time this individual was excreting 12,000 units per day, one of his co-workers was excreting 1,100 units per day, and the third man excreted less than 10 units per day.

Subsequent estimates of total lung burden made by Marinelli and Miller[3] using their *in vivo* counter were extrapolated back to the time the exposure ceased and indicated the initial value was 3.0 mg.

The data describing the excretion rate for the first 100 days post exposure may be fitted with the expression $0.0433\, e^{-0.0347\, t} + 0.0067\, e^{-0.00231\, t}$ (mg/day). To test the validity of extrapolating this expression, it was integrated and then evaluated for $t = 140$ days. This function predicted a total excretion of 2.04 mg over a period of 140 days, while the total accumulated urinary uranium determined by the bio-assay laboratory was 2.00 mg for the same period of time. Since the urinary excretion rate can be expected either to follow this curve or to lie above it sometime in the future, the integrated excretion as t becomes large should approach the total initial dose but not exceed it. The integrated dose for t approaching infinity is 4.1 mg and should represent a close lower limit to the initial dose.

The curve shown in Figure 8 was obtained by assuming that the initial dose was 4.1 mg and adjusting the lung burdens measured by the *in vivo* counter accordingly. Ratios of estimated lung burden to excretion rate are shown as an arbitrary function of time. In a case similar to this one it might be possible to obtain a rapid estimate of lung burden by using this curve. However, since this represents only one set of data, it is not offered as a usable framework for estimating lung dose, but rather to illustrate what might be done if more data such as these were made available.

SOLUBILITY IN LUNG FLUIDS

An average ratio of estimated lung burden to excretion rate for two uranyl nitrate inhalation incidents is also shown on Figure 8. The initial lung burdens were estimated both by comparison with the uranyl nitrate injection data and by numeri-

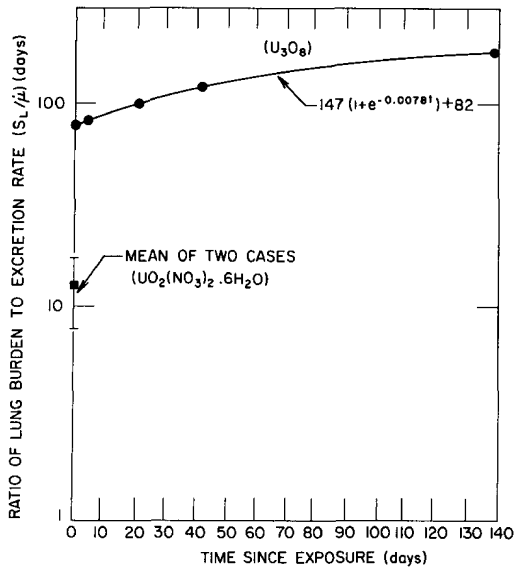

Figure 8. Ratio of lung burden to excretion rate following inhalation of uranium compounds.

cally integrating the measured urinary uranium over a period of three months. Because of gaps in the data it is very probable that the estimates for these two exposures are in error by as much as a factor of 2.

A very interesting relationship exists between the estimated initial ratio for the U_3O_8 inhalation and the average initial ratio for the uranyl nitrate exposures. The ratio of 82 to 13 is 6.3, which number might be interpreted to indicate that the rate of solubility of uranyl nitrate in lung fluids is about six times that of U_3O_8. Referring to work reported by Dounce[4] it may be seen that, using blood serum as an approximation to lung fluid, uranyl nitrate was dissolved about 5.8 times more rapidly than U_3O_8.

There are several pathways by which an inhaled uranium aerosol may reach the bloodstream and subsequently be detected in the urine. Particulates deposited in the pulmonary tract may be removed by ciliary action, phagocytosis, or dissolution in lung fluid. The action of the phagocytes is to transfer material either to the lymphatic system, where it may be deposited in a pulmonary lymph node, or to a ciliated area of the respiratory tree. It is possible that materials deposited in the lymph nodes may be slowly removed to the bloodstream. Furthermore, the particles removed by ciliary action may be swallowed, and a small fraction of the total amount may dissolve in the gastro-intestinal system, where it may then be transferred to the bloodstream. But, of all these mechanisms, the one that might be expected to lead to excretion in the urine immediately following an acute inhalation is removal by solution in lung fluids.

Considering the uncertainties involved in arriving at the various numbers used to show the apparent agreement in relative solubilities, it must be conceded that this agreement may, indeed, be fortuitous. On the other hand, if such a relationship exists, then it should prove to be very helpful in correlating single dose animal studies with human data from acute inhalation accidents.

CHRONIC EXPOSURE

A model for the distribution and excretion of the uranyl ion applicable to chronic exposures would not necessarily be the same as the model used to approximate the single dose data. A deposition site that receives only a small fraction of a single dose might be safely ignored in an acute exposure. But, if the elimination rate of that site is significantly slower than the rates of the other sites, then that compartment could take on primary significance in a prolonged exposure. A good example of this has been seen in Maynard's work[5] with the chronic inhalation of UO_2 in rats, dogs, and monkeys as compared to the work with the acute inhalation of U_3O_8 by dogs now under way at Oak Ridge National Laboratory. In the acute study the tracheobronchial lymph nodes were found to contain very small amounts of uranium, whereas the chronic study indicated that with time these lymph nodes accumulate uranium to reach concentrations much higher than that in general lung tissue.

At present there are not enough chronic data upon which to base a comprehensive model. Yet there is some indication that the basic model already described need not be radically changed. The biological half-time of uranyl ion in the kidney has been interpreted to be 12 days from the single injection study, and was estimated to be 10 days from a chronic exposure case reported by Butterworth.[6] Retention in the bone was seen in the Boston study to fall off according to two rates; initially an 18-day half-time dominated the elimination and then over a period of a few weeks the rate decreased until an approximately 300-day half-time became the major component. In the case of a model designed to represent chronic exposure, the 300-day component would be the more

significant. This value may be compared with the approximately 450-day component reported by Eisenbud[7] from the urinary uranium excretion pattern obtained following cessation of a chronic exposure.

SUMMARY

Urinary uranium excretion data from 12 humans receiving intravenous injections of hexavalent uranium have provided a pattern relating body burden to excretion rate. This relationship should be useful in inhalation exposures involving soluble compounds in addition to cases of wound contamination.

A U_3O_8 inhalation case has been studied by several methods: air sampling, blood sampling, *in vivo* gamma counting, and urine analysis. All these procedures yield tenable estimates of the exposure dose, but urinalysis and *in vivo* counting provide what are believed to be the best estimates.

A suggested framework for rapidly estimating acute inhalation exposure has been based upon two cases of accidental inhalation of uranyl nitrate and one U_3O_8 inhalation. It is evident that a great many more data are needed before such an approach as this can be made useful.

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Urinary Uranium as an Indicator of Dose to Exposed Personnel

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In previous papers the plant practice has been discussed of recording the integrated dose or relative hazard to individual workers calculated from measurements of urinary uranium. The purpose of this paper is to comment on the degree of reliance that can be placed on calculations of this sort.

Many pertinent factors might be considered, including accuracy and time of sampling, the validity of spot sampling, the adequacy of sampling over known time intervals <24 hr, and the accuracy and reproducibility of sample measurements. Instead, two other factors will be discussed, partly because there is a tendency to regard them as fixed and well-established. These two sources of uncertainty are, first, the biological factors involved in the estimation of the maximum permissible levels in the urine and, second, the individual biological variability from the average.

In Table 1 are listed suggested maximum permissible levels for the urine when the hazard is insoluble air-borne natural uranium. Three calculations are included, with somewhat different biological assumptions. In all three the assumption is used that half the insoluble uranium retained in the lung goes somehow into the circulating blood as the soluble form. In methods A and B it is assumed that a state of transient equilibrium exists, and that all excretion of systemic uranium is via the urine; whereas in method C it is assumed that about one third of the solubilized uranium is either excreted as feces or deposited. The half-times listed vary from 100 to 365 days. Each of the three sets of assumptions can be defended on the basis of data from animal experiments. Even supposing that a single set of biological values could be selected on its merits, there is still the problem of extrapolating to man. Some evidence has been presented in previous papers to the effect that animal experimental data lead to an overestimate of the hazard to man.

For the case of air-borne soluble uranium, a method of calculating the maximum permissible urinary level is shown in Table 2 which avoids specification of biological parameters. The only requirements are that the exposure be a reasonably constant one, that the time since the beginning of exposure be a month or more, and that the urine measurement be made during the work week (i.e., during exposure) and not after the week-end absence from work. It is based on the finding with animals that an exposure to 50 μg natural uranium per cubic meter air (1.7×10^{-11} μC/cc) is safe, and on the acceptance of damage to the kidney as the most sensitive mode of potential injury. This calculation gives no information about uranium body burden and in fact is made with the assumption that the skeletal uranium is in equilibrium at a concentration <25 μg/g (<300 mrem/week).

An alternative calculation for soluble air-borne uranium based on defensible values from animal data for body burden and effective half-time yields an estimate of maximum permissible urinary activity about one-fifth that given by the calculation shown in Table 2.

In view of the uncertainties indicated above and the other sources of error listed but not discussed, it would appear improbable that routine urine sampling could succeed in predicting absolute body burden or dose within a factor of two or three. The final establishment of the necessary biological information on man must come from health physics field research.

It may well be argued that relative rather than absolute numbers are wanted; that if a conservative urinary uranium level is chosen as the operational limit, individual body burdens can be evaluated in terms of this limit.

This brings us to a consideration of biological variability as a factor contributing to the uncertainty of relative evaluations. For an example I have made a simple analysis of the excretion data on six hospital patients at the University of Rochester injected with single doses of soluble uranium. It turns out, as pointed out in the preceding paper, that the best representation of the

Table 1

Calculations of Suggested Urinary MPL for Insoluble Uranium

	Method		
	A	B	C*
Effective half-time in lung, days	120	365	100
Fraction of U retained in lung which goes into systemic circulation	0.5	0.5	0.5
Fraction of systemic U excreted in urine	1.0	1.0	0.67
MPL in urine, d/m/24-hr specimen	107	35	86
d/m/l urine	76	25	61

In all methods of calculation it is assumed that the lung (weight=1000 g) is the critical organ; that the hazard is radiological; and that a dose to the lung of 0.3 rem/week is associated with a lung burden of 25 μg natural uranium per gram lung ($=8.3\times10^{-6}$ μC/g). By definition 1 μC in this context is equal to 1 μC U^{238}+1 μC U^{234}+0.024 μC U^{235}. The permissible urine activity level includes the sum of the α disintegrations from U^{238}, U^{234}, and U^{235}.

The formula used for calculation of μC in a 24-hr specimen is

$$\frac{\text{Lung conc.} \times \text{Weight of lung} \times 0.693 \times \text{Absorption factor} \times \text{Excretion factor}}{\text{Effective half-time in lung}}.$$

To convert to disintegrations per minute per 24-hr specimen, multiply by 4.48×10^{6}.

*In calculation C the factors used are those of Neuman, *Ind. Med. and Surg.* **19**, 185 (1950).

Table 2

Calculation of Suggested Urinary MPL for Soluble Uranium

Air exchange in 8-hr working day=10^7 cc.

$MPC_{air}=1.7\times10^{-11}$ μC/cc ($=50$ μg/m^3).

Percent of the inspired soluble U retained by the body=60%.

Therefore $1.7\times10^{-11}\times10^{7}\times0.6=1.02\times10^{-4}$ μC/24-hr urine specimen
$=450$ d/m/24-hr urine specimen
$=320$ d/m/l urine specimen.

It is assumed that the chemical toxicity to the kidney is the hazard of soluble uranium; and that the worker rapidly comes into equilibrium with his environment, particularly with reference to the kidney burden, i.e., that effective half-time in the kidney is of the order of 15 days. If the latter is true, the fractional amount of inspired uranium retained per day equals the amount excreted. Excretion by the fecal route is negligible.

It is to be strictly noted that, since soluble uranium is rapidly excreted, this calculation refers only to a *midweek* sample.

excretion rate as a function of time is arrived at by plotting the data on a log-log plot. When all the data from the six patients are plotted and the best single line is drawn, a general equation is obtained: $E_t=200\ t^{-2.13}$, where E_t is the percent of the intravenous dose excreted per hour at time t expressed in hours.

In Table 3 are listed for each patient the true dose known to have been injected; the average dose estimated by the above equation from analytical data on several 24-hr urine collections; the number of 24-hr urine samples at different times t used in the average; and the range of the individual estimates. It may be seen that the estimate

Table 3

An Estimate of Individual Biological Variation

Patient No.	True dose, μg	Average estimated dose, μg	No. of 24-hr urine samples	Range of single estimates, μg
1	385	310	3	185 to 431
2	476	223	3	170 to 292
3	584	482	6	275 to 657
4	1,918	2,143	7	1,690 to 2,920
5	2,746	3,047	3	1,842 to 4,560
6	3,910	14,286	6	7,840 to 19,400

is improved by increasing the number of samples; but even the average estimate is, in one case, off by a factor of 3 to 4. This is, as I have said, a simple example. The urine samples covered 24 hr and were collected with great care in a hospital metabolism ward by trained nurses. The uranium analyses were performed in the laboratory. The patients were screened so that those included in the experiment had normal kidney function. I therefore believe that the variations from the average are acceptable examples of individual differences. It is true that the sample is exceedingly small. This has the effect of making the general equation less reliable, but it also reduces the range of individual variation likely to be encountered.

It is therefore concluded that, with the present state of knowledge, the routine measurement of urinary uranium does not serve to specify the individual body burden or permit calculation of the body dose with a high degree of confidence. For accidents studied by laboratory procedures and especially in cases where other methods such as whole-body counting may be employed, the result may be more exact.

This emphatically does not imply that urinary uranium measurements are without value in a health protection program. Such measurement is perhaps the only good method of testing air contamination with respect to the individual exposure and the individual job. This may certainly be true for soluble uranium. For the evaluation of exposure to insoluble uranium, fecal sampling would be very much more appropriate and should be more generally used. The usefulness of excreta analyses in evaluating exposure and improving plant conditions has been amply demonstrated by the detailed examples presented earlier in this symposium and need not be further discussed here.

In conclusion, the importance should be emphasized of the human data which has been and is being collected in the field. It is apparent that there is a great need for more such data: for detailed measurements on accident cases; for job-rating exposure values arrived at by air sampling and by urinary and fecal measurements; for more autopsy data on workers whose exposure can be estimated with reasonable accuracy; and for medical vigilance in picking up late effects if they appear. In view of the circumstance that the exposure of workers to uranium in the early years was much greater than it is likely to be in the future, it is very important that these people be kept track of in a central file, that they receive regular medical examinations, and that every effort be made to obtain autopsy samples at their death. For the general elucidation of our remaining problems with uranium this human material is priceless and cannot be substituted for by experiments in the laboratory.

General Comments on Urinary Uranium Analysis

W.F. Neuman
University of Rochester, Atomic Energy Project, Rochester, New York

I feel that the Rochester data have been somewhat overused and perhaps misused from time to time. Nevertheless, I have gotten several distinct impressions from the papers I have heard. I would like to read the introduction of an article that I wrote eight years ago:

"The production of uranium has grown to be a large-scale industrial process. [That is still true.] Since uranium is both toxic and radioactive, particularly in isotopic preparations, the problem of determining the exposures is one of first order of importance. [That is still true.] Because of its specificity, analysis for urinary uranium is one of the most attractive methods of assessing exposure. [That is still true.] Urine specimens can be obtained simply and without the need of specialized personnel in attendance and without any inconveniencing or alarming the worker. [That is still true.] It may be determined accurately by a number of accurate methods." [I wonder about that.]

I would like to mention a maxim I learned in elementary mathematics; that is, to solve for two unknowns, at least two equations are needed, and preferably three; when there are five or ten or more variables, obviously they cannot all be covered by a single measurement. What we are stressing here, collectively and individually, is that a single measure – the analysis of urine for uranium – does not tell us the whole story. I don't think we should be very surprised about this. It is a poor measure of any kind of exposure – of air concentrations, of lung level, bone level, or any level. But it is one of the most convenient and one of the best we have; therefore, its shortcomings have been dwelt on.

I think we should refocus our attention on the fact that it is important and one of the most easily obtained and best methods we can get.

I also think we should focus our attention on the matter of more careful examination of methodology. I have in my files a report of a single case in which three different methods of analysis are used, with three different sets of data. I don't mean to emphasize or urge uniformity because I think a uniformly poor method is not to be desired. There might be some real advantage in having everybody try his own hand at it, but I do think it is worthy of a really intensive examination by scientific study. The people in this field never seem to realize how many thousands of urinanalyses are going to have to be made in the next ten years.

I also sense a certain disappointment that, based on the animal studies, largely done at Rochester, nobody seems to be getting hurt. This bothers me. I think we should be proud and happy that we aren't finding serious illness in the workers or administrators. It may be that we could be a little more optimistic. The fact is important, I think, that the permissible levels obtained from animal studies really weren't too bad, because they have proved to be reasonably operational and in fact safe, as far as we know.

We are using the nephrotoxic effect of uranium as our guide, and on this score we are fairly sure we are right; that is, the levels set are not causing any kidney damage. But there are many shortcomings to using animal data, and I think the surprising results in the dog presented by Mr. Downs illustrate the areas in which we cannot permit guessing. If you are finding accumulation of uranium in the experiments being done on the kidney, then it seems possible that there are long-term chronic effects from uranium which have not been uncovered by the animal studies so far. If you are somewhat surprised at not finding illnesses in people, they may yet appear to a much greater extent than we expect, but we are thankful that this has not happened so far.

I think it is perfectly obvious to all who have attended these meetings that there are too many occasions when the terminology used results in oversimplification. If you talk about soluble and insoluble, you have an oversimplification to start with. There are still too many variables involved; we are only beginning to find out how many. For example, in the matter of lung deposition, clear-

ance from the lung, and absorption from the lung, there are many gradients involved and many variables.

We know that as an element, uranium is beautiful. It has one of the simplest distribution patterns and one of the simplest metabolisms we have ever encountered, and its metabolism is one of the best documented. But, in spite of its simplicity, it is still quite complex. There is no single half-time for the deposition of uranium in bone; there are many. I think if you examine the whole spectrum, you will find that the half-time of uranium in the skeleton is a direct function of length of time of observation, and if you study the relatively short-term half-time, you find that it is very difficult to determine. This is because it is increasing with passage of time. It just isn't a simple log-decay relationship.

As to the matter of absorption, the generally accepted absorption figures have been questioned. Of course, they should be questioned. There is no such thing as a fixed percentage absorption rate. It depends on the level and kind of compound in question; some will be absorbed relatively well, others poorly; we still don't know why. We try to correlate the information we have and find that this is very complex.

Discussion of Session III Papers

Chairman, W.B. HARRIS

WATSON: Might not particle size data be of more interest than further urinalysis data? From some of the comments it seems that this might be the case.

NEUMAN: I think we are worrying more than we need to about whether we are justified in spending so much money on a urinalysis program. If you find that urinalysis has a positive value, then it should be continued. I agree that we should have further particle size data, but not that we should have particle size data instead of further urinalysis. The whole area needs to be covered.

I would like to suggest that as a result of this meeting a committee should meet to discuss this problem and decide how much emphasis should be put on dust sampling, urinalysis, and so on, to serve as a guide. I think this committee might set up a subcommittee on urinalysis to see whether or not all the programs being conducted are giving good results.

HARRIS: I take exception to Dr. Neuman's statement that it has been said that because we haven't created any injury, we are therefore in very good shape. The uranium industry is relatively new, and it has grown tremendously in the last five or six years. To lower the uranium exposure of people in this industry by a factor of 10 would probably cost something on the order of five million dollars. It is very important to understand this, and to decide whether we should raise our sights. Is it necessary to spend this five million (or perhaps even ten million) to reduce the levels from the point where they are now? I would guess that there have been exposures of the order of ten times the MPC in some cases; the question is whether this should be a cause for concern or whether we should just forget about it. Frankly, I don't know the answer to that.

It may be that when we have good information on humans (and perhaps we do have it, and the data need only be brought together and examined), we may find it possible even to raise the MPC. It is true that the early information on toxicity is very good. It has put us in a position to say now that in processing some hundreds of thousands of tons of uranium we have had no ill effects. I think this is an excellent record. The uranium industry in probably the first major industry dealing with a potentially toxic material in which nobody, as far as we know, has suffered any injury even during the start-up period. But, the question remains: do we have to spend more money on health protection from now on, or can we continue as we are?

QUESTION: Would Dr. Neuman comment on the accuracy of estimating body burden from urine specimens.

NEUMAN: This was discussed by John Hursh, who used excretion data to calculate the dosage figure, and then compared the results with the known dosage. His estimates were off by a factor of 1 to 7. These patients had received only one injection. Birney Fish also gave some examples of estimating body burden.

QUESTION: That was a nice experiment. Do you think urine samples give a low estimate or an overestimate of body burden?

NEUMAN: I can't answer that question directly. If we are called in, for example, in the case of an accidental exposure, and we have all the information (the kind of exposure, a good estimate of the air-borne concentration, and whether the material was soluble or insoluble) as well as the urine data, we can estimate just by using common sense the possible range of the exposure. But in such a case one is not using only urine data, but the other information also.

QUESTION: With reference to the paper by Maynard and Downs, it was interesting that the excretion levels in their data would tend to give an underestimate of body burden.

NEUMAN: I think the most significant feature was shown in the figure that indicated that, irrespective of the rise in lung burden, the urinary concentration had not changed.

QUESTION: With respect to that experiment, don't you think those dogs may have been con-

taminated, or that the results reflect contaminated samples?

NEUMAN: In these particular experiments extreme care is taken to avoid contamination because with lymph nodes the damage would probably be magnified; with this kind of tissue, a little contamination could influence the results a great deal. These analyses are done by one man, who is extremely careful and conscientious. I admit there is danger of contamination, but I believe in this case the results are reliable. In the lung sections black dust can be seen that has accumulated, and this is obviously uranium; it is visible on the histologic slide.

QUESTION: With respect to the low levels in urine, such a finding does not necessarily imply that there is no problem with reference to deposition or storage in the body.

NEUMAN: Exactly; that is just the point I tried to make. A high urinary uranium is meaningful; a low one may or may not be meaningful.

HARRIS: I think we might add, one high urinary value *may* be meaningful.

QUESTION: Would you care to comment on the types of long-term chronic effects to be expected from uranium exposure?

NEUMAN: I have nothing definite in mind. There are data on accumulation in the lung, and mention has been made of difficulty in the healing of fractures. I don't think there is anything to become alarmed about, but I would certainly like to see an analysis of the incidence of such occurrences in the general population in comparison with that in exposed people, because it might turn out that it is quite significant and should be followed up.

The problem of damage in the skeleton is always with us. No one has resolved the results of low-level radiation in the skeleton. The people analyzing radium data find, first of all, roentgenological changes at body burden levels which seem to be getting lower as more studies are made. Histologic changes are seen at exceedingly low levels for radium.

BUTTERWORTH: In considering chronic injury one shouldn't forget the kidney. In the old lead industry in Britain, chronic kidney damage has been found after many, many years' exposure.

There is one point I would like to question. The MPL's you use for air exposure are very different from ours. In Britain the MPL is higher than in the U.S. We use the ICRP 1954 figure. As a result we also use a higher urinary level for the so-called action point, with the addition of safety factors. The action level is 300 μg/l urine or 470 d/m/l. This does not mean that at a slightly lower level we do nothing; we do investigate the conditions of work. It does not mean that when we are designing a plant, we count on a higher level and therefore may not spend as much money as you do. However, we have no proof or evidence to date that, as a result of using the higher MPL, any of our employees have suffered. I think this is an important point.

HARRIS: Your MPL is higher by what factor?

BUTTERWORTH: A factor of 6: three times 10^{-11} d/m/cc, times another factor of 3 because only 8 hr of the 24 are spent at work. This is the more severe application that can be made of the ICRP recommendations. This does not mean we ever have such high levels, but we certainly do exceed your MPL.

HARRIS: This is really higher by a factor of two than the number we normally use.

BUTTERWORTH: I mention this because we have used this MPL (not necessarily to its highest level) since 1954, during operations similar to those in this country, apparently with no adverse effects. I am not saying that one should therefore be less careful in one's operations, but I think it is our duty not only to protect the individual but also to protect the community against unnecessary expense. I think this is the point you made earlier.

HARRIS: This is obviously true. Although a factor of two may not seem very large, in the design of a plant it could mean a great deal of money. An operation run with an MPL of 5 to 7 times 10^{-11} (your numbers) would not require any additional expenditure for safety, whereas to reduce the MPL to our level would require considerable expense. We must recognize that reduction of the MPL even by a factor of two would require a very large monetary outlay.

QUESTION: Do we know enough in the industry to design a plant with the exposure lower by a factor of two?

HARRIS: Yes.

QUESTION: Can we reduce the air count by a factor of two in an existing plant?

HARRIS: Yes, we can. As a matter of fact, in practice if levels are found to be 120 to 150% of the MPL, engineering control measures are introduced, which may in fact reduce the concentration by a factor of 10; however, the money is still spent. Many operations do result in levels 120 to 150%,

or perhaps even 200%, of the MPL; the question is, should we do anything about it?

NEUMAN: As a taxpayer I assure you I don't like to spend money; as a humanitarian, I find it a sticky problem to weigh human life and injury in terms of dollars and cents. I feel that this problem should be handled by an appointed body, presumably some legislative or recognized scientific society, and that it is peripheral to our scientific interests.

QUESTION: I wonder whether there are any suitable data to be examined by a committee. There is a large group of exposed people; between 1943 and 1949 or 1950 a relatively large number had exposures that could be considered significant, 5, 10, or 15 times the present MPL. These individuals do exist in the country, whether or not they are still working in the uranium industry. They are the only suitable source of data from which to obtain answers to these questions: people who have had an apparently fairly high level of exposure and, wherever they are now, are still available for examination.

An epidemiological study is needed. I would suggest that if the alternatives are to spend more money for more urine sampling or to cut down on urine sampling and spend the money for an epidemiological study, the latter is more important. Although we might find only a few hundred exposed individuals, and we might have to wait until all of them eventually died, we might get some valuable information. Certainly, if there are going to be symptoms in the group, they will start to appear before they die. This would seem to be a more fruitful line of endeavor.

NEUMAN: The point is perfectly well-taken; it is a very good one. Certainly, at this time, there is no new evidence to suggest that the MPL should be raised. The persons currently being exposed are not being damaged (or so it seems), but this is no basis for raising the MPL. The only way to obtain such a basis is to study the group exposed at higher levels. I would also suggest that the longer the data remain unexamined, the longer the safe MPL will remain in doubt. The study should be undertaken fairly soon.

QUESTION: Mr. Harris spoke earlier about the justification for spending five million dollars to lower concentrations by a factor of 10. Was that in reference to Part 20 as it relates to licensees?

HARRIS: It was merely a comment on the fact that we have a new industry and we must examine our data to see whether something should be done to improve conditions. It refers to the milling industry, not the contractors but the licensees.

QUESTION: The MPL for licensees is one-tenth that for contractors, is it not?

BARKER: I think that is true.

HARRIS: I didn't think that was true.

BARKER: The values used within the plant are identical to those in *Handbook 52*, the change being that they are applied to a 40-hr work week instead of a 160-hr continuous exposure period. The same values are used in contractor operations, I believe.

HARRIS: I think the statement is wrong that there is any significant difference between *Handbook 52* values and Part 20 values. There is a lower MPL for the general population, but this is consistent with the general view that exposure of the general population to any toxic material must be lower than industrial exposure. Variations in age, general health, etc., are taken into consideration.

SESSION IV

Is There Significant Correlation Between Alpha Surface Contamination and Air Concentration of Radioactive Particles in a Uranium Feed Materials Plant?

W.L. UTNAGE
Mallinckrodt Chemical Works, St. Charles, Missouri

INTRODUCTION

A study of alpha surface contamination and alpha air concentration was made at the Mallinckrodt Chemical Works, Uranium Division Feed Materials Plant, to determine whether or not there is significant correlation between surface contamination and air concentration in a plant where normal operations contribute sufficient alpha dust particles to the environmental air to produce average air concentrations greater than zero but less than the MAC established by NBS handbooks.

For many years various health physics groups working in connection with AEC programs have used alpha survey meters to detect leakage of radioactive particles from a processing or laboratory system. Surface contamination measurements are often used as a qualitative and quantitative measure of the efficiency of contamination control equipment, supplemented by other sampling and analysis methods to determine the potential health hazard to employees. There has never been sufficient agreement between health physics personnel throughout the AEC program to set meaningful limits on surface contamination as it relates to a dust health hazard. Most of us would agree that, if surface contamination were the only source of dust, it would contribute to the air concentration; on the other hand, if other sources of air contamination are more prevalent, surface contamination may have little importance.

Uranium feed materials plants, mills, and mines handle large tonnage quantities of uranium daily, and their production operations do not in any way resemble laboratory operations, but rather those of large commercial plants or metal foundries. Process equipment is of a type unsuited to remote control or completely enclosed operation. Such plants cannot be operated at zero exposure. Obviously it would be extremely expensive and in most cases impractical, if not impossible, to operate these plants like laboratories. Thus, some compromise must be made between no exposure and the MAC established by the National Committee on Radiation Protection.

DEGREE OF HAZARD

If the current methods of operating feed materials plants are accepted, then some leakage of material and some degree of air contamination must also be accepted. Limits on air concentration have been established, but a question remains regarding surface contamination. If surfaces are contaminated with radioactive materials that are beta and gamma emitters, there is definitely a problem of external radiation. However, if the contaminating material is predominately an alpha emitter, external radiation is not a problem; the problem becomes one of internal exposure in the event that the surface contamination becomes airborne. If alpha contamination is fixed on a surface, a high contamination reading is again meaningless.

These considerations make it evident that, with respect to surface contamination, one rule cannot fit all production or laboratory operations.

Normal uranium is a radioactive material of a low order of activity. It requires $\approx$2 tons of pure uranium oxide or fluoride to produce one curie of alpha activity. There is negligible gamma activity and very little beta activity, particularly when the product has just been refined. The major exposure problem is that of dust inhalation; surface contamination from normal uranium presents little in the way of an external exposure hazard.

DESCRIPTION OF CORRELATION SURVEY

Since a uranium feed materials plant is not operated like a laboratory, some surface contami-

Table 1

Alpha Surface Contamination and Air Concentrations of Natural Uranium in an Ore Sampling Plant

Air sampler location	Dust concentration, dis/min/m³ air	Floor type	Floor surface contamination, dis/min/100 cm²
Hopper loading control station	4	metal	10,100
Tare weight station (hopper)	5	cement	24,900
Drum lid bolt cutting station	6	metal	1,750
Auger station operating platform	13	"	11,600
Incoming drum conveyor	15	"	10,260
Drum lid removal station	19	"	3,200
Drum dumping station	26	"	17,400
Drum reclaiming station	27	"	7,000
Drum washer	30	"	12,350
Drum tare weight station	33	"	14,400
Lid securing station (auger)	33	"	8,350
Portable hopper loading area	34	cement	44,800

Table 2

Alpha Surface Contamination and Air Concentrations of Natural Uranium in a Uranium Metal Reduction Plant

Air sampler location	Dust concentration, dis/min/m³	Floor surface contamination, dis/min/100 cm²
Center of maintenance cage (no uranium processing in this area)	37	1200
Metal breakout chipping area	13	300
Reduction area	12	400
Crucible disassembly	12	2200
Jolter-filling machine	8	900
Saw area	29	3300
Metal breakout area	21	300
Upper filling machine area	72	500

nation can be measured in any production facility. The Mallinckrodt Health Department made a survey to determine whether or not there is a correlation between alpha surface contamination and air concentration, or, more precisely, to determine whether or not surface contamination contributes significantly to air activity in a plant where operational sources cause the bulk of air contamination.

Floor areas free from loose materials (floors which had been vacuumed) were chosen for the meter survey. Contamination was measured with a Victoreen 356 alpha survey meter. Beta and gamma contributions were eliminated. Air samples were taken in the same areas by drawing air through a sampling paper and counting the activity on an alpha scintillation counter. Air concentrations were then related to floor contaminations.

Data were taken in an ore sampling plant, in the waste products section of a refinery, in a UF_4 production plant, in a metals reduction plant, and in a uranium products warehouse.

RESULTS

Results of the survey show no correlation between surface contamination and air concen-

The Development of Surface Alpha Contamination Limits

A.F. Becher
Union Carbide Nuclear Company, K-25 Plant, Oak Ridge, Tennessee

Inhalation of the various uranium compounds processed or handled in large-scale production facilities is the primary health hazard associated with uranium, and, since the action of inhalation is an involuntary one, some type of external detection and control is required. Sampling of the environmental air is usually considered to be the most direct and efficient means of evaluating the personnel exposure potential, and at our plant this problem is considered first. The local program includes the use of commercially available high volume air samplers, low flow rate samplers (approximating the breathing rate of man) used for obtaining breathing zone samples for specific operations or service activities, and locally developed continuously recording air samplers (see Figure 1). These last record both 0.5-hr peak values and 8-hr average values on a tape, which is also identified as to time, date, and year for the recorded sample. An alarm device is under local development for use in conjunction with or independent of the above recording unit, which it is hoped will permit the detection of alpha air activity in the range of about 10 times the plant acceptable limit within a maximum elapsed time interval of 30 min.

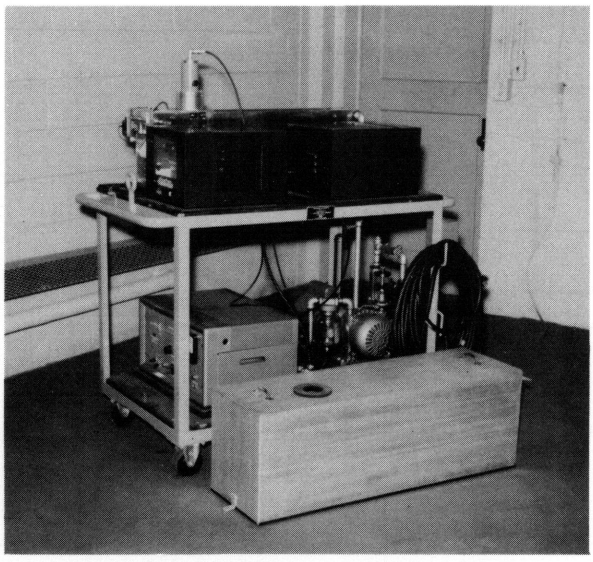

Figure 1. Air sampler.

It would thus seem that environmental air sampling provides a reasonably effective control technique, permitting prompt application of remedial measures, and this is probably true for locations where the work assignments can be well defined and are limited to relatively confined locations. However, the Oak Ridge Gaseous Diffusion Plant (ORGDP) includes many buildings potentially capable of becoming contaminated, comprising $\approx$15½ million ft^2 ($\approx$350 acres) of floor space, and of this more than one million ft^2 (23 acres) are routinely found to be contaminated to some extent; furthermore a large proportion of the operational and service personnel may be assigned to almost any location. Under these conditions it would be difficult if not impossible to keep track of each individual's internal dose during all his activities by any currently known air sampling technique in order to control exposures to NCRP limits.

Figures 2, 3, and 4 give some indication of the size of the building units involved; the U-shaped units of the original K-25 cascade are $\approx$400 ft wide, and the legs of the U $\approx$½ mile in length (Figure 2). Figure 4 shows a typical work area (upper sketch) surveyed within a unit (lower sketch) subject to contamination during normal work activity.

It was also recognized that evaluation of personnel exposures and estimation of body burdens by bio-assay techniques is difficult unless analyses are sufficiently frequent to permit averaging of the indicated exposures over a significant period, say 13 weeks, and therefore this type of program is probably more pertinent in plants where the jobs are of a more repetitive type and are done under closely controlled conditions. This type of analysis usually provides "after the fact" indicators of exposure, and consequently would be of little if any assistance in identifying the offending process or work activity in our widespread operations.

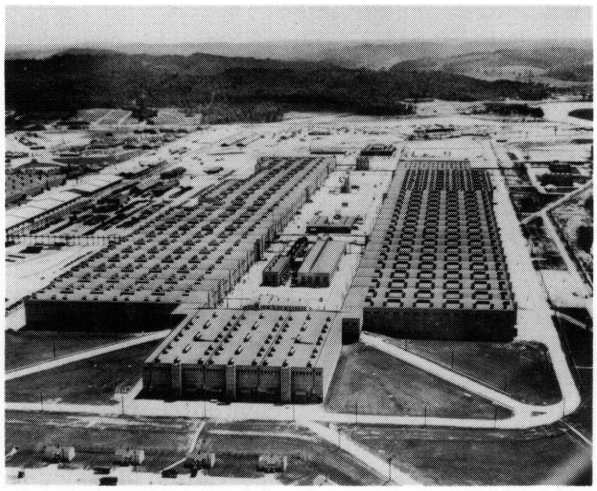

Figure 2. Union Carbide Nuclear Company, K-25 Plant.

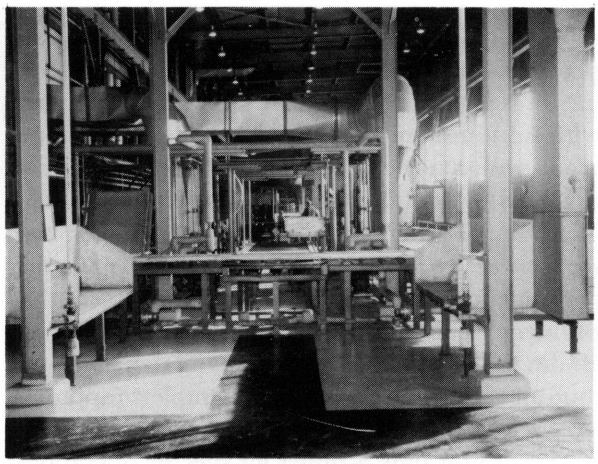

Figure 3. Interior of plant.

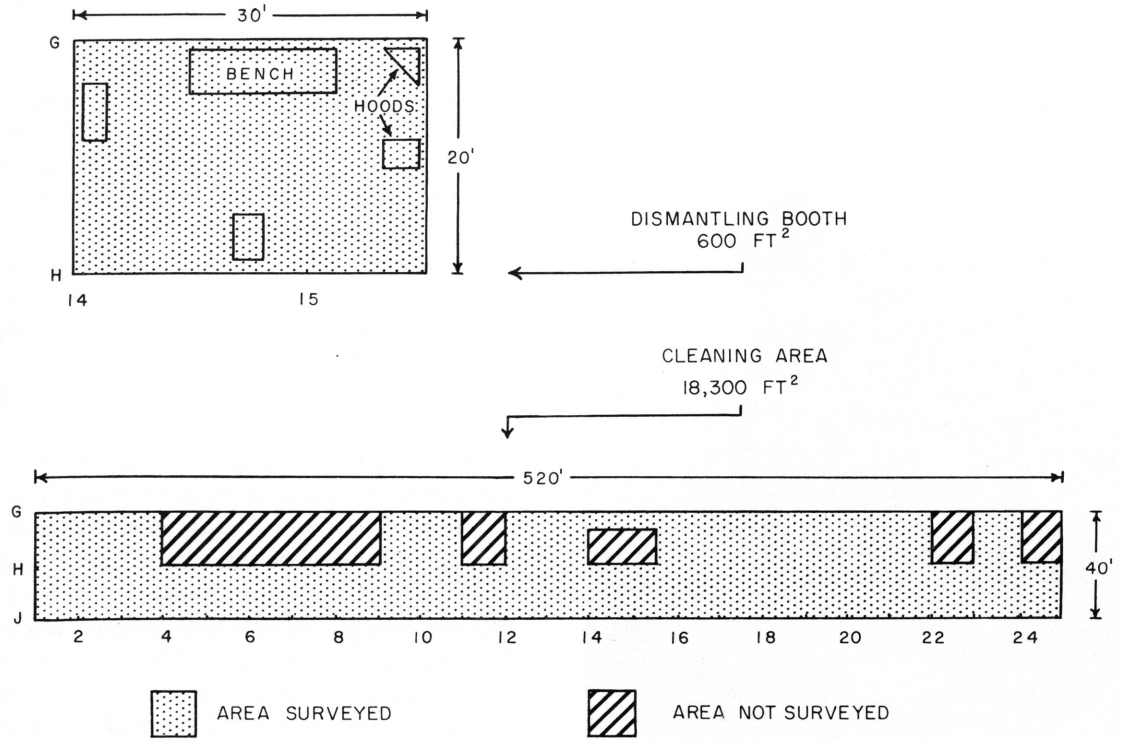

Figure 4. Typical cleaning facilities for small equipment.

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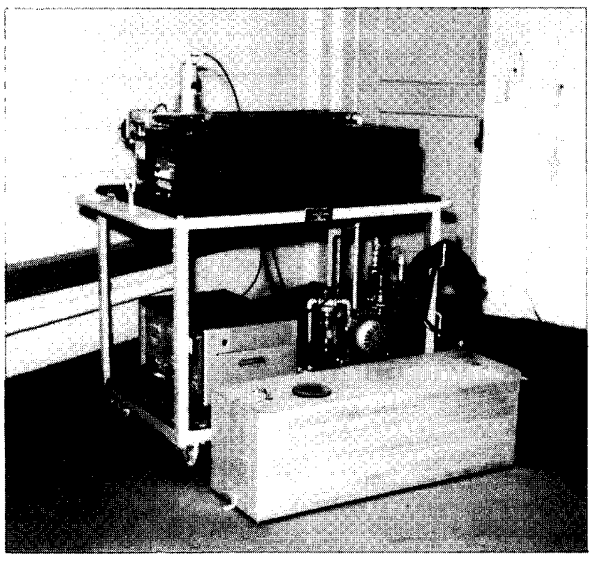

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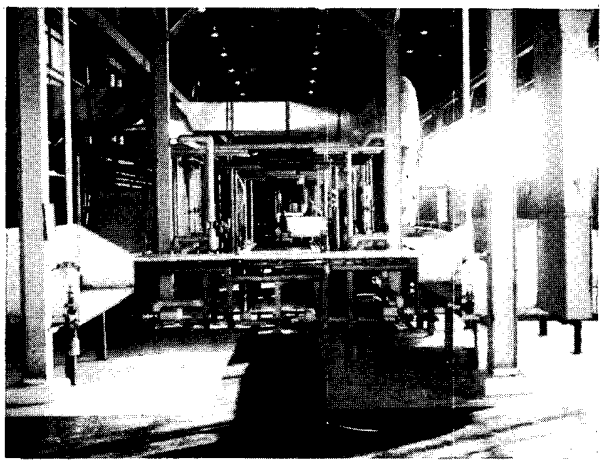

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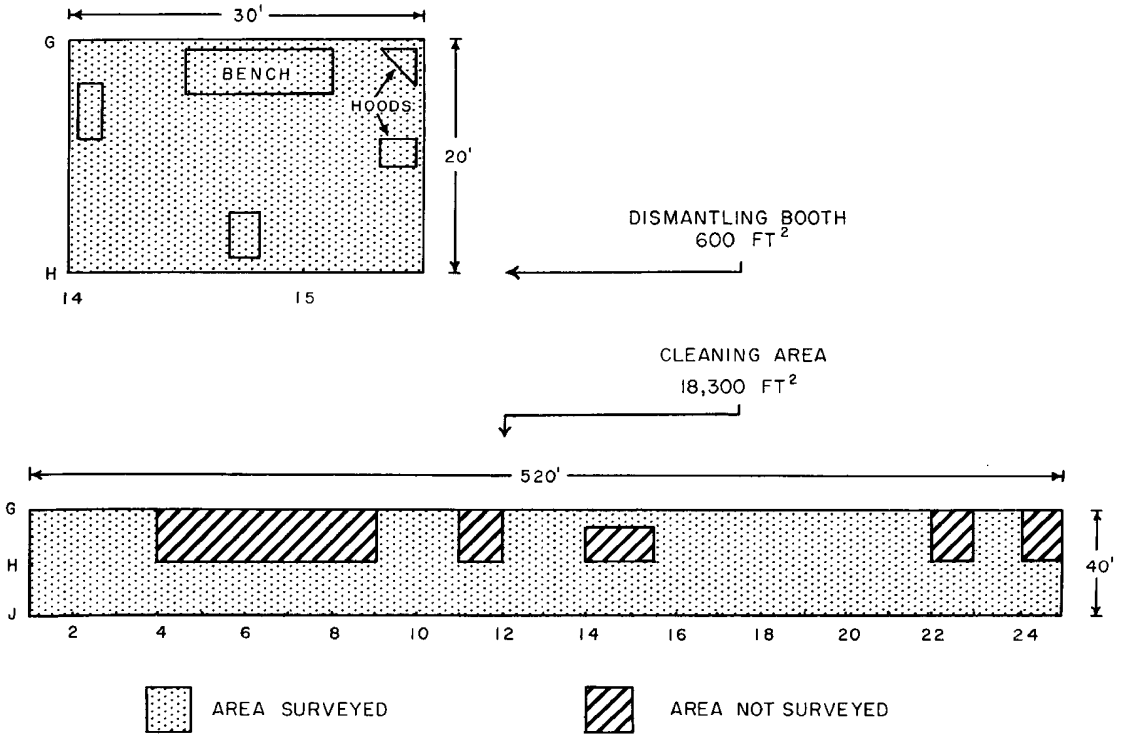

Figure 4. Typical cleaning facilities for small equipment.

In view of the above, since surface measurements are simple and generally meaningful in that the greatest personnel exposure potential should be related to locations with the most unconfined uranium, it became of interest to see whether surface measurements could be quantitatively related to probable intake and at the same time be made practicable administrative tools for the local management as an adjunct to the air sampling program.

Surface contamination within the work environment has previously been recognized as significant as a source of potential personnel exposure, and this problem has been specifically identified by Dunster[1] and by Bailey and Rohr.[2] Their findings establish fairly well that a given surface activity level can be related to a corresponding air activity level. Whether this air activity, as "fallout," subsequently contaminated the work surface or whether activities on the work surface produced the air problem because of ventilation, the worker's motions, etc., is irrelevant to the exposure problem involved. Dunster further reported on a specific case involving an area where pressurized suits worn by workers in contaminated areas were cleaned under a water spray. No transfer of air activity through the water-spray area was noted; however, tracking of contamination on the floor surfaces was reported. Workers whose sole source of exposure was due to this surface contamination were found to have detectable amounts excreted in the urine.

Instrument measurements reflecting changes in the surface activity levels are fairly simple to make and can be rapidly completed; thus they can be used to detect environmental changes, and permit rapid initiation of changes required to correct defects or equipment failure as well as operating deficiencies or errors. Obviously, the evaluation of surface contamination by survey instruments would be of little value if the work surfaces were allowed to become heavily contaminated, because the self-absorption due to the short range of the alpha-particles and the irregularities of the surfaces would tend to screen heavy deposits, so that significant quantities of uranium might be permitted to accumulate unless the surfaces were periodically surveyed and cleaned as necessary; such deposits are not only a potential health hazard but also may affect detection instruments. The requirements of various governmental agencies such as the AEC, ICC, etc., for the transfer or sale of equipment with suspected surface contamination as well as for the performance of work by subcontractor personnel without special precautions also specify the use of surface measurements of alpha activity and recognize the practice of averaging such contamination levels over the surfaces of equipment.

A simplified method of averaging both total surface activity and transferable activity from such work surfaces as floors, benches, table tops, etc., has been developed at the ORGDP; the result is referred to as the contamination index or CI (see Table 1). Since these indices reflect average values and a weighting factor is applied to compensate for specific ranges of measured activity, a rapid, uniform survey can be made of any plant area, and these can be totaled rather simply to comprise a larger component, if necessary, for reporting to the operating supervision concerned. As shown in Table 1, the CI reflects the sum of the fractions for any given work location within the survey area, weighted according to the range in which the measured activity levels fall, this being done for both the surface and transferable activity of concern.

For an area in which all measurements of surface activity were $>10{,}000$ counts/min/100 cm^2 and all the smears had values >2000 counts/min/100 cm^2, equal weights of 20 each would be applied to 100% of the area under survey, and this would yield a CI of 4000, the maximum value possible. At the ORGDP the CI of the most heavily contaminated areas has been lower than this by a factor of 2; in fact, the CI for 45 areas, comprising the most heavily contaminated plant areas, during 1957 was only 138. The survey form utilized provides space for recording the readings in various ranges of alpha activity, the total area involved in each class, etc., and gives a comparative index between the current and the previous audit which immediately reflects improvement or retrogression. A standard $\bar{\chi}$ graph (Figure 5) is used to evaluate the performance of the various operating groups by plotting the results of current surveys against the experience over the past several years, which minimizes normal fluctuations such as those due to cleanup, maintenance activities, etc.

Time does not permit a detailed discussion of controls and techniques; however, our experience in connection with the measurement of transferable activity seems worthy of mention. In our case, the transferable activity is determined by a

Table 1

Contamination Index Weighting Factors

The alpha contamination index is a weighted value which reflects both the degree and the extent of the contamination found. Essentially, it is the product of the area factor and a weighting factor depending on the contamination intensity, summed over the entire surface. For convenience at ORGDP, the contamination intensities for transferable and surface contamination are separated, each being divided into three classes with appropriate weighting factors; the ranges of these classes are given below.

Surface contamination range			Transferable contamination range		
Class	(counts/min/100 cm²)	Factor	Class	(counts/min/100 cm²)	Factor
I	500 – 2000	1	I	100 – 500	1
II	2000 – 10,000	5	II	500 – 2000	5
III	>10,000	20	III	>2000	20

In determining the over-all contamination index for any location, the contamination indices for the three classes in that location are summed by the following relation:

$$\text{Contamination index} = (A_{\text{I}}+5A_{\text{II}}+20_{\text{III}})_{\text{surface}}+(A_{\text{I}}+5A_{\text{II}}+20A_{\text{III}})_{\text{transferable}}.$$

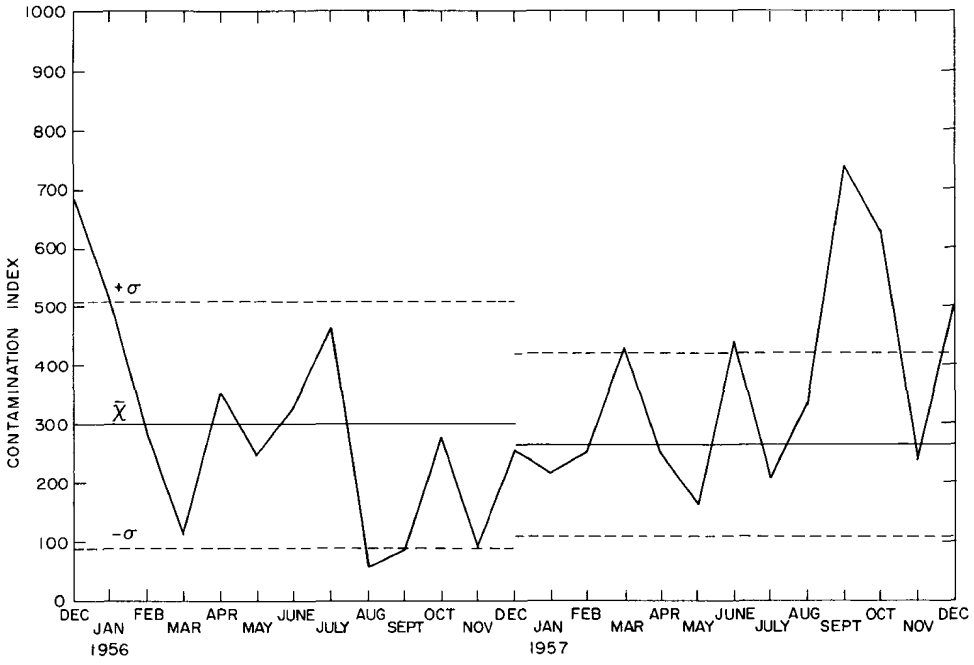

Figure 5. Contamination index in a typical operating area.

"smear" technique, and the differences in results due to variations in the technique as applied by different persons were checked. These differences included variation of the area smeared from 33 cm² to 320 cm², application of light to heavy pressure, and use of Whatman #41 filter paper or a 4×4-in. square of paper toweling. The results were calculated without correction for these differences; and, despite this, the standard deviation from the mean for 80% of the samples was ≈40%, which compares favorably with that reported by Solon and Barry,[3] who, under closely controlled test conditions, found 80% of the smear results within a 20% deviation from the mean. Since that

time the survey techniques have been standardized and give more accurate appraisals of the operations audited. Since the individual smear technique could be relied on to give a fairly accurate appraisal of the transferable surface activity at the point of smear, further study was initiated to evaluate the contamination index in terms of this average transferable activity, with the following results.

1. Even under the many varying field conditions, ≈50% of the numerical contamination indices of 100, 500, etc., are produced by the transferable surface activity component of the index.

2. Irrespective of the range calculated, the average value for the "smear" measurements will fall within the range of concern.

3. The average transferable activity, measured in counts/min/100 cm^2 by the Samson alpha survey meter with an over-all efficiency of 20%, agrees fairly closely with the calculated indices; thus, a contamination index of 10 approximates an average transferable activity of 10 counts/min/100 cm^2, etc. If the value is 10, we consider the location to be uncontaminated; a value of 1000 is considered the point at which uranium exposure may become significant.

It would be well to emphasize at this point the importance of standardizing the survey evaluation methods and actual measurement techniques to eliminate insofar as practicable the differences between persons doing the tests, since these differences could cause significant fluctuations in the calculated indices.

Bailey and Rohr[2] established the relationship of surface to air activity of "normal" uranium compounds both under simulated conditions and in selected operational areas, and reported it as the ratio (dis/min/m^3 air)/(dis/min/cm^2 surface). The results of this earlier work (Table 2) at the ORGDP are shown together with the more recent results obtained under actual operating conditions; the latter are seen to validate the earlier findings. This is perhaps the more significant considering that the later field conditions involved both soluble and insoluble uranium compounds and also the fine UO_2F_2 particulates formed by hydrolysis of UF_6 released to the atmosphere.

On the above basis, if a straight average is computed for the transferable activity, 6000 counts/min/100 cm^2 would correspond to the air value at the plant acceptable limit (PAL), which corresponds to the NCRP value for a 40-hr/week exposure of 5.1×10^{-11} $\mu C/cm^3$; however, it is recognized that the use of average values would permit individual instances exceeding the PAL. Furthermore, the most reliable range of the contamination index, which would correspond to an air activity at the PAL, is between 1000 and 3000, or a mean value of 2000, which is felt to be not only acceptable but also safe. It should be noted that none of these values is used as a control limit, since individual exposures can not be adequately controlled by average conditions, and there are other considerations such as the control of clothing and hand contamination, smoking, etc. (as will be explained in the next ORGDP paper). Thus, the contamination indices of general concern locally are as follows: <10, the area is considered uncontaminated; between 10 and 100, good housekeeping and personal hygienic practices are encouraged; between 100 and 1000, these practices are made mandatory and close control is exercised over personnel activities; and >1000, this level is considered the potential hazard point for purposes of control. (These will also be discussed in more detail in the next paper.)

It has been found practicable and in fact economically desirable at the ORGDP to aim the control limits at preventing essentially any expo-

Table 2

Relation of Air-Borne Alpha Activity to Surface Contamination

Source of data	Ratio (d/m/m^3 air)*/ (d/m/cm^2 surface)**
Two operating areas (1953) having highest contamination levels	
Shift-length air samples	0.64
Spot air samples	1.90
Plant-wide operations, 415 surveys (9 mo. 1958)	
Shift-length air samples	0.36
Spot air samples	5.05
Special test conditions (1953)	
Simulated operations	13.0
Worst possible conditions (short periods)	20.0

*Air activity as measured from a 9-cm disc of Whatman #41 filter paper, with fixed counting equipment, yields a counting and collection efficiency of 30%.

**Transferable activity from surfaces as measured by the Samson alpha survey meter gives an efficiency of 20% (including geometry factor).

sure, and not at integrating an individual's exposure to permit accumulation of the maximum permissible internal dose for uranium materials.

The work reported by Dunster gives similar limits for uranium-contaminated surfaces, although it should be recognized that this latter work at AERE was reportedly based on exposure to the most hazardous isotopes such as plutonium, radium, and strontium, with an increase in the reported control values by a factor of 10 being permitted for all other alpha emitters excepting radium, polonium, plutonium, and actinium. Their suggested surface limit of 1.5×10^{-4} $\mu C/cm^2$, which would yield the maximum permissible limit in air, as well as the value of 10^{-5} $\mu C/cm^2$, which is the limit at which they consider surface contamination to be no health problem, are in good agreement with the similar values established at the ORGDP.

It appears therefore reasonable to conclude that:

1. Surface contamination levels, as expressed by the contamination indices, will reflect operational changes or inadequacies in the control methods.

2. High surface contamination levels are almost invariably identified with correspondingly high air activity measurements.

3. Standardization of survey techniques and areas permits a rapid, simple determination of the surface contamination levels to be made by relatively inexperienced personnel, which, when expressed in terms of the contamination index, provides management with an effective yardstick to measure performance and evaluate the effectiveness of the control measures employed.

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Personnel Contamination as a Uranium Hazard

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INTRODUCTION

In discussing the question of personnel contamination, it is recognized that the various possible routes by which uranium may enter the body cannot be considered as being individually independent of all other routes of intake, and that the total "radiation environment" of employees must be considered when establishing limits for contamination. Probably the primary factors as related to an individual's radiation exposure are the direct external radiation he may receive and the direct inhalation of air-borne radioactive materials. Secondary factors include the intake of uranium from contamination on the individual's clothing and person.

In order to maintain safe conditions with respect to radiation at the Oak Ridge Gaseous Diffusion Plant (ORGDP), air-borne concentrations of radioactive materials and exposure to penetrating radiation are controlled at or below the limits given by the National Committee on Radiation Protection. An attempt has also been made to relate the intake of radioactive materials from secondary sources of exposure to the NCRP values, and to control conditions so as to make it highly unlikely that any individual will receive as much as 1/10 of the maximum permissible exposure from any one secondary source. Although it is recognized that the intake of radioactive material from two or perhaps three secondary sources may represent additive effects, we consider it very improbable that any individual will be continuously exposed to conditions such that the limits for all types of exposure are closely approached; thus, as long as the appropriate limits are observed, it appears unlikely that the total contribution from such secondary sources will appreciably exceed 1/10 of the maximum permissible value.

Since uranium contamination on work surfaces or personnel presents no hazard except as a source from which uranium may be taken into the body, the problem of evaluating personnel contamination is one of determining such intake as a function of contamination levels. This paper concerns some studies undertaken at the ORGDP to determine the intake of uranium as a function of hand and clothing contamination, and the relation of this intake to the limits as given by the NCRP for air and water concentrations, these being the only NCRP limits applicable to this problem.

The problem of uranium dust inhalation is of much more concern than the problem of ingestion because the quantity of uranium representing the maximum permissible exposure due to inhalation is very much smaller that due to ingestion. In addition, the prevention of ingestion is much more easily controlled by the individual involved than is the prevention of inhalation. In view of this relative importance of inhalation, our attention has been directed primarily toward the control of this type of entry into the body. The studies relating uranium intake to surface,[1] hand,[2] and clothing[3] contamination have been reported previously and will only be summarized here.

SUMMARY OF EXPERIMENTAL STUDIES

Hand Contamination

One of the principal mechanisms by which material on the hands may be inhaled appears to be transfer to cigarettes and subsequent inhalation with the smoke. In order to determine the amount of uranium that might be inhaled in this manner, a work surface contaminated with uranium tetrafluoride was rubbed with the hands, and the uranium picked up in this way was rubbed into the hands to simulate conditions that might be experienced during normal plant work. Cigarettes were handled for about 1 min each, the hands being recontaminated before each cigarette was handled. The amounts of uranium on the hands were determined by means of a Samson alpha survey meter, and the cigarettes were counted in a laboratory-type proportional counter. Supple-

mentary tests indicated a counting geometry of 15% for the Samson for hand contamination and a geometry of 30% for the laboratory counter in the counting of the cigarettes. These tests indicated that the transfer of uranium from the hands to the cigarette amounted to $\approx 1\%$ of the material on the palmar surface of the hands.

To determine the amount of uranium that would be inhaled as a result of smoking contaminated cigarettes, simulated smoking of cigarettes contaminated with a known amount of uranium was done by air-sampler devices which collected the material in the smoke drawn through the cigarette while an additional sampler collected the smoke escaping from the end of the cigarette. Several types of collecting devices were used including two water bubbler collectors in series, two Whatman #41 filter papers in series, and, in final tests, single Millipore filters. Uranium compounds used were uranyl nitrate, uranyl fluoride, and uranium tetrafluoride. The fraction of uranium on the cigarettes which appeared in the smoke varied from 0.02 to 0.69% for various series of tests, the highest value being found for uranium tetrafluoride. It was concluded that $<1\%$ of the uranium material on a cigarette would be inhaled by smoking.

On the basis of these test results, it is computed that the hand contamination value that would produce the maximum permissible inhalation of uranium from this source alone would amount to 5.7×10^5 dis/min on the hand surfaces contributing to this type of intake for an individual smoking 20 cigarettes a day with contaminated hands. In arriving at a hand contamination limit for plant applications, only the palmar surfaces of the hands were considered to contribute appreciably to this type of intake; as far as inhalation from smoking is concerned, this would permit hand contamination of 2.8×10^5 dis/min on each hand surface.

In evaluating the ingestion hazard from hand contamination, the rather conservative assumption was made that, through repeated contact with contaminated surfaces and repeated ingestion from the hands, a man might ingest an amount equal to the total amount on the palmar surfaces of the hands. On the basis of the NBS *Handbook 52* values, this would permit a contamination of 2.5×10^5 dis/min on each hand surface.

There is no information which would permit a direct evaluation of the effects of skin absorption, but a comparison of the lethal effects of uranium by ingestion and by absorption through the skin (taken from some of the early animal studies at Rochester[4]) indicated that solutions of some uranium compounds may be approximately as toxic by skin application as by ingestion. The assumption was therefore used that material on the backs of the hands is absorbed to the same extent as if it had been ingested, but that absorption through the thicker palmar surfaces of the hands is negligible. On this basis, the hand contamination which would lead to the absorption of the maximum permissible amount is the same as that calculated on the basis of ingestion, i.e., 2.5×10^5 dis/min on each hand surface.

If it is assumed that the effects of ingestion, inhalation, and skin absorption are additive, which may be the case if soluble compounds are involved, the amount on the hands to produce the maximum allowable combined intake is found to be 8.6×10^4 dis/min per hand surface, the reciprocal of this value being simply the sum of the reciprocals of the individual values for the various types of uranium intake indicated above. With the 15% geometry for instruments used at the ORGDP in monitoring hand contamination, this corresponds to a counting rate of 13,000 counts/min per hand surface.

The limit for hand contamination at the ORGDP is 1000 counts/min per hand surface, corresponding to 6700 dis/min per hand surface. On the basis of the evaluation just outlined, this would correspond to a uranium intake from hand contamination of $<8\%$ of the maximum permissible amount with hands contaminated continuously to the limit for five days per week. Safety factors inherent in this evaluation include the assumption or experimental use of loosely bound uranium materials with respect to inhalation and ingestion, and the assumption of a very soluble uranium compound in the consideration of skin absorption.

Clothing Contamination

In the past the question of the significance of alpha contamination on clothing has presented some serious administrative problems, and widely divergent views were held with respect to the health aspects of such contamination. Some felt that any contamination at all on clothing represented a serious hazard, while others felt that

clothing contamination was quite insignificant as a health hazard.

In order to evaluate the inhalation of uranium from contaminated clothing, the test procedure was to have personnel perform routine work in essentially uncontaminated locations while wearing clothing purposely contaminated with uranium tetrafluoride powder, and to determine the amount of uranium that would have been inhaled from this source by analyses of filters from the respirators worn. The release of material from these test coveralls was compared with the release from coveralls contaminated during normal plant work by having personnel perform standard series of calisthenics while wearing test coveralls contaminated with uranium tetrafluoride and then while wearing coveralls contaminated during plant work, and determining the air-borne activity by analyses of filters from respirators worn during the tests. The results of these tests could be related to plant conditions through routine survey data. On routine surveys the highest reading on an individual's clothing is recorded as a measure of his clothing contamination; and, under plant conditions, this maximum was found to be about 3.5 times the average reading on the front surfaces of the coveralls. The pertinent relation can be expressed by the two equations,[3]

$$C_A \text{ (dis/min/m}^3\text{)} = 0.07 C_C \text{ (dis/min/cm}^2\text{)},$$

$$C_A \text{ (fraction of MAC)} = 1.03 \times 10^{-4} C_C \text{ (counts/min/100 cm}^2\text{)},$$

where C_A is the air-borne contamination and C_C is the high-spot clothing contamination in the units shown. The MAC is the NCRP value for a 40-hr week.

From these equations the high-spot reading corresponding to the inhalation of the maximum allowable air-borne concentration is 1620 dis/min/cm^2, or, with the geometry of 6% found for the Samson alpha survey meters at our plant, 9700 counts/min/100 cm^2. At the ORGDP 4000 counts/min/100 cm^2 is used as the limit for clothing contamination, which would correspond to about 40% of the maximum permissible uranium inhalation with continuous exposure to clothing maintained at this level by continual recontamination. It should be noted, however, that uranium materials were found to drop off the test contaminated clothing fairly rapidly, about half coming off in the first 2 hr, and the calisthenics tests indicated that the uranium materials on the plant-contaminated clothing came off at about the same rate. It is perhaps obvious that if the material on the clothing decreases, the amount of material in the air will also decrease with time, and therefore, once sources of repeated contamination are removed, inhalation from clothing will average below this maximum figure. On the other hand, if the material does not come off, it is also obvious that there will be no air-borne contamination from this source. Thus, it appears that clothing initially contaminated to the plant limit represents no hazard to the individual wearing it or to those with whom he associates. However, as will be indicated below, the plant controls on contamination make it extremely unlikely that any individual will be contaminated to this degree routinely.

An analysis was made of routine survey data accumulated over a period of one year to determine the amount of contamination found on an employee's clothing under different levels of surface contamination. The clothing contamination was found to vary approximately as the square root of the contamination index (CI); this relationship is shown in Figure 1, where points representing the average clothing contamination are plotted against the average CI for each of four decades of the latter. The line represents the product of the square root of the CI and a constant which was found by statistical analysis to be 41.

If this expression is combined with the one relating inhalation to clothing contamination, the result is the expression represented graphically in Figure 2. The contamination index that would result in clothing contamination sufficient to cause the maximum allowable uranium inhalation is seen to be actually outside the range of CI values that we use, as the degree of contamination necessary to produce it is far greater than the levels found at our plant. The CI of 1000 corresponds to an intake of 16% of the maximum allowable intake.

A similar study of hand contamination gave the results shown in Figure 3. The visually fitted curve shows a rather definite curvature; this is probably due, at least in part, to the routine use of gloves by employees working with highly contaminated equipment. The use of gloves would tend to reduce the degree of hand contamination, particularly in

highly contaminated locations, and would cause the curve to have this shape.

DISCUSSION

The above studies provided the basis for the plant's present contamination limits and verified the adequacy of the contamination control program at the ORGDP. The contamination control levels and the calculated intake of uranium at these levels are summarized in Table 1.

These values indicate that, without any special protective measures, the *average* exposure could be above the maximum permissible value only in areas where the contamination index was >1000. It is emphasized that this is on the average only: the control of the average exposure of a group of individuals to a value below the limit may not assure that each individual's exposure is below the limit. Accordingly, controls for individual jobs or operations are utilized as warranted by contamination conditions, and these controls may be over and above those normally used routinely in a particular area.

Probably the best example of such specific action is the use of respiratory protective equipment where air-borne uranium concentrations may exceed the long-term maximum allowable values. Plant experience indicates that, in the absence of special measures, short-term exposures of this nature would constitute a large fraction of the total exposure of employees, and the use of respirators during these short periods greatly reduces the over-all intake of uranium. Although we emphasize the use of respirators where air-borne contamination may be present, plant operations are, in general, engineered to maintain air levels below the NCRP limits. The vast majority of the plant areas have essentially no routine air-borne contamination, but respiratory protection is required when equipment containing uranium materials is opened, since experience has shown that air concentrations can be high for short periods during such operations.

An additional important check on the entire contamination control program and on possible individual exposures is the routine urinalysis program. The low levels of uranium intake that are maintained make it possible to keep urinary excretion rates to low levels also. Under these conditions a rather infrequent urinalysis sampling schedule appears adequate, with individuals

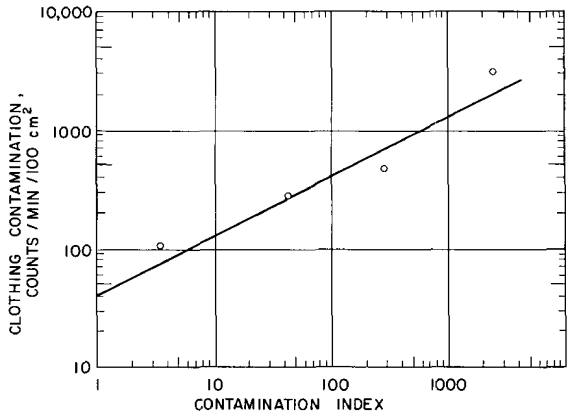

Figure 1.

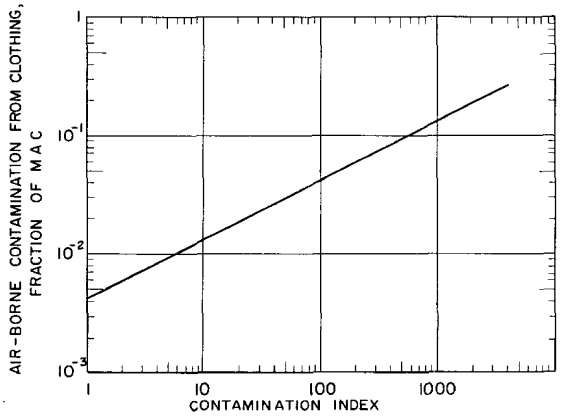

Figure 2.

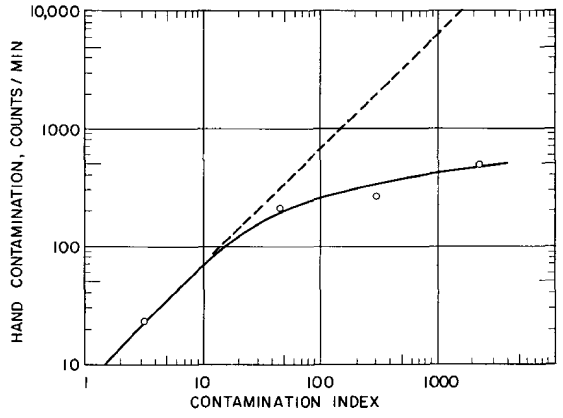

Figure 3.

Table 1

Average Employee Exposure at ORGDP Control Values (With No Special Protective Measures)

Contamination index	Calculated intake (Percent of maximum permissible)		
	Direct air contamination	Hands	Clothing
10	0.5	0.5	1.7
100	5	2.0	5.1
1000	50	3	16

checked at intervals of three to six months depending upon the probable degree of uranium intake.

With regard to routine operations where air contamination itself presents no problem, as Mr. Becher pointed out in the preceding paper, we consider any location with a contamination index of 10 or less to be uncontaminated, this being the point at which decontamination or minimal precautions are instituted. At each decade above this level, control and monitoring measures are increased. For example, routine air monitoring is carried out in locations where the CI is >100; and, in the one or two locations where it is >1000, respiratory protection and issued clothing are mandatory, operations in these areas being short-term only. Protective clothing is never required unless respiratory protection is also required, although issued clothing is available to anyone who wants it. Specific limits for air, hand, and clothing contamination have already been mentioned, which serve as control points for these specific items when they are above the ranges of contamination normally encountered. Thus, considering the additional controls used on special jobs, the actual exposures are probably lower by a factor of perhaps two than indicated in Table 1.

In summary, we feel that measures of surface contamination provide a very useful indicator of general contamination conditions, and that on the basis of such measurements reasonable control requirements for general operations can be established in locations varying widely with regard to the degree of uranium intake that may be experienced therein. We also conclude that where personnel contamination by uranium represents a source of any significant exposure, the control of direct air-borne contamination is a much more important consideration.

The delineation of areas where clothing contamination might represent a significant source of uranium intake has permitted technically sound administrative decisions concerning the conditions under which company-issued clothing should be regarded as protective clothing and where it should be regarded simply as work clothing.

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Figure 1. Wash room area with washer on the left, hydraxter in right foreground, and dryer in right background.

are shown in Table 2. The laundry scalers were set to pass only items contaminated to <100 counts/min. Alpha contamination results are also shown in Table 2. The acceptable level for alpha emitters was <500 dis/min. The 55-min wash cycle consisted of a series of rinses at temperatures up to 180°F, a 20-min solution wash, and a series of hot and cold rinses as shown in Table 3. The temperatures shown apply only to cotton cloth items. Rubber goods are processed at temperatures <140°F.

Shortly after these tests were conducted, the Turco Company modified the 4182 formula by changing from the normal salt to the acid salt of ammonium carbonate because the latter is commercially more available. Subsequent tests showed the new formula 4182-A to be as effective for decontamination as the original formula, and this agent is now being used in our regular laundry process. A complete description of the studies is given in the unclassified document HW-38218-Rev, July 29, 1955.

The wash cycle currently being used consists of three 4-min rinses, one 15-min wash in a solution of 15 lb Turco No. 4182-A in 150 gal water, and four 4-min rinses. All hot rinses and the solution wash are at temperatures of 180° to 185°F.

Before considering the problems involved in operating a laundry of this type, it will be useful to outline the flow of apparel from user to clean storage. Clothing worn in radiation zones or in other regulated areas is usually removed at "change stations" and placed in bags which are picked up at specified intervals by the laundry truck. Preliminary surveys are made by the customer group, on the basis of which heavily contaminated clothing is discarded and moderately contaminated clothing is separated from that with nominal contamination. All rubber goods are separated from canvas goods.

At the laundry, incoming bagged clothing is first stacked in a receiving area. The moderately contaminated clothing is processed separately to prevent cross contamination. The bags are dumped into a 400-lb-capacity, self-dumping, end-loading commercial washer in 250-lb loads. All operators are required to wear respirators. After the wash cycle, the canvas goods are dumped in 125-lb loads into a hydraxter in which water pressure on

Table 1

Efficiency of Various Solutions Tested

Decontaminant	Efficiency (% removed) Pu	U	F.P.	S.B.
Calgon (sodium hexametaphosphate)				
Formula A	85	—	38	44
Formula B	92	99.2	92	52
Turco				
Formula A	95	79	—	93
Formula B	96	92	—	91
Formula C	99.6	100	98.3	92.7
Formula D	98.5	100	99.3	91.7
Versene T				
Formula A	59	84	65	74
Formula B	55	89	76	84
Formula B (rewash)	64	99.4	95	93
Formula C	94	93	—	91

results indicated that complexing agents are most effective in removing metallic ions strongly fixed to cotton cloth. It was also shown that large quantities of water in the washer wheel provide greater possibilities for floating away loose radioactive particles and also give better results in removing complex or inactivated ions from cotton fabric. Soaps, clays, silicates, and other recognized commercial laundering agents were found to have little value in removing normal radioactive contaminants; whereas compounded agents containing glassy phosphate salts and carbonate and ammonium ions proved to be most effective. Some of these are listed in Table 1.

The best Calgon formula consisted of 4 parts Calgon to 1 part Hilco powder and 1 part soap (Lever Brothers 770). The best Versene T formula (formula C) consisted of 3 parts Versene T, 2 parts Hilco powder, and 1 part soap (Lever Brothers 770). The most effective of all decontaminating agents was found to be Turco No. 4182 (formula C). To determine the complexing effect of a fluoride, formula D, consisting of 3 parts Turco No. 4182 to 1 part ammonium bifluoride was tested, and almost the same efficiency was found.

All these tests were conducted on a laboratory scale (in a 5-gal stainless steel basin) with standard sized pieces of cotton material contaminated to known levels. The laundry cycle generally used consisted of one 3-min cold water rinse, one 3-min rinse at 120°F, one 3-min hot rinse at 180°F, a 15-min wash in the test solution, one 3-min hot rinse, two 3-min split rinses at 120°F, and one cold rinse. Rinsing, washing, and wringing were done by hand; the cloths were dried in laundry type hot air dryers at about 180°F.

In order to test Turco No. 4182 on a regular laundry process scale, all protective clothing received from one plant in one week was washed in a solution of 15 lb Turco No. 4182 in 150 gal water. All bags were premonitored and showed contamination to be at a normal level as measured with a standard VGM instrument. After processing, the load was monitored on regular laundry scalers for beta-gamma contamination; the results

Table 2

Field Test Results for Formula C, Turco No. 4182

	β-γ Measurements		α Measurements	
	Acceptable	Rejects	Acceptable	Rejects
Coveralls	705	3	702	1
Laboratory coats	2		2	
Rubber gloves	831		831	4
Misc. canvas goods	4110	1	4109	
Rubber shoes	663	2	661	3
Total	6311	6	6305	8

Table 3

Typical Wash Cycle

	Minutes	°F
1. Split rinse	5	120
2. Hot rinse	5	180
3. Hot rinse	3	180
4. Solution wash	20	180
5. Hot rinse	5	180
6. Split rinse	5	180
7. Split rinse	3	120
8. Cold rinse	3	60
9. Cold rinse	3	50
10. Cold rinse	3	40
Total	55	

Figure 2. Beta-gamma monitoring operation.

Figure 3. Alpha monitoring operation.

the opposite side of a diaphram is used to squeeze out the excess water. The damp-dry clothing is then dried in a 200-lb capacity, self-dumping, tumbler dryer in 125-lb loads. (See Figure 1.)

The area used for sorting, washing, and drying is considered a radiation zone. When dry, the clothing is moved to a contamination-free area for radiological survey, the type depending upon the customer operation. Some clothes are monitored for fission products and others for plutonium or uranium; many articles are surveyed for both alpha and beta-gamma emitters. For beta-gamma surveys a bank of 8 GM tubes is recessed into a table top under a wire mesh screen covered with paper to guard against contamination. The tubes are wired in pairs through a conventional counting circuit, and each of the resulting four registers is observed for appropriate release limits. (See Figure 2.) Surveys for alpha contamination are performed with an air proportional counter (Figure 3).

Current release limits for clothing depend upon the type of contamination found. For uranium not in equilibrium with its decay products, apparel may be released for further use if the survey indicates <2000 dis/min/100 cm^2 on rubber or <1000 dis/min/100 cm^2 on all other articles. For plutonium the release limit is 1000 dis/min/100 cm^2. For beta-gamma emitters the release limits are 3000 counts/min for uranium and 1000 counts/min for fission products. Clothing which does not pass these limits is rewashed, and clothing not acceptable after the third rewash is discarded. Acceptable clothing is inspected, repaired if necessary, and then folded and bagged by type for reissue.

The phases of this operation that have presented the most problems have been the sorting in the receiving area and the washing operation. In the early days, it was customary to dump each bag of soiled clothing on the floor at the washer and sort out the items desired for that washing cycle. This was necessary because each bag contained all types of wearing apparel, but is was potentially hazardous because items were encountered with all degrees of contamination. Air samples in the vicinity taken over a two-month period indicated concentrations ranging from 10^{-11} to a maximum of 4.4×10^{-10} μg Pu/cc air. To improve these conditions, customer cooperation was enlisted in surveying clothing before shipment to the laundry.

The following criteria were established: For beta-gamma contamination all articles released to the laundry are required to read <50 mrads/hr and the bag surface <6. Articles contaminated to >50 mrads/hr but <500 require special handling, including appropriate tagging, segregation, and shipment on a radioactive shipment release form with prior notification to the laundry. All wearing apparel contaminated to >500 mrads/hr is discarded to contaminated waste. For alpha contamination, the conditions of release of the articles to the laundry vary with the different facilities. In general, clothing contaminated with uranium is processed regardless of the original level, but clothing subject to alpha contamination is kept separate according to the facility. In addition, clothing potentially contaminated with plutonium is kept separate from that contaminated with uranium when possible. All persons working within the radiation zone of the laundry are required to wear protective clothing, and when moderately contaminated clothing is being sorted, respiratory protection is also required.

Experience gained in the solution of these problems over the years has resulted in the present laundry layout and design. The regulated receiving area is of adequate size to handle the volume of business anticipated. To prevent the spread of contamination, the washing area at HAPO consists of a large room physically separated from all but the receiving area. The floor drains in the washing and receiving areas are connected to a contaminated drain system to facilitate routine flushing of all floor areas. The exhaust air from the washing and receiving rooms is mechanically filtered to remove lint particles. The air stream through the dryers is filtered with a water-type precipitator connected to a contaminated waste drain.

To avoid the spread of contamination during the transfer of clothes from the washing area to the testing room, separate sets of laundry carts are used, one only within the washing and receiving areas, and the other only in the testing area plus a small part of the washing area near the dryers. Clothing is unloaded from the dryers directly onto the testing room carts for transfer from the washing area to the testing area. The testing area is considered a regulated area but with a contamination hazard appreciably less than that encountered in the washing area, since the clothing brought in has been previously washed and dried and the contamination remaining is relatively well fixed. Proper ventilation around each work area is

Figure 2. Beta-gamma monitoring operation.

Figure 3. Alpha monitoring operation.

required to remove the ever present lint which is potentially contaminated.

One additional feature required is the provision of changerooms where employees can change from street clothing to the protective clothing required within the regulated areas, including facilities for removing potentially contaminated protective clothing and issuance of clean protective clothing.

In summary, the operation of a Decontamination Laundry involves problems not normally encountered in routine commercial laundry operations, including the necessity for a suitable detergent that not only cleans clothes but also removes radioactive contamination. The problem of cross contamination is ever present and must be continually guarded against. Special precautions must be taken in disposing of waste solutions. Once processed, all apparel must be surveyed by suitable radiation monitoring instruments, and proper facilities must be provided for disposing of clothing that cannot be decontaminated.

The authors hereby express their gratitude for the assistance and cooperation of B.B. Evans, Manager of the Laundry Operation, and his entire staff, particularly J.L. Norwood, who supplied the data on decontaminating agents.

Laundry Operations in a Uranium Feed Materials Plant

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INTRODUCTION

This report is a summary of the interpretation at Mallinckrodt Chemical Works of the recommendation of the National Committee on Radiation Protection for laundering clothing contaminated with uranium. In-plant laundering, as practiced by Mallinckrodt, is briefly described, and some comparative cost data on in-plant laundering vs commercial laundering are presented.

Regulated clothing is required in many AEC facilities, under certain conditions, as a means of personal protection and to control the spread of contamination. Literal interpretation of the NCRP recommendations would require some percentage of persons engaged in uranium refining operations to wear regulated clothing. The National Bureau of Standards handbooks also contain recommendations for laundering contaminated clothing. The opinion at Mallinckrodt, based on interpretation of handbook recommendations, is that some in-plant laundering is necessary to reduce contamination to a level prescribed by the handbooks before release to a commercial laundry. Under these conditions the necessary monitoring and separation facilities would be part of the cost of laundry operations. On this basis there are data to support the argument in favor of using an in-plant laundry for all plant clothing, both from a contamination control and an economic standpoint. Some of these data are presented in this report.

NCRP RECOMMENDATIONS CONCERNING LAUNDERING

The NCRP recommendations as published in the NBS handbooks are summarized below with comments by the Mallinckrodt Chemical Works Health Department.

A. General

1. *Handbook 42* and *Handbook 48* contain most of the requirements for clothing and laundry; however, both state that "the handling of alpha emitters is not specifically included in this report." Consequently uranium, being an alpha emitter, is not specifically included. Because uranium is always in some degree of equilibrium with its beta-gamma emitting daughters, we at Mallinckrodt feel that it must be included in the application of these requirements.

2. The handbooks refer to operations in "laboratories" but do not specify factory operations such as the radiochemical processes in a uranium feed materials plant. We at Mallinckrodt feel that the Committee intended these recommendations to apply to any facility or operation handling radioactive materials and that our operations are directly involved.

3. There are no specific requirements for protective clothing where alpha contamination is concerned; however, *Handbook 42* defines "significant" alpha contamination and *Handbook 52* sets permissible concentrations of alpha emitters (including uranium) in air and water. We at Mallinckrodt feel that these criteria establish limiting factors for alpha activity that should not be exceeded in any public laundry as a result of processing clothing from a uranium feed materials plant.

4. The possibility of laundering in a licensed laundry has not been included in this summary because at the time of the writing we did not know of any licensed laundry.

B. Recommendations of Handbooks 42, 48, and 52 Pertaining to Clothing and Laundry

1. (A) (42-III) Persons working with radioactive materials *shall* be required to wear clothing which is restricted to that work.

(B) (42-I, 2) Common sense precautions dictate the use of protective clothing when there is danger of contaminating the person.

2. (A) (48-III, 1) Clothing which is contaminated *shall not* be removed from the work area until decontaminated.

(B) (48-III, 1) Under no conditions *shall* contaminated clothing be worn into clear areas.

3. (A) (42-I, 3) Extreme care should be taken to prevent contamination from reaching halls and offices where personnel not normally working with radioactive materials may come in contact with them.

(B) (48-III, 3) Effort *should* be devoted in the use of laundry facilities to insure the minimizing of spread of contamination.

(C) (42-III, 7) With long-lived isotopes, the *management is committed* to a prevention of contamination of the public domain.

4. (42-V, 3) Protective clothing should be washed and monitored under controlled conditions.

5. (48-III, 3) Protective clothing *should not* be released to public laundries until contamination is below permissible levels *and* is of short half-life.

6. (42-V, 3) Before contaminated garments are considered for release to public laundering service, the extent of hazard *shall* be carefully evaluated.

7. (42-V, 3) Special laundry facilities *should* be used by all groups regularly engaged in radioisotope work.

8. (48-III, 3) Note: Although not specified, it is implied that self-operated, commercial type laundry facilities are practical for large industrial concerns engaged in radiochemical work.

9. (48-III, 3) When monitoring reveals that clothing is contaminated above permissible levels, laundering may then be attempted. In the selection of any decontamination procedure, the chemistry of contamination shall be considered.

10. (48-III, 2) PERMISSIBLE LEVELS OF CONTAMINATION. Note: Uranium and daughters are assumed by Mallinckrodt to be in Group 1.

(A) 1 mrep/hr for beta or gamma radiation measured as the average near the surface of the garment. Note: It is undetermined whether "average" refers to the entire garment having an average of 1 mrep/hr or whether it means a maximum of 1 mrep/hr averaged over any 2-in.^{2} area of the garment.

(B) $\approx$1000 counts/min when a Geiger-Mueller counter having a flat plate area of 2 in.^{2} is placed against the garment.

(C) These levels are the same as those for contamination of the skin given in (48-II, 2). If the body is generally contaminated, special efforts should be made to reduce the level. If the body (or clothing) is entirely covered with contamination, it should be reduced to 0.1 mrep/hr regardless of the group of isotopes involved. Note: This appears to establish the target level of cleanliness to be achieved for laundered clothing.

11. HANDBOOK 52 PERMISSIBLE AIR-BORNE CONCENTRATIONS.

(A) (52-E, 3) The Committee recommends that every effort be made to keep the concentration in air and water and in the body to a minimum. *The goal* should be *no* radioactive contamination if it can be accomplished with reasonable effort and expense. *Operating levels should be kept as far below the recommended value as possible.*

(B) (52-A, 1) It is the opinion of this subcommittee that all unnecessary exposure to radioisotopes should be avoided. Note: It is suggested that a safety factor as large as 10 be used in the design and operation of permanent facilities.

(C) (52-TABLE 3) PERMISSIBLE MAXIMUM AIR DUST CONCENTRATION. For continuous exposure: 1.7×10^{-11} μC/ml = 35 d/m/m^3 = 26 μg/m^3. For 40 hr/wk, 49 wk/yr: 105 d/m/m^3 = 78 μg/m^3.

(D) (52-TABLE 2) PERMISSIBLE MAXIMUM AIR CONCENTRATION. When the components are not determined, alpha emitter $=5\times10^{-12}$ μC/ml = 11 d/m/m^3 = 8 μg U/m^3.

DESCRIPTION OF CONTAMINATION SURVEY

In order to determine the contamination level of clothing used in a uranium processing plant, a survey of clothing surface activity was made using a Victoreen 356 alpha survey meter and a Thyac beta-gamma meter. All clothing at Mallinckrodt may be worn by any person, so that a uniform contamination condition is eventually obtained. Clothes used in contaminated and noncontaminated areas are kept separate and washed separately.

A surface contamination study was also made in the laundry and on laundry equipment to determine the equilibrium contamination level. The laundry had been operated $\approx$10 years without a decontamination cleanup at the time of survey.

Air samples were taken in the laundry by drawing air through a filter paper and counting the alpha activity on an alpha scintillation

counter. These data represent the air concentrations likely to be encountered in a laundry handling clothing contaminated with uranium.

RESULTS

The results of the survey are as follows.

A. After Wearing but Before Washing

1. Regulated Coveralls

(a) 70% show visible soil which appears to be uranium materials. Spots range in size from 10 cm^2 to 30% of the total garment.

(b) 80% have one or more 2-$in.^2$ areas reading >3000 c/m with a thin-wall Geiger-Mueller tube.

(c) The estimated average whole-garment level for all coveralls as measured with an air ionization meter was 1.5 mrep/hr with a range of 0.2 to 12.

(d) Only 40% of all coveralls have an estimated average for the entire garment <1 mrep/hr, most of these having one or more spots >1 mrep/hr.

(e) All coveralls show a whole garment average >100 c/m per 2 $in.^2$.

2. Nonregulated Coveralls

(a) 5% show visible "color" of uranium material, but in no case was this considered heavy.

(b) 70% show no spot >1000 c/m.

(c) 20% show an over-all average >1.0 mrep/hr but <3 mrep/hr.

(d) No garment was found with an average <100 c/m.

3. Soft Wear (Underwear, Socks, Handkerchiefs)

(a) Socks show nondetectable to 800 c/m; the average for all socks is estimated at 200 c/m.

(b) Underwear is essentially the same as socks.

(c) Handkerchiefs show a slightly higher average than other soft goods, 10% being visibly contaminated and having spot readings >1000 c/m, but the average for all handkerchiefs is <300 c/m.

4. Bath Towels

Only an occasional detectable reading is found, and no spot is >1000 c/m. The average for all towels is <100 c/m.

5. Gloves

All gloves are heavily contaminated, the average being ≈ 5000 c/m per 2 $in.^2$.

6. Caps

Caps show low to moderate contamination, the average being ≈ 300 c/m.

7. Blue Smocks

These show low to moderate contamination with an occasional spot >1000 c/m, but the average for all garments is ≈ 200 c/m.

8. White Smocks

These are similar to blue but somewhat higher with more spots >1000 c/m. The average for all garments is ≈ 300 c/m.

9. Shoe Covers (Destrehan)

These show high contamination, $<10\%$ giving an average <1000 c/m.

10. Lab Smocks

These read low with occasional spots >1000 c/m. The average is <300 c/m.

B. Typical Laundry Surface Contamination

Table 1 shows typical beta-gamma readings on laundry equipment used ≈ 10 years with clothing containing normal uranium.

Table 1

Surface Contamination in a Laundry Handling Clothing Contaminated With Normal Uranium

Unit	$\beta+\gamma$,* c/m
Surface of presser pad	2000
Surface of cloth hamper	5000
Surface of wooden hamper	1000
Work tables and surfaces	200– 1000
Inside of washer	300
Inside of dryer	1000– 4000
Dryer lint screen	2000
"Handful of lint"	4000
Under dryer, inside housing	100–40,000
Under washer (soap cake)	20,000–60,000
Floors, av	300– 500
Walls	100– 300
Overhead pipes, etc.	300– 1000
Lint trap on roof	3000

*Measured by Thyac beta-gamma meter with thin-wall tube.

C. Air Concentration in Laundry

Table 2 shows typical air concentration of alpha dust in a laundry handling clothing contaminated with normal uranium.

Table 2

Alpha Air Concentration in a Laundry Handling Clothing Contaminated With Normal Uranium

Operation	Concentration, α d/m/m³ Av	Worst
Breathing zone		
Load washer with coveralls	560	820
Load dryer with coveralls	50	140
Unload dryer with coveralls	20	70
Press white coveralls	40	55
Repair white coveralls	60	90
Sorting to wash*	20	870
Weighted average		
Washer operator	50	
Presser	40	
Repairman	40	
Sorting and handling*	30	140
Average general air in laundry**	20	110

*Worst condition obtained with no ventilation. This job is regularly done at a ventilated location to give the average value.

**Worst condition obtained by turning off ventilation fans for 4 hr. Worst general air was taken in the vicinity of handling and loading into washer.

COSTS

A. Equipment

The laundry equipment used at Mallinckrodt consists of the following.

1 Large washer, Champion Cascade, 42×72
1 Small washer, Norwood, 30×30
2 Centrifuges (4-ft-diam and 3-ft-diam)
4 Dryers, American, 36×30
1 Presser, American
1 Hot water surge tank, 1000-gal size

It is estimated that this equipment could be purchased and installed in an existing building for ≈$75,000.

B. Operation

Operating costs at the Mallinckrodt Destrehan laundry during 1957 were estimated, per piece, as follows.

Coveralls	$ 0.19
Towels	.027
Socks, pair	.02
Undershirts	.02
Undershorts	.02

These costs include labor (9 men), laundry supplies, and overhead. They do not include utilities, maintenance, and supporting activities such as purchasing, accounting, health supervision, and porters. The monthly rate was ≈25,000 coveralls, 50,000 towels, and 25,000 pairs of underwear and socks.

Operating costs at the Mallinckrodt Weldon Spring laundry are somewhat less.

Coveralls	$ 0.17
Towels	.03
Socks, pair	.014
Undershirts	.014
Undershorts	.014

These costs include labor (3 men), laundry supplies, overhead, maintenance, and supporting activities such as purchasing and accounting.

Firm bids from outside commercial laundries ranged from $0.30 to $0.50 each on coveralls and $0.02 to $0.05 each on soft goods (undershirts, shorts, and socks). Comparison with the operating costs listed above shows that it is more economical to operate an in-plant laundry. If savings in costs of monitoring and separation by in-plant laundering are included, the payoff period on the initial investment would be between two and three years.

CONCLUSIONS

1. Clothing contaminated with significant amounts of uranium may not be released to public laundries according to the recommendations of the NCRP.

2. Release of clothing to commercial laundries would require some prewashing in most cases and extensive monitoring and separation facilities. Contamination levels would have to be below maximum levels established by the NCRP.

3. When contamination levels are the controlling factor, cost advantage is irrelevant, whether favoring in-plant or outside laundry service. NBS handbook recommendations should be followed.

4. For long-term consideration there is no cost advantage in using outside laundry service. In fact, in Mallinckrodt's operation, there is a definite cost advantage in using an in-plant laundry.

5. Better control of contamination is achieved with an in-plant laundry.

6. No commercial laundries were found that were "licensed" by the AEC to handle uranium and had the necessary ventilation equipment installed to achieve good health conditions.

Experience With Commercial Laundry Operations

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INTRODUCTION

M & C Nuclear is a privately owned and operated nuclear fuel fabrication facility producing reactor fuel in the form of plates, rod, tubing, foil, etc., both clad and unclad, alloyed and unalloyed, from depleted uranium, natural uranium, enriched uranium, thorium, and their oxides. In addition, complete reactor cores are now being fabricated and assembled. Both government contract and licensed operations are carried out in the plant. As a result of this activity, a sizable number of employees working in the 136,506-ft^2 plant require protective clothing which may become contaminated with these materials.

At M & C Nuclear we do not wish to wash clothes unless a definite advantage exists. We were, therefore, pleased to learn of the formation in the fall of 1957 of a company which would launder radioactively contaminated garments. To my knowledge, there are now three such firms in the United States licensed by the AEC. The firm whose services we use is Nuclear Laundry Service, Inc. of Waterbury, Connecticut. In addition to being licensed by the AEC, NLS also has city and state approval of its operation. The service performed includes pickup and delivery of all garments, laundering, and monitoring to the requirements of the customer. NLS carries liability insurance for all phases of their operation outside of our plant as specified by contract. Garments may be either owned by the customer, or rented from NLS at additional cost.

OPERATION OF LAUNDRY SERVICE

At M & C Nuclear, the employees are issued shirts and trousers rather than the more conventional coveralls. Underwear and socks are not provided, although safety shoes are. The garments become contaminated mostly with highly enriched uranium and to a lesser extent with natural uranium, depleted uranium, and thorium. All garments are collected in 55-gal steel drums in the locker room which are subsequently used as shipping containers. Gloves of various types are collected in steel drums located in a different part of the plant.

The drums are sealed, monitored as are all outgoing shipments, and sent to Waterbury by common carrier. All shipments to date have been sent as exempt quantities of radioactive material, which requires that there be $< \approx 19$ g highly enriched uranium per drum. Originally, NLS trucks were to be used for transporting the laundry between Attleboro and Waterbury, but the existence of excellent overnight trucking service made this unnecessary.

The laundry itself is not very different from any other industrial laundry except for a few items such as liquid storage tanks, a protective clothing change point, and some survey and counting equipment. The clothing is washed, extracted, dried, inspected, repaired if necessary, sorted by size, and packed into the original containers now lined with paper to prevent contamination from residual dust. Several randomly selected garments from each wash load are monitored with an alpha survey meter. We are notified if any garments monitored show >500 dis/min/100 cm^2, but we have not requested that the garments be relaundered. It should be mentioned that the monitoring requirements of a customer may vary from zero to 100% inspection depending on the nature of the customer's facility and the type of contamination. The drums of clothing are sealed, monitored, and shipped back to M&C Nuclear. The complete cycle takes about four working days.

COSTS

When M&C Nuclear began using NLS, we already had on hand a large stock of clothing and therefore arranged for laundering only, making replacements and additions to the stock ourselves as necessary. We have recently changed to a rental program. The cost data presented here cover the

period January through June 1958, with allowances made for the addition of new clothing caused by a large increase in the number of employees.

Exclusive of handling and distribution costs and overhead, each set of shirt and trousers or lab coat costs $\approx$\$0.83 to use, at a rate of 3500 sets per month. Under the rental program, each set will cost $\approx$\$0.85, and we will be relieved of the task of inventorying and purchasing garments. It must be emphasized that these costs are specific for M&C Nuclear and would not necessarily apply to any other installation because of possible differences in transportation charges, quality and quantity of garments used, monitoring requirements, etc.

While these costs do not represent the entire operating costs, as mentioned previously, they should be of value for purposes of comparison with plant operated laundries.

HEALTH PROTECTION

By contract, NLS is liable for any damages which may arise from the time contaminated clothing leaves our door until the cleaned clothing is returned to our door; therefore, health protection in the laundry is not our responsibility. It is difficult for us to interpret the various health protection measures taken at NLS, since they launder many customers' garments contaminated with natural uranium, highly enriched uranium, fission products, and other isotopes, all in varying amounts.

Urinalyses of NLS employees are made every three months for uranium alpha and gross beta-gamma activity. Air samples are collected during various operations, with specific attention to the loading of the washing machines, and counted for both alpha and beta-gamma activity. Floor and other surface contamination surveys are made on a routine basis. In over a year of operation, no results $>10\%$ of maximum permissible levels have been obtained. Since the levels of contamination on our garments are not measured before laundering, no correlation between internal exposures and clothing contamination could readily be established. Such correlations have been attempted before by others.

Wash water from the laundry is held in a large underground tank. Prior to discharge, the water is pumped to an above ground tank, and a continuous sample is withdrawn, evaporated to dryness, and counted for both alpha and beta-gamma activity. All samples analyzed to date have been below the dumping tolerance level.

PUBLIC RELATIONS

The problem of public relations has been negligible. Few people outside M&C Nuclear have reason to know that contaminated clothing is sent out to be laundered. Those that do know accept the fact without question. No concern exists on the part of the common carrier, M&C Nuclear, or NLS. If the AEC can convince people that no hazard exists from dropping and exploding unarmed nuclear weapons containing many kilograms of uranium, we should have no trouble with a few drums of clothing containing a few milligrams of uranium.

ECONOMICS OF URANIUM HANDLING

As mentioned previously, the wash water from the laundry has been found to contain little activity; the amount is below the nonoccupational level for any radioisotope in drinking water and is not economically recoverable. Two drums of sludge recently scraped off the bottom of the underground holding tank were found to contain significant amounts of uranium. These findings are classified; however, this much can be said: assuming that all the uranium came from M&C Nuclear, which it did not, the amount recoverable would not, by any stretch of the imagination, justify the construction and operation of a plant laundry.

COMPARISON WITH A PLANT OPERATED LAUNDRY

On the basis of comments made by other contractors, including one of the national laboratories, it appears that the cost of operating plant laundries is higher than that of operating commercial laundries. At least one contractor of moderate size is now using the services of a commercial laundry in preference to its own existing laundry facility.

A study made by M&C Nuclear last fall showed little difference between in-plant and outside laundering, at least on paper. The things which did not show on paper, however, were:

1. The plant laundry was predicated on the existence of a liquid waste disposal system which

was planned but did not and still does not exist. This represents a sizable capital investment.

2. We did not know anything about running a laundry.

3. We did not want to be in the laundry business.

Since we began using the services of a commercial laundry, we have been completely satisfied. Under the newly adopted rental plan, our work has been even further reduced and our problems are truly minimal.

Evaluation of Environmental Uranium Contamination at the Feed Materials Production Center

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The processing of normal uranium with its extremely low specific activity ($\approx 7\times10^{-7}$ curies/g) does not suggest a significant health problem when compared with that of radium or some of the longer-lived fission products. The possibility of renal damage due to its chemical toxicity, however, justifies considerable effort to control uranium and its associated decay products. At the AEC Feed Materials Production Center operated by National Lead Company of Ohio at Fernald, Ohio, the emphasis has been upon engineering into process operations a high degree of control of radioactive materials in order to keep to a practical minimum the release of such materials to the total environment.

PLANT LIQUID EFFLUENT

The total plant liquid effluent varies between 1 and 2 million gal/day. The combined effluent stream, carrying industrial, storm, and treated sanitary sewer effluents, is sampled continuously. Composite samples are analyzed daily for chemical and radioactive content. Table 1 shows the uranium concentration and the total alpha and total beta activities of the plant effluent for the first half of 1958 as sampled daily before release to the Miami River.

The highest uranium concentration, sampled through a 24-hr period, was 1.0×10^{-5} μC/cc. The over-all average for the 7-month period was 1.3×10^{-6} μC/cc. The recommended maximum permissible concentration[1] for natural uranium (soluble) in water for continuous exposure is 7×10^{-5} μC/cc. As an AEC facility, FMPC is governed by the recent Chapter 0524 of the *AEC Manual*[3] which limits radioactive liquid effluent released to the environment to a uranium concentration of 2×10^{-5} μC/cc (assuming the worst possible condition of no dilution whatever). The uranium concentration in the plant effluent is within the limits for human consumption.

The highest total alpha activity in the plant effluent, averaged over a 24-hr period, was 1.5×10^{-4} μC/cc. The over-all average for the period January through July was 5.7×10^{-6} μC/cc. Similarly, the highest beta activity was 2.5×10^{-3} μC/cc, and the over-all average was 5.2×10^{-5} μC/cc. If it is assumed that virtually all the beta activity is from the immediate daughters of U^{238} (Th^{234} and Pa^{234}; see Table 2), then these beta results can be compared with the MPC for this combination,[2] which is 5×10^{-2} μC/cc.

The obvious conclusion from these data is that the uranium concentrations and the Th^{234} plus Pa^{234} activities in the plant effluent waste stream to the river do not exceed the recommended MPC's for the respective radioisotopes in drinking water for continuous exposure. The large dilution (by a factor of $\approx$2000) afforded by the Miami River is therefore an unimportant asset.

The combined plant effluent is discharged via pipeline into the Great Miami River $\approx$1 mile east of the project. River water samples are taken semimonthly at the Venice and New Baltimore bridges, respectively above and below the plant effluent outfall. Each sample point is $\approx$2 miles distant from the outfall point. These river samples are taken jointly with Ohio State Health Department personnel. The samples are split and then analyzed both by the State Health Department and by our Health and Safety Analytical Laboratory, and the results are compared. Table 3 shows Miami River sample data for the first half of 1958.

These data indicate that the uranium, alpha, and beta concentrations below the plant effluent outfall point are lower than the plant effluent concentrations by two or three orders of magnitude. Why the river samples above the plant effluent outfall point sometimes show slightly higher con-

Table 1

FMPC Combined Liquid Effluent Data

1958		U, μC/cc	Total α, μC/cc	Total β, μC/cc	Effluent volume, gal×10³/day	River flow, gal ×10⁶/day
Jan.	High	5.6×10^{-6}	2.2×10^{-5}	7.0×10^{-4}	2564	8337
	Av	1.1×10^{-6}	4.1×10^{-6}	5.4×10^{-5}	2142	2537
Feb.	High	2.7×10^{-6}	1.5×10^{-4}	2.5×10^{-3}	2375	3361
	Av	1.1×10^{-6}	1.4×10^{-5}	1.1×10^{-4}	2150	1510
March	High	4.9×10^{-6}	1.7×10^{-5}	5.3×10^{-5}	2286	5687
	Av	1.2×10^{-6}	3.6×10^{-6}	1.1×10^{-5}	1686	2075
April	High	1.0×10^{-5}	4.5×10^{-5}	3.3×10^{-4}	1995	7691
	Av	1.8×10^{-6}	4.6×10^{-6}	2.7×10^{-5}	1573	3021
May	High	6.9×10^{-6}	3.8×10^{-5}	5.7×10^{-4}	1770	9048
	Av	1.4×10^{-6}	4.1×10^{-6}	4.3×10^{-5}	1347	3613
June	High	3.8×10^{-6}	4.8×10^{-5}	1.4×10^{-3}	1490	9694
	Av	1.0×10^{-6}	3.6×10^{-6}	6.5×10^{-5}	1086	6321
July	High	5.0×10^{-6}	7.9×10^{-5}	3.2×10^{-4}	1862	13894
	Av	1.7×10^{-6}	1.3×10^{-5}	4.0×10^{-5}	1307	5367

centrations or higher activities than samples below is unexplained. Probably at these low levels analytical accuracy is minimal.

Water samples are collected three times daily from a small surface stream called Paddy's Run, which drains the west side of the project. These samples are composited every third day and analyzed. This stream is of concern because occasionally a waste chemical pit is decanted into it for a short period when the discharge line to the river is not available. Table 4 shows the monthly averages of samples taken at a point near where the stream leaves the project. The highest uranium concentration, averaged over a month's time, was 1.2×10^{-7} μC/cc. The highest 3-day composite (not shown in Table 3) had a uranium concentration of 6.2×10^{-7} μC/cc. On the basis of the above data, average uranium concentrations (6.5×10^{-8} μC/cc) and total alpha (1.4×10^{-7} μC/cc) and total beta activities (5.5×10^{-7} μC/cc) in Paddy's Run are not exceeding recommended MPC's. Table 5 shows Paddy's Run water data upstream from the propect.

It is true that concentrations of contaminants in industrial effluents will vary from time to time unless they are controlled by elaborate retention and dilution systems. Though our sample analyses are usually 24 hr or more "after-the-fact," we have not seen significant variations in effluent radioactivities, and we have seldom had occasion to restrict the release of liquid effluent to the off-site environment. The few such occasions were predictable, and each time controlled plant operations prevented excessive radioactivity discharge. It is not felt that the plant is taking advantage of the averaging of sample collection and data to permit, as it were, the occasional release of large quantities of radioactive or chemical contaminants.

Three deep wells supply water for the plant. These wells are sampled and analyzed monthly for uranium and total alpha. Table 6 presents well water data for the first half of 1958. The uranium concentrations and alpha activities of the well water may be considered as background (naturally occurring concentrations), and are of the same order of magnitude as those in the Miami River water and in Paddy's Run.

Table 2

			Immediate daughter	
Isotope	% Abundance	% Activity	Emitter	Half-life
U^{238}	99.28	49	β	24 days
U^{235}	0.715	2	β	25 hr
U^{234}	0.006	49	α	10^5 yr

Table 3

Miami River Water Data in μC/cc

	U		α		β	
1958	A*	B	A	B	A	B
Jan.	9.8×10^{-9}	3.4×10^{-8}	n.d.	3.2×10^{-8}	2.0×10^{-6}	1.6×10^{-6}
Feb.	4.2×10^{-9}	1.8×10^{-8}	3.6×10^{-8}	n.d.	n.d.	n.d.
March	2.1×10^{-9}	6.3×10^{-9}	4.6×10^{-8}	4.1×10^{-8}	1.6×10^{-7}	4.1×10^{-7}
April	5.6×10^{-9}	9.1×10^{-9}	2.5×10^{-7}	8.6×10^{-8}	5.4×10^{-8}	2.7×10^{-8}
May	7.0×10^{-9}	9.1×10^{-9}	2.7×10^{-7}	1.5×10^{-7}	2.3×10^{-7}	1.6×10^{-7}
June	n.d.	2.1×10^{-9}	4.6×10^{-8}	5.9×10^{-8}	5.9×10^{-8}	5.0×10^{-8}
July	4.2×10^{-9}	5.6×10^{-9}	8.6×10^{-8}	6.8×10^{-8}	6.4×10^{-8}	7.7×10^{-8}
Aug.	4.2×10^{-9}	7.7×10^{-9}	1.8×10^{-8}	2.3×10^{-8}	7.7×10^{-8}	7.7×10^{-8}

*A and B indicate above and below the plant site.
n.d. = none detectable.

Table 4

Paddy's Run Water Data in μC/cc
Downstream From Project Boundary

1958	U	Total α	Total β
Jan.	5.8×10^{-8}	1.3×10^{-7}	8.5×10^{-7}
Feb.	3.6×10^{-8}	1.2×10^{-7}	7.1×10^{-7}
March	8.9×10^{-8}	2.0×10^{-7}	2.2×10^{-7}
April	6.2×10^{-8}	1.5×10^{-7}	2.0×10^{-7}
May	7.2×10^{-8}	1.3×10^{-7}	6.3×10^{-7}
June	1.5×10^{-8}	5.9×10^{-8}	6.3×10^{-7}
July	1.2×10^{-7}	2.1×10^{-7}	6.2×10^{-7}
Av	6.5×10^{-8}	1.4×10^{-7}	5.5×10^{-7}

Table 5

Paddy's Run Water Data in μC/cc
Upstream From Project Boundary

1958	U	Total α	Total β
Feb.	9.7×10^{-9}	4.0×10^{-8}	n.d.
May	1.9×10^{-8}	1.0×10^{-8}	7.9×10^{-7}
June	8.5×10^{-9}	6.5×10^{-8}	6.3×10^{-8}
July	3.9×10^{-8}	1.1×10^{-7}	4.5×10^{-8}
Aug.	6.8×10^{-10}	2.1×10^{-8}	n.d.
Sept.	1.4×10^{-9}	4.0×10^{-8}	1.5×10^{-8}
Av	6.8×10^{-9}	5×10^{-8}	2×10^{-7}

Table 6

FMPC Well Water Data in μC/cc

	Well #1		Well #2		Well #3		Averages	
1958	U	α	U	α	U	α	U	α
Jan.	1.4×10^{-9}	n.d.	4.2×10^{-9}	n.d.	4.2×10^{-9}	n.d.	3.5×10^{-9}	n.d.
Feb.	7.0×10^{-9}	n.d.	1.7×10^{-8}	n.d.	5.6×10^{-9}	n.d.	9.8×10^{-9}	n.d.
March	7.0×10^{-9}	n.d.	7.7×10^{-9}	n.d.	2.8×10^{-9}	n.d.	5.6×10^{-9}	n.d.
April	5.6×10^{-9}	n.d.	1.4×10^{-9}	n.d.	1.4×10^{-9}	3.6×10^{-8}	2.8×10^{-9}	1.4×10^{-8}
May	7.0×10^{-10}	2.3×10^{-8}	4.9×10^{-9}	n.d.	6.3×10^{-9}	n.d.	4.2×10^{-9}	9.1×10^{-9}
June	1.4×10^{-9}	9.6×10^{-8}	n.d.	n.d.	1.4×10^{-9}	1.2×10^{-7}	1.4×10^{-9}	7.3×10^{-8}
July	n.d	n.d.	2.1×10^{-9}	n.d.	8.4×10^{-9}	4.1×10^{-8}	3.5×10^{-9}	1.4×10^{-8}
Aug.	6.3×10^{-9}	5.9×10^{-8}	5.6×10^{-9}	1.4×10^{-8}	3.5×10^{-9}	1.2×10^{-7}	4.9×10^{-9}	6.4×10^{-8}

n.d = none detectable.

PLANT AIR-BORNE EFFLUENT

Evaluation of air-borne uranium concentrations at FMPC is done by intermittent high-volume air sampling (1.5 m^3 air/min) and by gummed-paper fallout collectors. Since normal uranium processing seems to lack any major hazard or catastrophy potential, continuous air sampling procedures and equipment have not been considered necessary.

Table 7 shows air sample data from eight locations along the security fence surrounding the production area of the project (about 1200 ft from the center). The uranium concentrations and total alpha and total beta activities given are monthly averages of data from all the stations. The original data (not given in Table 7) show that the individual high uranium concentration was 2.5×10^{-12} μC/cc air. The high for total alpha activity was 1.4×10^{-12} μC/cc and for total beta, 3.5×10^{-12} μC/cc. The samples were taken for one to three days at a rate of $\simeq$1.5 m^3/min. The over-all average uranium concentration in air at the production area perimeter was 1.9×10^{-13} μC/cc; alpha activity, 3.1×10^{-13} μC/cc; and beta activity, 9.1×10^{-13} μC/cc. The averages in Table 7 differ slightly from the weighted averages because dur-

Table 7

FMPC Perimeter (Production Area) Air Sample Data (Eight-Station Averages)

	U, μC/cc	α, μC/cc	β, μC/cc
1957			
Oct.	4.0×10^{-13}	1.4×10^{-13}	No data
1958			
Feb.	5.1×10^{-14}	6.4×10^{-13}	1.4×10^{-13}
March	2.8×10^{-13}	3.8×10^{-13}	5.0×10^{-13}
June	1.1×10^{-13}	1.1×10^{-13}	2.1×10^{-12}
July	1.8×10^{-13}	8.8×10^{-14}	7.5×10^{-13}
Aug.	1.6×10^{-13}	4.8×10^{-13}	1.1×10^{-12}

Table 8

Off-Site Air Sample Data (1958)

		U, μC/cc air	
Location	No. of samples	Individual high	Average
Venice area	6	5.0×10^{-13}	1.4×10^{-13}
North of plant, Ohio 126	8	4.6×10^{-13}	3.2×10^{-13}
East of plant, Ohio 128	2	7.0×10^{-14}	7.0×10^{-14}
South of plant, Willey Road	6	2.8×10^{-13}	1.3×10^{-13}
New Baltimore area	2	7.0×10^{-14}	7.0×10^{-14}
West of plant, Atherton Road	2	2.8×10^{-13}	1.4×10^{-13}

Table 9

FMPC Gummed-Paper Fallout Data

	U, μg/ft^2/day			
1958	Group I	Group II	Group III	Group IV
Jan.	455	104	31	6
Feb.	700	163	21	12
March	485	96	18	14
April	460	103	54	23
May	209	63	20	11
June	339	111	52	17
Av	441	107	33	14

Table 10

FMPC Gummed-Paper Fallout Data

	U, μg/ft^2/day	
1958	Highest single collection	Weighted monthly av, all stations
Jan.	641	104
Feb.	1714	162
March	842	109
April	586	114
May	321	61
June	406	99

Table 11

Incinerator Area Soil Data

Sample No.	U, μg/g soil
1	425
2	54
3	186
4	155
5	496
6	155
7	36
8	17

ing some months samples were taken at only four of the eight stations.

Off-site air samples were taken infrequently during the first half of 1958. Sampling points were from 1 to 3 miles from the project boundary. The results of these surveys are summarized in Table 8, which is self-explanatory. The recommended MPC for uranium in air for continuous exposure[1] is 1.7×10^{-11} μC/cc, and for AEC-released effluent to the environment[3] the MPC is 5×10^{-12} μC/cc.

Twenty-four gummed-paper stations are located on a rough grid over the project area. The papers are collected and analyzed monthly for uranium. Table 9 shows the data for the first half of 1958. Group I fallout stands are located from 250 to 800 ft from the center of the production area; Group II, 1100 to 1800 ft; Group III, 2400 to 2900 ft; and Group IV, 3200 to 6200 ft. Table 10 shows the highest uranium collection on a single fallout stand (month's collection), expressed as $\mu g/ft^2/day$, and also the monthly averages of all the stations.

The only soil survey data available are from a special study done in May 1957. This survey covered the vicinity of the combustible waste incinerator at varying distances up to 300 ft from the stack. The data are shown in Table 11. Without data on pre-operational uranium concentrations in the soil it is difficult to evaluate these findings. Further soil sample data are needed before any conclusions can be drawn.

In summary, we feel that there is no significant uranium contamination in the plant or the off-site environment, significant amounts being taken to mean amounts approaching maximum permissible levels. This is due not to chance but to the engineering of controls into the process. Only a small part of the cost of the liquid effluent treatment can be related directly to radioactive contaminants, the major part being due to the usual industrial chemical wastes that result from the operation of any chemical plant. Furthermore, in our experience, dust collection systems, besides holding air contamination to a minimum, pay for themselves by the recovery of uranium.

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Environmental Contamination

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INTRODUCTION

Facilities for the handling and processing of tonnage quantities of uraniferous materials have been in continuous operation since 1942. Uranium materials have been discharged continuously to the environs at these plant sites. Studies have been directed by the Health and Safety Laboratory (HASL) toward assessment of short-term and long-term effects of these plant effluents on contamination of air, surface water, and soil. Facilities investigated include Lake Ontario Ordnance Works, Middlesex Sampling Plant, Harshaw Chemical Company, Mallinckrodt Chemical Works, and many milling plants in the Colorado plateau area.

METHODS OF STUDY

Air Pollution[1]

Two different sampling techniques were used in making the surveys, stationary and mobile. With stationary sampling, continuous or intermittent samples were taken at the same locations over an appreciable length of time. With mobile sampling, samples were taken at different distances upwind, cross-wind, and downwind. The locations of sampling stations depended on wind direction and other weather conditions at the time of sampling. Stack sampling was synchronized with sampling at mobile and fixed stations. Air dust samples were collected by drawing air through 1⅛-in.-diameter Whatman #41 filter discs with standard sampling equipment and techniques normally employed by HASL. Radioactive dust collected on filter discs was analyzed for alpha activity on scintillation counters.

Surface Water[2]

Duplicate samples of water and mud were collected at selected points, the former to provide an instantaneous view of stream quality and the latter to indicate the average condition. Uranium was determined in both water and mud samples by photofluorimetric analysis.

Soil[3]

Duplicate soil samples from the surface and subsurface were obtained by conventional soil sampling techniques along equally spaced radii at selected distances from the site being investigated. Studies[4] were made to determine the long-term accumulation of uranium in soils surrounding processing sites in order to determine the concentration gradient of uranium residing in the soil with respect to depth, distance, and direction from the source of pollution.

DISCUSSION OF RESULTS

Soil, Water, and Mud

Although no standards exist for uranium contamination in soil and mud,[5] a conservative maximum can be estimated.[2] Ten CFR Part 20[6] specifies maximum allowable concentrations in water above natural background for radioactive materials released into water in unrestricted areas equivalent to 10.4 μg U per g water. The natural soil uranium as reported by D.E. Lynch[4] is 3 to 9 μg/g. It has been suggested[4] that the permissible concentration level for soil might safely be set at 100 times the value for water, i.e., 1040 μg U per g soil (ppm).

Results[4] of a soil and water uranium survey conducted in 1949 are listed in Table 1. Few points, if any, outside the site boundaries were found to be contaminated.

Soil[3] sample surveys were conducted at three AEC facilities during 1951 to 1954 to determine the concentration gradients of uranium in the soil with respect to depth, distance, and direction from the source of pollution. Survey results are listed in Table 2 for comparison. The soil concentration in each case decreased linearly with distance and was

Table 1

Soil and Water Uranium Survey Results (1949) in μg/g

Medium	Sample location	Lake Ontario Ordnance Works	Middlesex Sampling Plant	Harshaw Chemical Company
Soil	Background	4.8	3.6	7.8
	Property boundary	8.2	170	265*
				7.1**
	1000-ft circle		107.8	
	8000-ft circle	4.8		
	Distant points		3.6	7.8**
Mud	Background	4.4	3.0	3.4
	Ditch	12.2		
	Branch		16.0	
	Upstream	4.4	3.0	3.4
	Downstream	6.1	2.6	3.5
Water	Background		0.0025	0.001
	Ditch	0.22		
	Branch		0.005	
	Sewer outlet			0.86
	Upstream		0.002	<0.001
	Downstream	0.047	0.003	0.001

*Top ¼ inch.
**¼ in. to 6 in.

Table 2

Comparison of Results of Soil Surveys (1951 to 1954)

Item	Middlesex Sampling Plant	Mallinckrodt Chemical Works	Harshaw Chemical Company
Years of operation	7	13	11
Average annual rainfall (in./yr)	45	38	38
Estimated U in stack discharge since start of operation (tons)	47–45	18.5	52.3
Area sampled (mi^2)	2	2	2
Soil content (μg/g): Upper soil, 0 to 1 in.	4.3	3.0	0.1
Lower soil, 1 to 6 in.	2.3	1.0	0.1
Tons U in total soil, 0 to 6 in.	16	8	0.6
Tons U in soil per ton in stack effluent	0.32	0.40	0.012

indistinguishable from the local background beyond about 2000 ft from the center of plant activities. The material from both Middlesex Sampling Plant[3] and Mallinckrodt Chemical Works was found to remain in the soil to the extent of between 30 and 40% of the estimated initial fallout. This would indicate that where insoluble materials were discharged, there was relatively small reduction in the soil content due to weathering. Also, most of the material appeared to remain in the upper portion of the soil. About 2 to 3 times as much was found in the first inch as was in the next five. Harshaw Chemical Company, which discharged almost entirely a soluble form of uranium (UO_2F_2), showed markedly different results. Here the residual concentration in the soil was

Table 3

Environmental Pollution Surveys (1956 to 1958) of Plant Effluents
Figures are average uranium concentrations in water in $\mu\mu$C/ml.

Mill	Upstream	Plant effluent	Downstream	
			1 mile	3 miles or more
A	–	–	–	–
B	0.004	0.0015	0.002	0.001
C	0.0017	6.0	0.021	0.013
D	0.007	0.0064	0.0019	0.0064
E	–	–	–	–
F	–	–	–	–
G	0.0017	0.002	0.0013	0.0017
H	0.0011	0.0011	0.00027	0.00017
I	0.0058	0.297*	0.0064	0.0061
J	–	–	–	–
K	0.002	0.095**	0.006	–
L	–	–	–	–

*Only seepage from tailings ponds.
**Mostly insolubles as slurry.

estimated to be $\approx$1.2%. The soluble material also differed in that there was no significant difference between the concentration in the upper inch and in the next five inches.

A superficial study[7] was made during 1956 to 1958 of the contribution of plant effluents from 12 uranium concentrating mills to local surface waters. Samples from the local streams were taken along the banks on the plant side. No attempt was made to obtain samples across the width of the stream nor through its depth. Samples were taken upstream as well as downstream to obtain background levels. Data were also obtained, where possible, directly from the effluent flow. Table 3 shows these data. Ten CFR Part 20 specifies a concentration of 7×10^{-6} μC uranium per ml water for continuous exposure. It can be seen that the uranium contribution was negligible. Table 4 shows concentrations in soil samples obtained concurrently with the water samples. Here it can be seen that there was a significant accumulation of uranium in the soil which would tend to deplete the contribution to the surface waters.

Air

Results of an air pollution survey[8] conducted in the vicinity of the Harshaw plant during June 1949 are shown in Figure 1. The data presented were standardized to a uniform wind velocity of 3 mph. During the survey the average rate of uranium emission from the plant stacks was 0.097 g/sec. Ten CFR Part 20 specifies a concentration of 1.7×10^{-12} μC U per ml air for continuous exposure, which corresponds to 2.5 μg/m^3. Few of the reported concentrations were above maximum permissible levels. Application of Sutton's equation[9] to data plotted in Figure 1 indicates a maximum air-borne concentration with a 3-mph wind of 42 μg/m^3 at about 500 ft. The actual value found was $\approx$10 μg/m^3, with the MAC occurring at about 1000 ft.

Results of an air pollution survey[10] conducted in the neighborhood of the Mallinckrodt Chemical Works during November 1949 are shown in Figure 2. All concentrations plotted were standardized for unit wind velocity. Although no stack samples were taken, data[11] accumulated previously indicated a probable average rate of emission of uranium from the plant stacks of about 0.011 g/sec. Nearly every reported concentration was below maximum permissible levels. Here, too, 1000 ft would circumscribe the MAC.

Economic Aspects

As shown in Table 2, Middlesex Sampling Plant, Mallinckrodt Chemical Works, and Harshaw Chemical Company have been in operation for several years and have discharged 50, 18.5,

Table 4

Environmental Pollution Surveys (1956 to 1958) of Plant Effluents
Figures are uranium concentrations in stream muds and sediments in μg/g.

Mill	Upstream	In discharge canal	Downstream	
			1 mile	3 miles or more
A	–	–	–	–
B	–	–	–	–
C	0.5	250	12	5.5
D	3.0	1.8	1.5	2.3
E	–	–	–	–
F	–	–	–	–
G	0.38	61.8	3.5	3.5
H	4	210	1.2	1.1
I	0.8	38	1.8	1.5
J	–	–	–	–
K	5.0	128	9	–
L	–	–	–	–

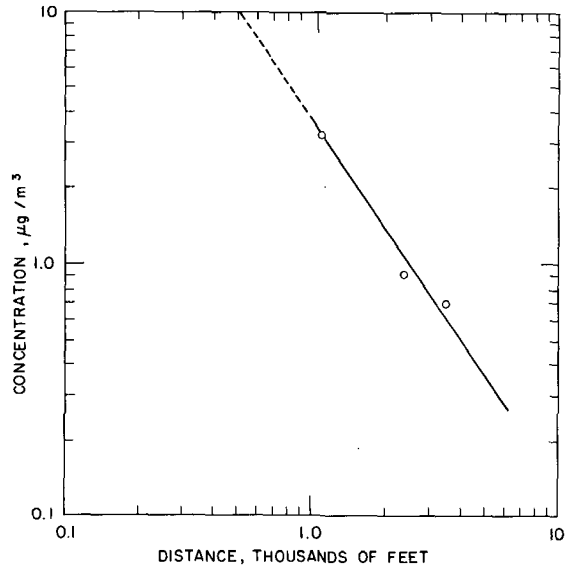

Figure 1. Air pollution survey in the vicinity of the Harshaw Chemical Company (1949). Fall-off of maximum uranium concentration with distance, standardized to a uniform wind velocity of 3 mph.

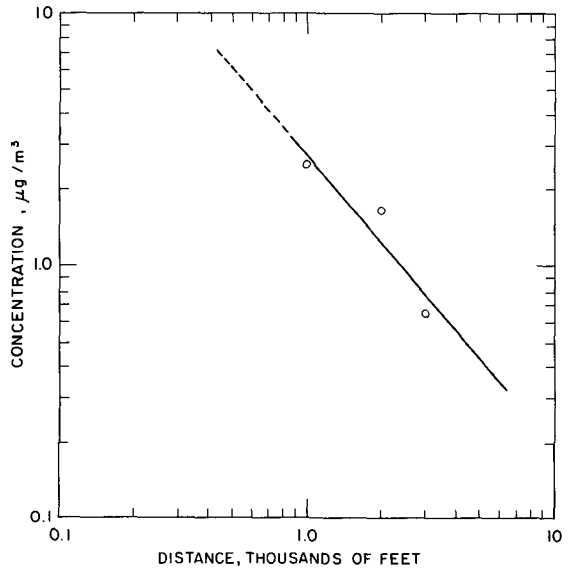

Figure 2. Air pollution survey at Mallinckrodt Chemical Works, Plant 4 (1950). Fall-off of uranium concentration with distance, standardized to unit wind velocity.

and 52 tons respectively of uranium as metal in stack effluents. Based on 1956 costs,[12] these losses represent ≈$500,000, $430,000, and $1,230,000, or a grand total of ≈$2,160,000. It is therefore useful at this point to examine the economic aspects of the MAC for off-site air and water as specified by 10 CFR Part 20 in light of the data that have been collected.

At the Harshaw plant, the discharge rate of metal was 0.097 g/sec, or, for a 24-hr/day operation, ≈3600 kg/year as recoverable oxide. At an assumed value of $10 per pound, this represents a loss of ≈$80,000 per year. The total air moved was about 80,000 cfm. The installation[13] of air cleaning equipment with an efficiency of 99% would have resulted in an annual saving of $79,000 to be balanced against an installation cost of a maximum of about $100,000 and an annual operating and maintenance cost of about $13,000, or a 1½-year write-off.

Similarly, at the Mallinckrodt Chemical Works, where the effluent was about 12% and the air quantity about 25% of the above, the saving could have been $9600 per year, with an installation cost of ≈$25,000 and an operating and maintenance cost of $3200, which would yield a 4-year write-off.

Thus it can be seen that at two plants from which the maximum ground level concentration did not exceed the MAC at 1000 ft, with a probable average concentration below MAC at 500 ft, it would have been very profitable to install air cleaning equipment to reduce these values by 99%.

With stream pollution the situation is generally similar. The average flow in a very small stream is 450 gal/min or 1 cfs, and the permissible concentration of uranium in drinking water is 10 ppm. The annual loss required to average 10 ppm in a stream with 1-cfs flow is 10 tons, or about $200,000 per year.

SUMMARY

Results of environmental pollution studies conducted by HASL at natural uranium processing facilities indicate little, if any, contamination of soil, water, or air. Estimates indicate a loss of about $2,000,000 worth of uranium in stack effluents at three major facilities over a 14-year period. Examination of some of the economic aspects of environmental pollution control indicates the practicality of such measures.

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Discussion of Session IV Papers

Chairman, D.M. Ross

QUESTION: What is the limit on disposing of material into a stream?

WEINSTEIN: I believe I mentioned previously that CFR Part 20 gives a limit of 7×10^{-6}.

QUESTION: How confident are you on the estimates of the total stack emissions?

WEINSTEIN: As confident as the data permit. We made a detailed study of this matter, which is reported in our paper on soil contamination, and we used whatever figures were available from these plants.

QUESTION: How did they arrive at the figures? Are these average concentrations?

WEINSTEIN: They did stack sampling at various times in these plants.

QUESTION: What allowable surface contamination limits are applied to equipment resold for unlimited use?

UTNAGE: The AEC has established legal limits. I think the I.C.C. regulation for release to private parties requires surface contamination <500 alpha dis/min/cm^2. In our paper we dealt with actual working areas rather than with articles to be released.

QUESTION: At what concentrations do you require respiratory protection for routine operations?

UTNAGE: Our approach is perhaps different from that of many laboratories in this respect. We try to engineer our operations so that the concentration is <1 MAC. This does not necessarily mean that if it is >1 MAC for a specific operation we immediately require the operator to wear a respirator. This would depend on his integrated exposure, taking all operations into consideration. However, if we do find some short-term high alpha concentrations in the air, we make it a practice to have personnel wear respirators temporarily until the situation is corrected. We do not subscribe to the use of respirators as standard control equipment.

BROBST: In deference to Mr. Utnage, while it is true that there is probably little correlation between air and surface contamination in plant operations, I think one of the reasons he may have found so little correlation is because the surfaces were vacuumed before the surface contamination was measured, and thus, the activity built up over a period of time was compared to air activity measured on an instantaneous basis.

I have another comment on scrap. It is all right to decide that a certain amount of contamination is allowed on scrap going out, but a sharp differentiation must be made between the kinds of contamination. It is one thing to send out material that has fixed contamination, such as jacketing material, fuel plates, etc., from which the contamination cannot be removed; it is another thing to send out to a junk dealer material that has any transferable contamination on it at all which can be spread around. I am sure you are all aware of the problem with transferable contamination in Houston, Texas. If it is fixed, it is of very little concern; but if even $\frac{1}{10}$ of it is transferable, it can be very mobile and can lead to some complicated legal situations.

Another point in discussing surface contamination: many contractors tell us that we accuse them of having a "dirty" plant, whereas they know of other plants where more uranium is spread around. One of the things they forget is that another plant may be working with natural uranium while they are working with enriched uranium with a specific activity perhaps 100 times as high. Also, one plant may be able to handle large amounts of enriched material and not have much activity, while another may handle a small amount and have a great deal of activity.

SESSION V

Beta Exposure During Uranium Processing

R.C. Baker
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This report concerns the beta radiation dose received by personnel employed in the operation and maintenance of UF_6 manufacturing, uranium isotopic separation, and uranium metal casting facilities. The sources of exposure are the beta-emitting daughter products of U^{238} and small quantities of fission products remaining in decontaminated spent reactor fuels. Although exposure to beta radiation is usually considered secondary to uranium toxicity and even to alpha radiation problems, in these operations and processes beta-emitting impurities are concentrated to a high degree, and beta radiation exposure and monitoring present the most significant and difficult problems. First the processes and equipment in which significant quantities of beta emitters accumulate will be described, and then how the exposure occurs and how it is monitored.

SOURCES OF BETA RADIATION

In the radioactive decay scheme of normal uranium an alpha-particle is emitted by both U^{238} and U^{234}, and a beta-particle is emitted by both Th^{234} and Pa^{234}. The specific disintegration rate is 1500 alpha and 1500 beta dis/min/mg U. This has been shown to produce about 240 mrad/hr at the surface of uranium metal, about 208 mrad/hr at the surface of UO_3, and 183 mrad/hr at the surface of UF_4. During the preparation of our UO_3 by our suppliers, much of the beta-active material is removed, but by the time the uranium enters UF_6 production facilities, the beta activity is from 50 to 100% of the equilibrium value. The beta activity of the reactor tails material ranges from 60 to 150% of the equilibrium value for normal uranium.

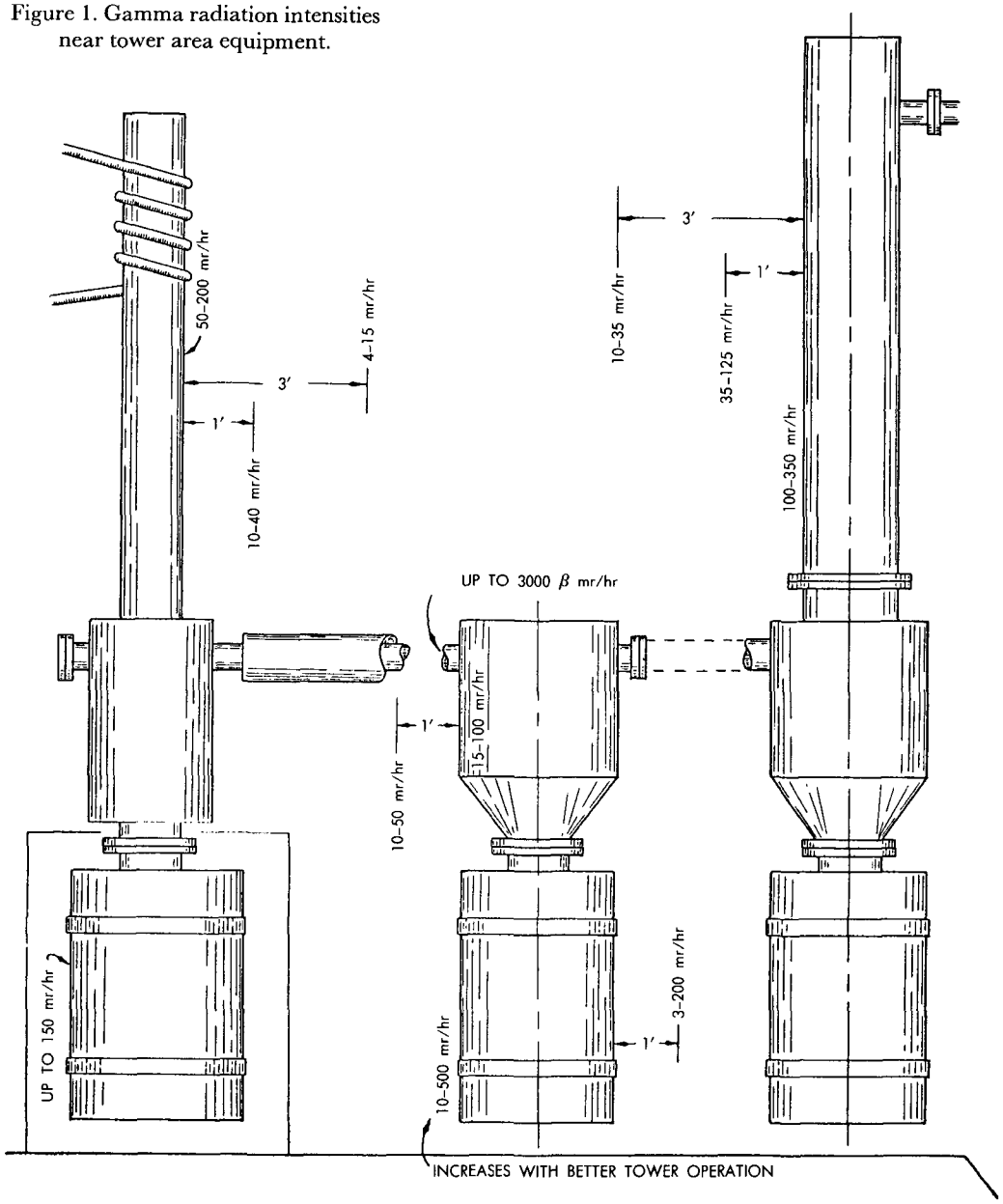
UF_6 Manufacturing

The decontamination of reactor tails is remarkable. The results of analyses at Hanford and at Savannah River show the beta activity of decontaminated reactor tails to be a small fraction of, and the gamma activity about equal to, the normal uranium equilibrium value. These activities increase during shipment and storage to about 100% of the equilibrium beta and 200% of the equilibrium gamma activity.

During the processing of this material little, if any, change in activity is noted until the fluorination of UF_4 to UF_6. The UF_6 formed is gaseous and flows on through the system. Some of the UF_4 passes through a molten state and does not react but falls out in the fluorination tower ash receiver with other solid fluorides of feed impurities. The velocity of the gas stream is sufficient to transport a significant fraction of solids through the gas cooler lines, cyclone separators, and dust collectors to a sintered metal filter. The solid particles are unreacted UF_4, fluorides of thorium, long-lived fission products, and metal impurities such as iron, sodium, calcium, nickel, and copper. Figure 1 shows the equipment involved and some typical radiation readings. To a large degree the efficiency of UF_4 conversion affects the intensities read. If the conversion efficiency is low, the unreacted uranium provides a good shield for gamma radiation, and the specific beta activity of spilled material is low. However, in the past two years improvements in production techniques have outstripped that in the ash handling system. A routine conversion efficiency $>99.5\%$ has been reported. Since little beta activity is carried beyond the system shown, this conversion produces almost a 200-fold over-all increase in specific beta activity. This increase is not uniform among the dust collectors; the material in the third dust collector is about 15 times as active as that in the first. Typical beta activities are as follows:

	Activity, mC/g	
Sample source	Normal U line	Reactor tails line
Tower ash receiver	0.002	0.002
First cyclone receiver	0.015	0.015
Second cyclone receiver	0.010	0.100

Figure 1. Gamma radiation intensities near tower area equipment.



This activity is made accessible whenever the process system is opened. The tower ash receiver has to be replaced every 2 or 3 days, and the cyclone dust collectors are replaced every 2 or 3 weeks. The cooler lines frequently become restricted or plugged so that the system must be opened and cleaned. To date the most effective cleaning tools have been 12-ft-long, 1-in. steel rods and vacuum hoses. Dust control is difficult and never complete; some of the beta-active material gets on process equipment, tools, gloves, and work clothing. The basic cleaning equipment and work methods have remained essentially unchanged through the years as the UF_4 and UF_6 conversion efficiency has increased. Because of the significant beta exposure from contaminated work clothes, work methods were slightly modified in 1958, and new equipment is currently being investigated.

UF_6 Cylinders

The UF_6 produced by the fluorination tower is cold-condensed, liquefied, and drained on a batch basis to a 10-ton cylinder. The filled cylinder is eventually moved into a vaporizor where the UF_6 is reliquefied and vaporized. During the interval that the cylinder is full, there is a build-up of Th^{234}

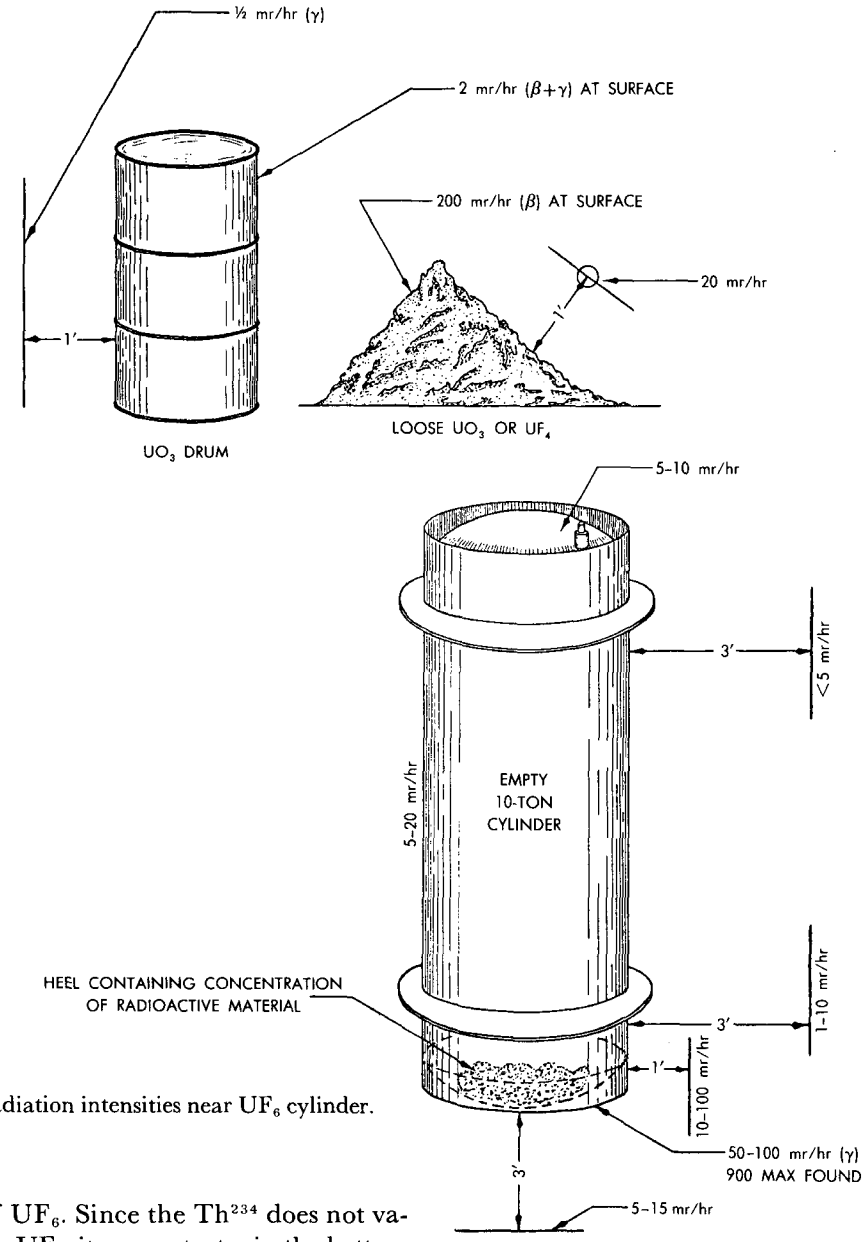


Figure 2. Radiation intensities near UF_6 cylinder.

in the mass of UF_6. Since the Th^{234} does not vaporize with the UF_6, it concentrates in the bottom of the cylinder. The maximum equilibrium content is about 2 curies each of Th^{234} and Pa^{234}. The maximum gamma reading at the outside surface of the cylinder may be 900 mrad/hr. Beta radiation readings of 50 rad/hr were taken at about 2 ft from the inside surface of the end of a cylinder cut open for corrosion inspection. Much of the beta-active material in the bottom end of the cylinder is redistributed when fresh UF_6 is drained into the cylinder; and, although it may appear strange, there is usually much more radiation from empty cylinders than from full ones. (See Figure 2.)

The Diffusion Cascade

Vaporized UF_6 is fed to the diffusion cascade where it is pumped, diffused, pumped, and diffused through many stages. Each stage of the cascade consists of a pump or compressor and a diffuser. Some of the uranium undergoes radioactive decay while going through the cascade just as it does anywhere else. Again the decay product is not volatile but attaches itself to the inner surfaces of cascade equipment apparently by impingement. There is a concentration of beta activity at the points of greater pressure and turbulence, but the

beta activity on the inner surfaces of equipment presents no problem during normal operations. Cascade components failing in service usually acquire accumulations of uranium compounds such as UO_2F_2 or UF_4 at the time of failure and require decontamination, after which the surface beta intensities are about 2 mrad/hr. However, during major cascade improvement programs, equipment in good operating condition and having no significant uranium accumulation is removed from the cascade for alteration. This equipment is moved directly from the cascade to the maintenance shops where it is opened and disassembled. Some pieces of equipment, such as the turbine-like compressors, require long hours of close work. The beta radiation at the surface is about 50 mrad/hr while that at from 1 to 2 ft from the surface may be 10 to 20 mrad/hr. The total beta exposure is that received directly from the equipment and that received from work clothing contaminated by contact with the equipment.

Uranium Metal Production

Part of the cascade tailings, UF_6 depleted in the U^{235} isotope, is made into uranium metal by reduction to UF_4 with hydrogen and to metal with magnesium. When there is no holdup of material in the production line, very little build-up of Th^{234} occurs, and the concentration of the Th^{234} at the tops and sides of the first melt (derbies) produces beta radiation of about 30 to 50 mrad/hr at the surface. However, the usual practice at the Paducah Plant is to run portions of the plant in campaigns, which results in the Th^{234} building up in UF_4, in derbies, and in metal castings. It is in the casting or remelt area that the most significant concentrations of beta-active material occur. Apparently the traces of thorium migrate to the surface of the melt and adhere to the graphite crucible walls and to any slag that may be present. The contact beta radiation readings at the inside surface of the crucible and at the surface of the crucible burn-out ash may be 3 to 10 rad/hr. The crucibles are cleaned by hand, and the radiation and radiation-emitting material are accessible to personnel.

PERSONNEL EXPOSURE

Monitoring Methods

The primary method of monitoring the exposure received while working around beta emitters in the metals plant and other areas is the film badge. We use the old style badge (PF-1B) which has an open window for detection of low energy beta radiation. A thin plastic envelope is used to keep beta-active dust out of the badge. Because of the contamination probabilities, the badges are exchanged on a weekly basis.

The film badges are calibrated for beta radiation by placing the film packet directly on a slab of uranium metal. We have found the film density due to the dose calculated from the published surface dose rate of the uranium metal to be approximately the same as that produced under the cadmium shield by the same dose of gamma radiation from Co^{60}. Our open-window beta calibration density-vs-dose curve lies on our gamma shield curve.

Because of the wide variation in temperature from one part of the plant to the other, we checked the variation of film sensitivity with temperature and found the sensitivity to increase with increasing temperature at a rate of 10% per 16°F over the range 20° to 100°.

In addition to the film monitoring, spot checks are made of the contamination on the clothing of the personnel whose work involves contact with beta-emitting material. Tests were conducted to determine the relation of the reading of portable beta instruments to the skin dose rate. These tests indicated that the Geiger-tube instrument (NICC-2610) and the Juno readings had to be multiplied by 3 to give the approximate skin dose rate. The following contact instrument readings are considered as levels below which contamination is insignificant in determining an employee's total dose.

	MRAD/HR
Issued clothing	0.75
Issued shoes, inside	3.0
Personal clothing	0.25
Gloves	3.0

Significant Area

Contamination of clothing raises the question of what is a significant skin area. The permissible exposure to the skin of the extremities, 75 rad/year, is much greater than that for the whole body, an average of 10 rad/year. Should the dose received by the skin under an area of contamination of about 10 cm^2 on the front pants pocket be added into a person's exposure record as whole-body exposure? If checks show one side of a man's

clothing to have significant beta radiation one week and the other side to have it the next, should the readings be additive? I am sure that some of you agree with an affirmative answer. In the fluorination tower area where our problem was the greatest, we found that the reading at the right front pocket averaged the highest, and this reading was recorded for calculating the additional skin dose to be recorded with the regular film badge results. In NBS *Handbook 59*, any part of the trunks and thighs is considered as the whole body for dose evaluation. (On page 76, 1 cm^2 is given as a significant area.)

Typical Exposure Record

Typical exposure experience as recorded by film badges is shown in Table 1.

Table 1

Skin Dose, mrad/year (1957)

	Maximum	Average
UF_6 manufacturing plant		
Operators	10.565 rad	2.96
Maintenance	8.020	3.34
Cascade equipment		
Maintenance (1955, 5 mo only)	5.65	4.94
Metal production		
Operators	5.87	1.82
Maintenance	0.91	0.476
Uranium recovery operators	8.825	3.5

Skin Dose From Clothing Contamination

Our record of skin dose due to beta emitters on clothing was started in July 1958. Personnel checks just prior to this time indicated that the combination of increased specific activity and the unconcerned attitude of the operators had increased the extent of clothing contamination and was resulting in skin exposure from this source of greater magnitude than that registered on the film badges. During July and August an average value of 22 mrad/hr of tower area operation was added to the operator's record. Usually this job is performed by two men on each of four rotating shifts. Rotation of personnel on this job was rapid. As a result of intensive efforts on the part of supervision and operators, the conditions changed to the extent that our checks during September showed an average value of 5 mrad/hr of tower operation to be appropriate. Current checks indicate the exposure rates to be insignificant.

EXPOSURE CONTROL

Incidental Shielding

The magnitude of the beta radiation sources, the frequency of open systems, and the contact type of maintenance indicate that exposure control must be exercised. We make use of inherent and deliberate shielding, mechanical equipment, and various means of control of clothing contamination.

Beta radiation is very easily shielded. Safety glasses, which are required on all maintenance work and chemical operations, provide about 90% attenuation of the beta radiation encountered. Shielding is also provided by sides and lids of drums, process piping, hoods, and catch buckets.

Shielding Devices

At the crucible cleaning station in the metal remelt operation, a down-draft ventilation and catch basin was designed with a deep-slotted grating to provide shielding from beta-emitting material in the catch pan. The reblading of UF_6 compressors required one man to work inside the rotor for several hours per day. An easily moved, easily attached sheet metal disk was employed to shield the mechanic. The disassembly of the sintered metal filter from the fluorinator ash system is performed under water for maximum shielding.

Mechanical Equipment

In general, the larger the containers used to handle our active material, the lower the personnel exposure. Fewer transfers are required, and, with large containers, mechanical equipment such as overhead cranes and fork lifts are needed and provide the advantages of remote handling.

Clothing Contamination Control

Some of the work mentioned above results in rather heavy clothing contamination. In some cases the simplest control is provision of company issued coveralls and the requirement of an immediate change following the dirty job. We also use plastic aprons. We first used a light-weight but

large apron, but the operators objected to its interference with work, additional heat, and general discomfort. We now use an apron shortened at the bottom, cut back at the sides, and equipped with strong cotton cloth ties. The modifications have reduced the objections, the apron is easily removed and cleaned, and the beta exposure from clothing has been greatly reduced.

Exposure from beta emitters on clothing in other areas has been reduced by rather simple means. A welder working inside cascade diffusers during a major alteration program had high beta readings on the seat of his pants until he was given a small wooden stool. General reductions in beta exposure are obtained by employee education. The factors involved in exposure reduction are usually simple and readily demonstrable and can change the operator's attitude from unconcern to alert confidence.

SUMMARY

The beta-emitting materials associated with uranium processes are concentrated by fluorination of UF_4 to UF_6, vaporization of UF_6, and impaction on process components, and during metal casting. This concentration does not prohibit contact maintenance and operation, but increases the problems. The resulting exposures respond to the applied control measures and are within acceptable limits. Clothing contaminated with concentrated beta emitters presents a difficult monitoring problem, in which the accumulated dose is approximated by applying statistics from spot checks.

In Vivo Counting as a Device for Evaluating Uranium Exposure*

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Abstract

This paper concerns whole-body counting of individuals following enriched uranium inhalation. The general configuration is described of a 7×7×7-ft shield with 8-in. steel walls. The data, derived primarily from measurements in this shield, are coded on a 250-channel spectrum analyzer. Background measurements, phantom measurements, and measurements of certain individuals are discussed. Some comparisons are made of whole-body measurements and urine data following inhalation. Whole-body counts are presented for 15 to 20 patients, but it is emphasized that the data are preliminary and unevaluated.

*Because of the preliminary nature of the data presented, only an abstract of this paper is printed here. It will be published in final form as *In Vivo Gamma Counting Method of Determining Uranium Lung Burden in Humans*, Y-1250, by R.E. Cofield.

Possibilities and Limitations of Whole-Body Counting in Assessing Burdens of Natural Uranium

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This paper describes the possibility of measuring natural uranium in the human body by *in vivo* counting. The predictions are based on several years' experience with a whole-body counter at Harwell, although, as we have never had occasion to measure a subject contaminated with natural uranium, some of the following remarks must of necessity be speculative.

MEASUREMENT OF BODY RADIOACTIVITY

The equipment in use at Harwell for the measurement of body radioactivity has been described in detail elsewhere,[1] and only a brief description will be given here. The measurements are made inside a shield constructed from interlocking lead bricks 4 in. thick (Figure 1). In order to reduce the chance of observing surface contamination (on skin, in hair, etc.), the subject showers and changes into clean pajamas and socks before the measurement, during which he lies on a special stretcher, as shown in Figure 1. Four sodium iodide crystals 4.25 in. in diameter by 2 in. thick are used as the detectors, and these are disposed above and below the body in such a way as to minimize dependence of response on distribution of activity within the body. The outputs from the 4 photomultipliers are fed in parallel to a linear amplifier and thence to a multichannel pulse amplitude analyzer; the gain of the system is set to scan the energy range 25 kev to 1.675 Mev in 33 channels of 50-kev width.

The main gamma radiation from the human body is due to naturally radioactive potassium (K^{40}) and to a small amount of the fission product Cs^{137} from nuclear weapons fallout, acquired from food. In Figure 2 are shown gamma-ray spectra from two subjects (experimental points) superimposed on the spectra (smooth curves) from a phantom containing the amounts of potassium and Cs^{137} indicated. The subject of curve A was measured in late 1956 with our first arrangement using 4 small sodium iodide crystals; this man was not occupationally exposed to radioactive materials, and the amount of cesium found was typical of the average levels at that time. Curve B is the spectrum from a man slightly contaminated with Cs^{137}, taken with the present arrangement of 4 large sodium iodide crystals. In both cases the agreement between the calculated and experimental spectra is good, indicating that scattering and absorption of gamma-rays are the same in the phantom as in the subject. Thus it is possible to use the response above 0.5 Mev to calculate the contributions from potassium and cesium below this energy. When a subject is contaminated with a nuclide emitting gamma-rays of energy <0.5 Mev, the net contribution from the contaminant can be determined in this way. This also applies in the case of bremsstrahlung from beta-particles,

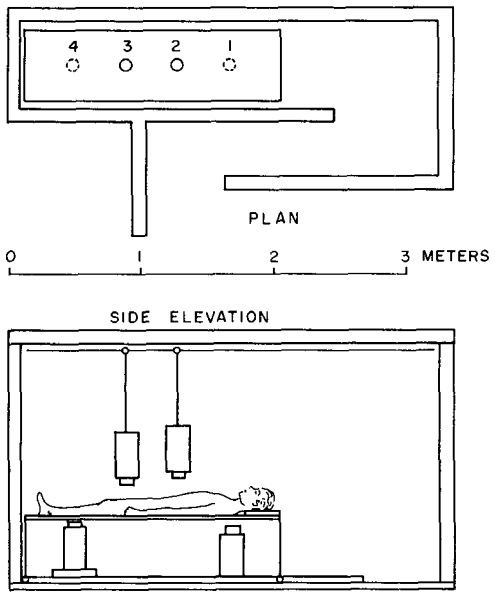

Figure 1. Layout of the lead shield and disposition of the four detectors for measurement of body radioactivity.

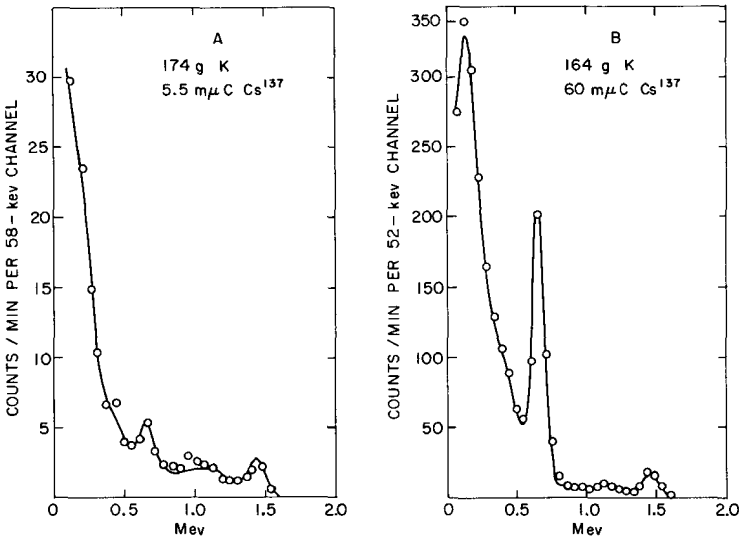

Figure 2. The smooth curves are the spectra of gamma radiation from the phantom containing the amounts shown of potassium and Cs^{137}, compared with the observed spectra (experimental points) from two subjects. A: Uncontaminated subject (four small crystals). B: Subject slightly contaminated with Cs^{137} (four large crystals).

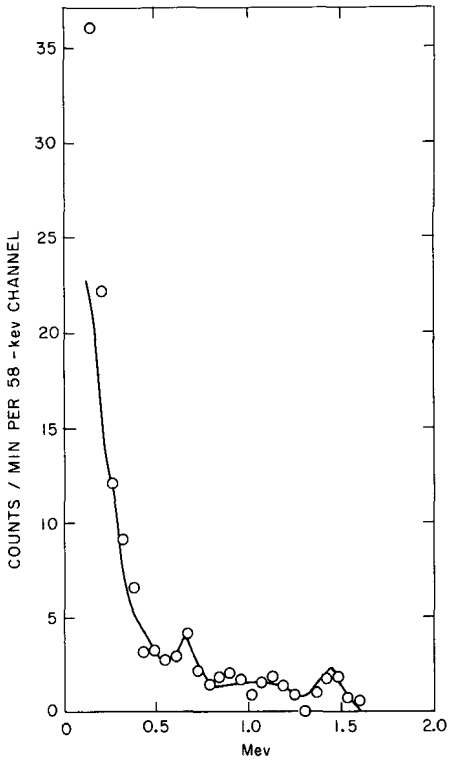

Figure 3. Calculated spectrum (smooth curve) of radiation from 132 g potassium and 3.7 mμC Cs^{137} in the phantom, measured with the four small crystals. The experimental points are the observed spectrum from a subject exposed to Sr^{90}.

and in Figure 3 is shown the spectrum from a man slightly contaminated with Sr^{90}. The smooth curve is the spectrum from 132 g potassium and 3.7 mμC Cs^{137}; above about 0.4 Mev, the spectrum from the subject (experimental points) is in reasonable agreement with this curve, but below 0.4 Mev there is a considerable excess of counts from the subject, amounting to 68% at 0.15 Mev. The excess corresponded to about 0.36 μC Sr^{90}.

ELECTROMAGNETIC RADIATION FROM URANIUM

The radioactive decay of U^{238} and U^{234} proceeds as follows:

$$U^{238}\,(UI) \xrightarrow[4.5\times10^{9}\ yr]{\alpha} Th^{234}\,(UX_1) \xrightarrow[24\ day]{\beta}$$

$$Pa^{234}\,(UX_2) \xrightarrow[1.2\ min]{\beta} U^{234}\,(UII) \xrightarrow[2.5\times10^{5}\ yr]{\alpha} Th^{230}\,(I_0).$$

The long half-life of Th^{230} prevents a significant activity from building up (except over a period of very many years), and the series can be regarded as stopping at U^{234}. All the nuclides shown emit some low energy nuclear gamma-rays, but of low intensity, and many of them are highly internally converted. There are also some characteristic x-rays. In addition, there is a weak branch in the chain via a metastable state of Pa^{234}, which emits several gamma-rays of energies up to 1.8 Mev, but their low abundance ($<1\%$) precludes their use in estimating small amounts of uranium *in vivo*. The gamma radiation from U^{235} is also too low in intensity in natural uranium to be usable in this connection, although it is of great value in cases of internal contamination with highly enriched uranium, as shown by Miller,[2] and discussed by Burkhart in the previous paper.

The only practicable means of measuring natural uranium in the body by *in vivo* counting is to make use of the bremsstrahlung accompanying the beta decay of Pa^{234}, which emits beta-particles of maximum energy 2.3 Mev in more than 90% of its disintegrations. In Figure 4 is shown the spectrum from 140 g potassium and 7 mμC Cs^{137} in a phantom in our equipment, together with the extra response from 90 mg natural uranium (the current maximum permissible body burden for skele-

tal deposition, corresponding to 0.03 μC of each of the four nuclides U^{238}, Th^{234}, Pa^{234}, and Th^{230}). The level of Cs^{137} chosen is about the average currently found in persons resident in the Harwell area with no occupational exposure to this nuclide. The increase in the response above 0.5 Mev amounts to 8 counts per minute (cpm), compared with 218 cpm from the cesium and potassium, whereas below 0.5 Mev the response from the uranium is 197 cpm, compared with 491 cpm from the cesium and potassium. The latter counting rate would be overestimated by about 4% or 20 cpm when calculated from the 226 cpm above 0.5 Mev. The estimated contribution to the counting rate from the uranium below 0.5 Mev would thus be 10% low. However, the maximum permissible body burden can be measured in this way with acceptable accuracy, and it would be possible to measure somewhat less than this quantity.

It should be noted that the maximum permissible body burden when the kidneys are the critical organ is likely to be reduced to 0.005 μC or 15 mg uranium. It is doubtful if this quantity could be measured by external whole-body counting, and urinalysis would be much more sensitive. Further, since only 6.5% of the body content is in the kidneys, a counter placed close to the kidneys would still not be able to detect the uranium.

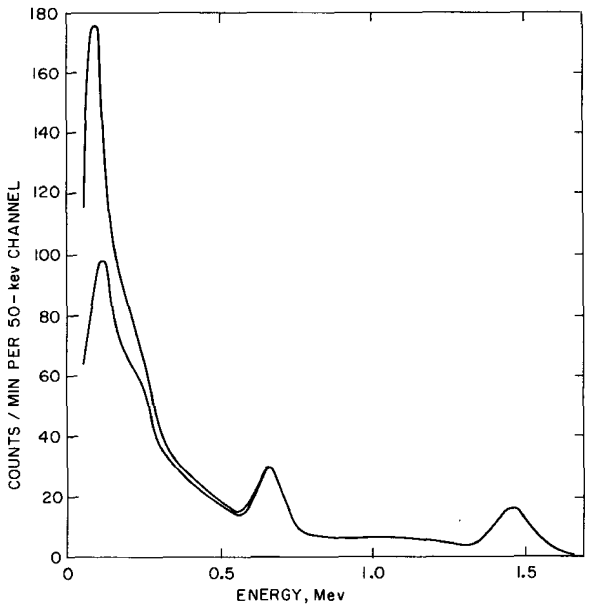

Figure 4. Spectrum from 140 g potassium and 7 mμC Cs^{137} in the phantom measured with the four large crystals, and the extra response from 90 mg natural uranium.

LIMITATIONS OF THE TECHNIQUE

Three conditions must be fulfilled if the maximum sensitivity is to be achieved.

First, the state of equilibrium of the 24-day Pa^{234} must be known. Freshly purified uranium has little or no beta activity, and it reaches 94% of equilibrium in 96 days. Internal contamination with uranium purified more recently than this can be assessed only by comparison with a standard of the same specific beta activity, and 90 mg uranium will give a lower response than that indicated in Figure 4 and above. This aspect may also give rise to some very misleading results. The Pa^{234} tends to migrate to the surface of uranium during heat treatment; inhalation of material with high specific beta activity is possible, and a body radioactivity measurement might suggest a substantial uranium burden. Conversely, if the material with high beta activity is removed, a surface with low beta activity is exposed, and a subject could have a large burden of uranium without giving a greatly increased response in the whole-body counter.

Second, there must have been no selective excretion by the body of the beta-active daughter products; in cases of inhalation of insoluble uranium of medium particle size this is unlikely to occur, but the possibility must be borne in mind. It could be checked by estimating the uranium in the excreta by a bremsstrahlung measurement, as well as by the usual method.

Finally, the problem of surface contamination must be mentioned. Sodium iodide crystals are somewhat sensitive to beta-particles, and the counting rate is about one order of magnitude greater for their direct detection than for their detection via the bremsstrahlung they produce. Where surface contamination is suspected, shielding the counters with sufficient low-Z material to absorb all the beta-particles enables its presence to be checked. A substantial drop in the counting rate with the counters shielded indicates surface contamination.

CONCLUSIONS

The detection of uranium in the body by *in vivo* counting is possible by using sodium iodide crystals and gamma-ray spectrometry, but the sensitivity is not very great and calibration presents a formidable problem. However it should be possible to measure down to about one third of the

maximum permissible level for skeletal deposition, with an error of perhaps 25 or 30%. This would be satisfactory for checking occasional cases where contamination has been demonstrated by the finding of uranium in urine or feces; by following reduction in the body activity and determining the amount of uranium excreted, a check on the calibration would be obtained, which would lend confidence to future measurements. However, the time required to interpret the gamma-ray spectrum would probably preclude the use of the technique as a routine tool, and it is unlikely ever to supplant urinalysis.

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Air Sampling Procedures in Evaluating Exposures to Uranium*

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The many types of air sampling equipment and procedures used to evaluate exposures to air-borne uranium aerosols would seem to indicate that there are several solutions to the problem, all equally good. However, closer examination reveals that the data from many laboratories and plants do not accurately represent the workmen's exposure and are not comparable. It has been suggested that the differences in the air sampling procedures are due to two distinct philosophies of air sampling, termed the "industrial" and the "laboratory" viewpoints in the report *Health Protection Practices Survey,* October 1957, by W.B. Harris of HASL.

Standard air sampling procedures used to evaluate exposures during routine industrial processes are well known and have been described for the nuclear industry by Klevin and Harris.[1] In fact, standard air sampling procedures for industry were described by Bloomfield[2] of the U.S. Public Health Service in 1935. However, air sampling procedures used in research and development laboratories have not been standardized and are known to vary widely from one laboratory to another. Perhaps the many procedures developed in the laboratories were an outgrowth of the methods developed by physicists during the early part of the Manhattan Project when secrecy prevented them from hiring or obtaining the services of trained industrial hygienists.

From the standpoint of air sampling, the most important difference between routine industrial operations and research and development laboratories is the wide variety of jobs performed by individual workers. In industry, one or more men may work on the same process every day throughout the year. In research and development laboratories, an individual may work with different equipment each day. If he does use the same equipment, the exposure may vary because the mass or shape of the material worked is constantly changed. Our experience at Los Alamos indicates that each man will perform many jobs in an 8-hr shift, and the jobs will differ from day to day. Thus, it is obvious that air sampling procedures in research and development laboratories must differ from those used for industrial processes. This paper will be limited to a discussion of the former.

Air is sampled for air-borne uranium aerosols to evaluate personnel exposures and the effectiveness of control measures, to measure the quantity discharged from stacks, and as a check on the proper operation of processing equipment. To evaluate the exposures of workmen breathing contaminated air, it is necessary to determine the mass concentration and the particle size distribution of the uranium aerosol. Air samples should be collected in the breathing zone (BZ) of the workman during each type of work performed during the day to calculate the average weighted concentration. In general, it is agreed that this is the only way that personnel exposure records can be correlated with medical records. In actual practice, most laboratories measure only the mass concentration by a combination of BZ and general air (GA) sampling. The particle size distribution is usually determined with a cascade impactor or a Millipore filter.

Burnett and Hatch[3] stress the nonuniform retention and deposition of air-borne activity and the necessity of determining the particle size of the dust breathed, and conclude that none of the air sampling procedures in use today permits the correlation of exposure data and medical records. They propose[5] the use of a 2-stage air sampler simulating the collection of aerosols in the upper respiratory tract and the lungs. Air sampling instruments of this type have also been suggested by Harris and Eisenbud,[4] May,[6] and others. At this symposium J.E. Ross has discussed the use of a pre-impinger, representing a 2-stage sampler. Since current air sampling procedures do not provide exposure data that can be correlated with

*This work was done under the auspices of the U.S. Atomic Energy Commission.

in dry boxes or enclosures and any air-borne contaminants are dispersed uniformly into the room air.

In some situations both BZ and GA samples are required to evaluate the workmen's exposure. In a uranium foundry, BZ samples would be necessary for the workmen who stand over the casting during its removal from the mold. During the many hours of the melting and pouring cycle when the workmen move about the foundry, GA samples will adequately evaluate their exposure. Generally speaking, if there is any question regarding the type of air sample that should be collected, a BZ sample would be the choice. It is obvious that this presents a real problem for persons confronted with air sampling for the first time. The first emphasis then should be on obtaining industrial hygienists or health physicists trained in industrial hygiene air sampling procedures.

AIR SAMPLING EQUIPMENT

An instrument for measuring air concentrations of uranium aerosols should have certain characteristics for general use, the most important being a known or high collection efficiency for the aerosol. The instrument should collect samples in such a medium that the amount collected can be readily evaluated by counting or other means. The sensitivity should be such that a concentration 1/10 the maximum permissible can be measured in a sample collected in a reasonable period of time. This, in turn, is determined by the value of the MPC and the sensitivity of the counting procedure. The results should be obtained rapidly so that corrective measures can be started quickly whenever high concentrations are found. Finally, the instrument should be light in weight and readily portable so that it can be moved close to the BZ of the

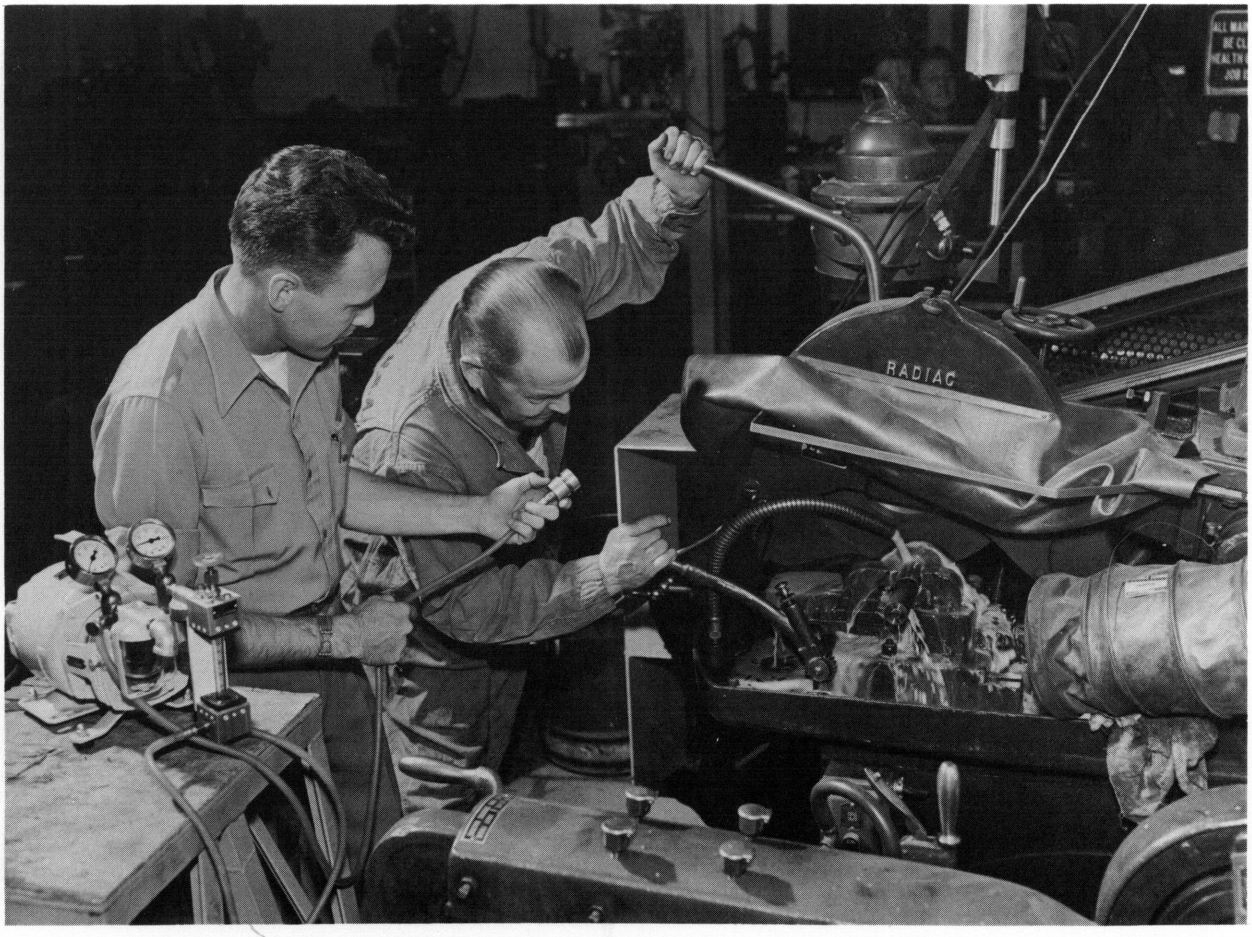

Figure 3. Breathing zone air sampling in foreground and general air sampling with Filter Queen in background.

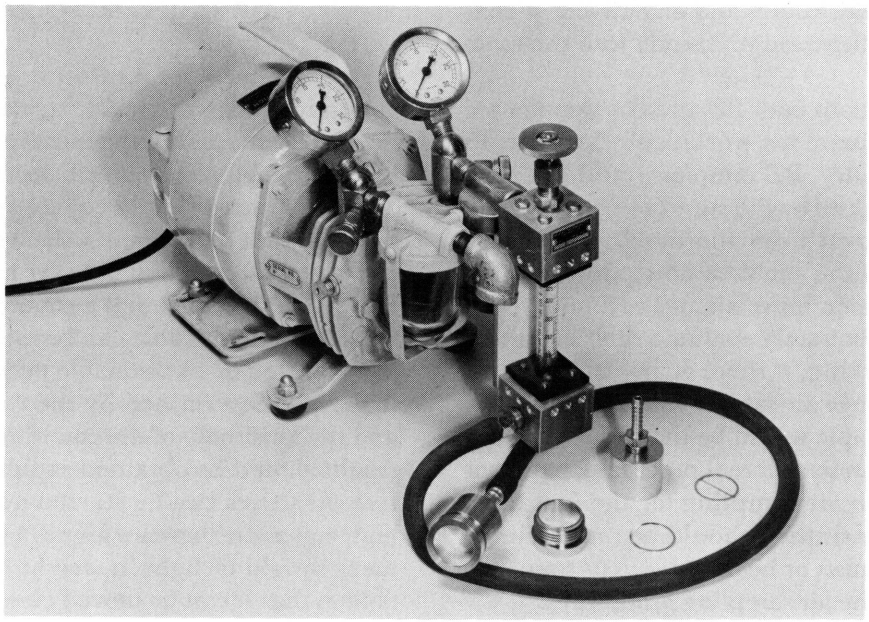

Figure 4. A 1⅛-in. sampling head with a Gast pump and rotameter.

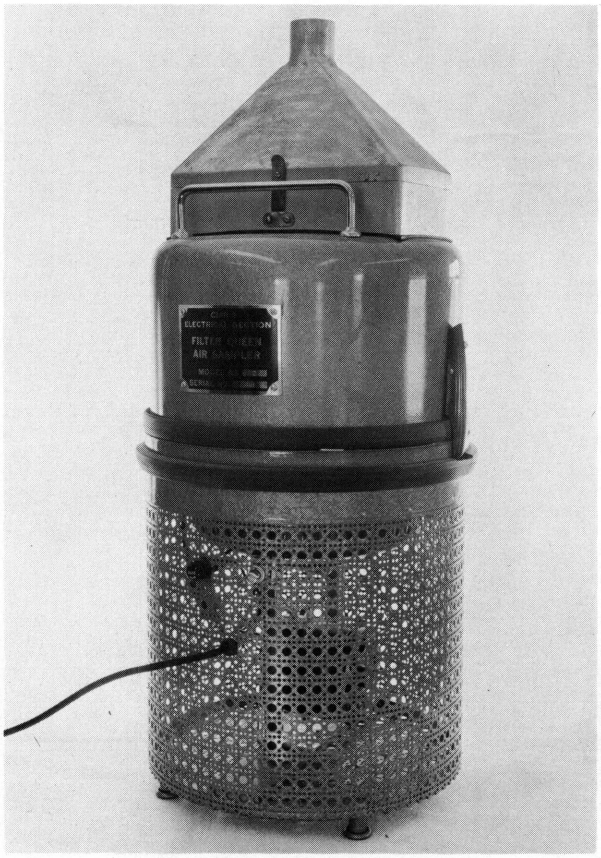

Figure 5. Los Alamos modification of the Filter Queen air sampler.

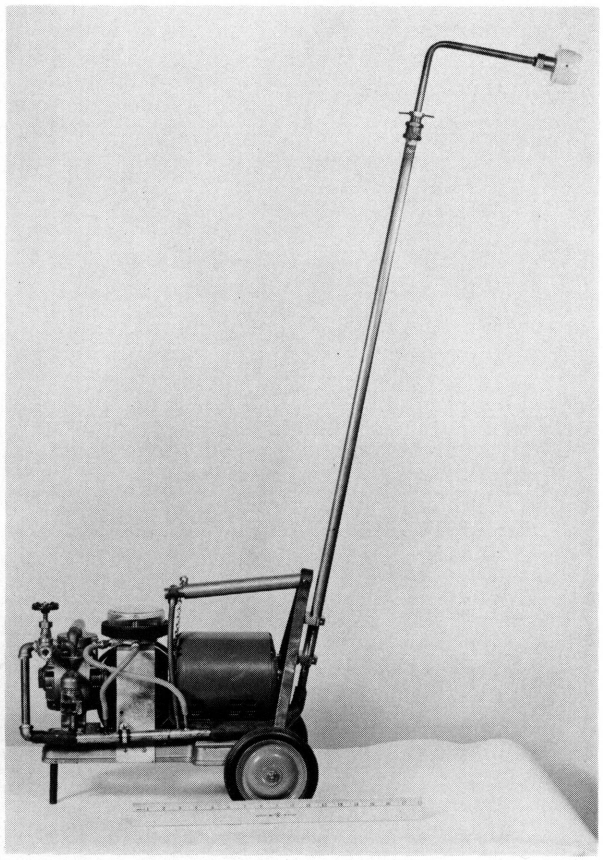

Figure 6. A portable Gast air pump equipped with an orifice meter, sampling through a 2⅛-in. plastic head.

in dry boxes or enclosures and any air-borne contaminants are dispersed uniformly into the room air.

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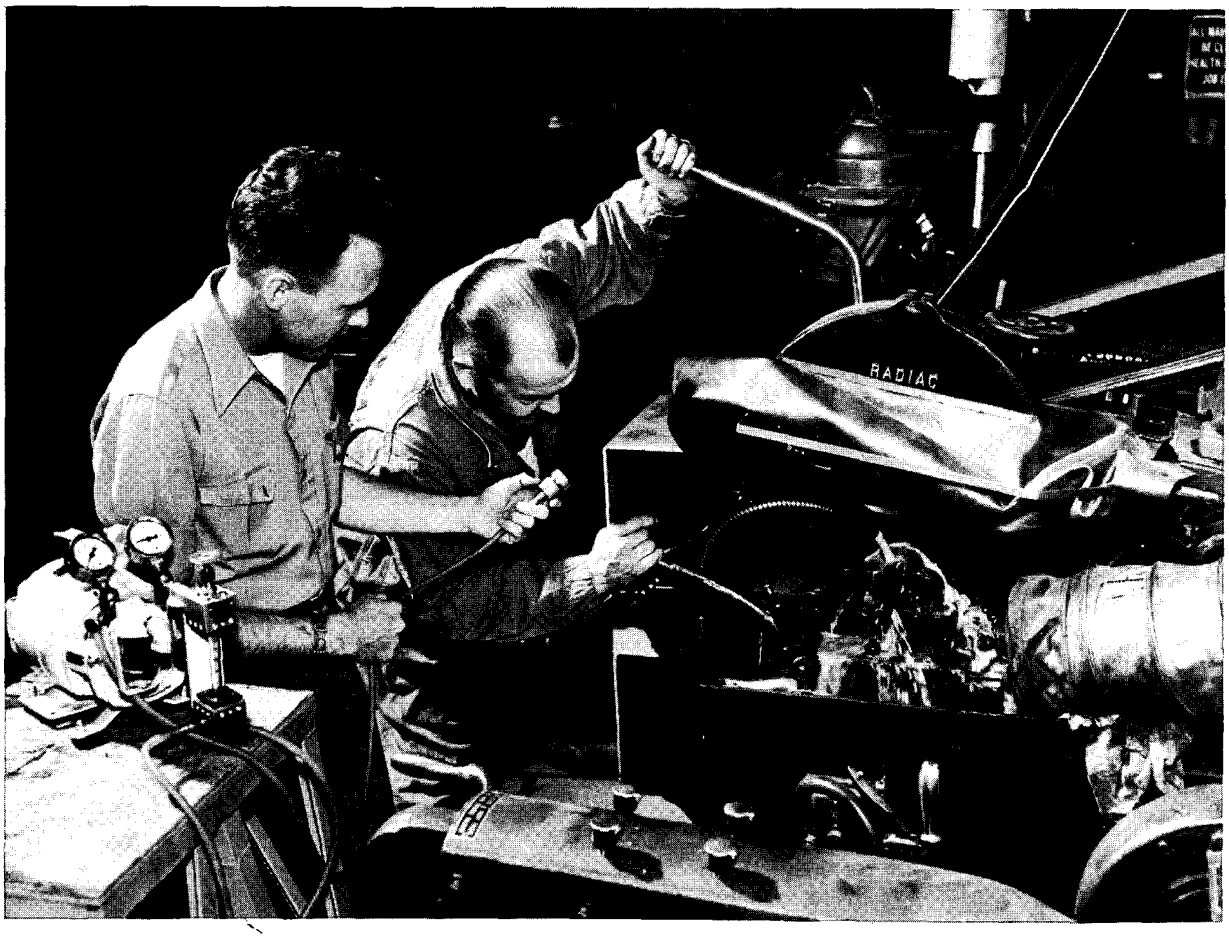

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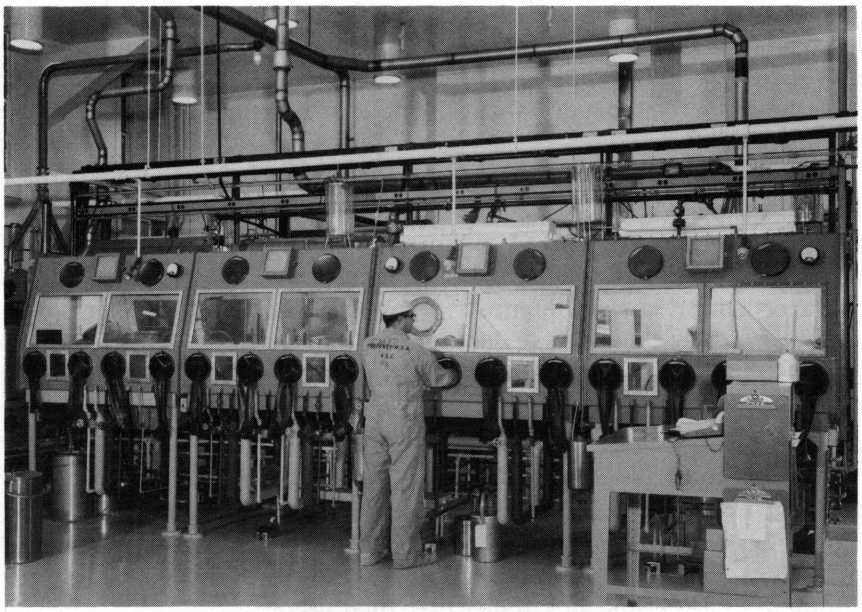


Figure 7. Central air sampling system on line of dry boxes. A plastic sampling head is shown ≈10 in. above the workman's head. The 2-in. white air sampling line above the dry boxes is one of many such lines operating from one central multistage air pump.

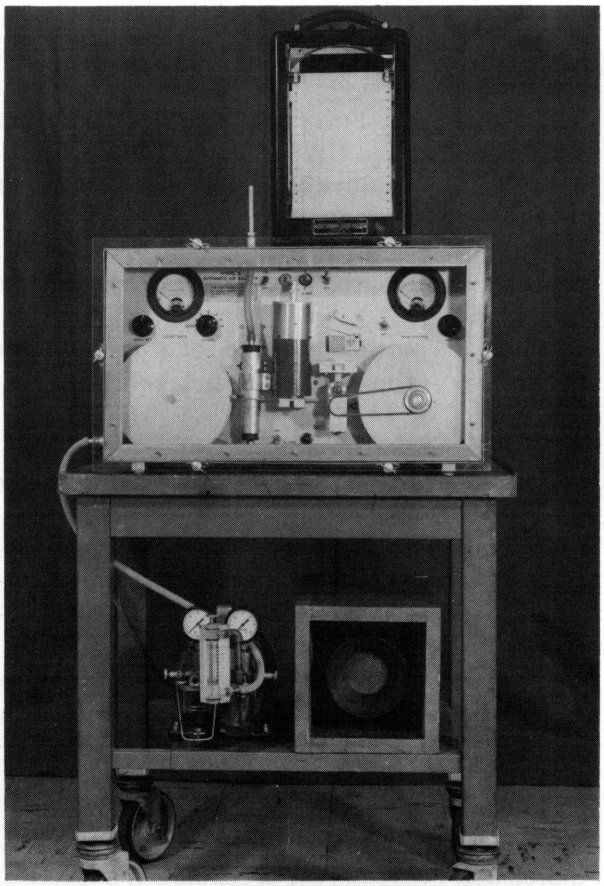


Figure 8. A recording automatic intermittent air sampler for alpha aerosols that sets off an alarm at dangerous concentrations.

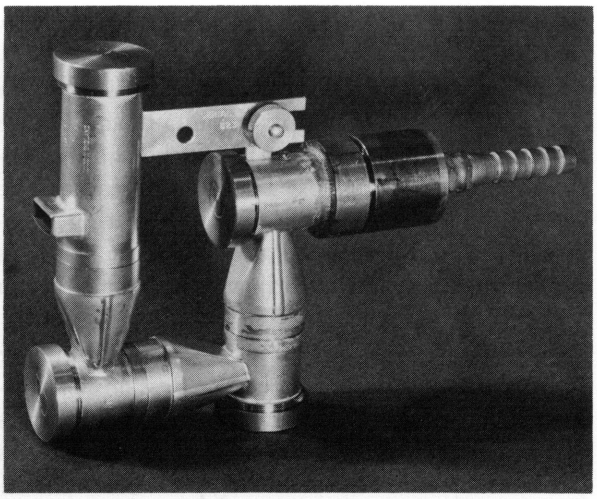


Figure 9. Los Alamos modification of the Casella cascade impactor.

exposed worker. The most common method of air sampling is to draw air through filter paper at a known flow rate by some type of pump; the complete instrument includes the filter paper holder, the air flow rate indicator, and a pump.

Types of air sampling equipment used at Los Alamos are illustrated in the following figures. Figure 4 shows a 1⅛-in. sampling head with a Gast pump and rotameter, a unit introduced by HASL now in common use to collect BZ samples. Figure 5 shows the Los Alamos modification of the Filter Queen air sampler commonly used to collect GA samples.

Figure 6 shows a portable Gast air pump equipped with an orifice meter, and sampling through a 2⅛-in. plastic head, called the Giraffe. The plastic sampling head and pump were designed by the Rocky Flats, Colorado, plant of the Dow Chemical Company. The Giraffe air sampler makes it possible to collect continuous or short-term BZ samples without holding the sampling head by hand in the BZ. We modified the Dow design by adding an orifice meter to measure the air flow, which ranges from 0 to 2.5 cfm but is normally 2 cfm.

To permit the collection of several continuous air samples in a given area, we have installed central air sampling pumps capable of drawing 2 cfm through as many as 60 air sampling heads. Figure 7 shows a sampling head $\approx$10 in. above the workman's head at a dry box. The plastic head can be easily changed to collect BZ samples during a given operation or can serve as a continuous process sampler. The air flow is adjusted at 2 cfm with an orifice meter as shown.

Figure 8 shows an automatic intermittent air sampler that may be used as a process or GA sampler. This unit may be adjusted to collect samples intermittently for 10-min to 6-hr periods and has proven very useful in measuring peak dust concentrations. Figure 9 shows the Los Alamos modification of the Casella cascade impactor used to sample aerosols for particle size analysis. Note the metal end caps replacing the original rubber ones.

The air flow rate indicator and the air pump are generally standard items of equipment and do not represent a significant source of error in air sampling. However, the filter media used and the air sampling rates vary widely from one laboratory to another and are frequently the source of large differences in the results reported.

A survey of air sampling practices and filter media among AEC contractors was made by the A.D. Little Company[7] in 1953. They found 37 laboratories using 22 different filter media for 85 applications. The group most frequently used was Whatman papers, #41 being most used in the group. The single most widely used paper was HV #70 (Hollingsworth and Vose). Next were membrane filters and glass fiber; other media were used in much smaller quantities.

After a thorough study of the requirements set forth by the 37 laboratories and performance tests of all media, 5 filter papers were selected by the A.D. Little Company and presented for comment. To date, no effort has been made to standardize on these 5 filters or on any other group of filter media. The 5 papers recommended are listed below, with comments on appropriate situations for their use.

1. WHATMAN #41

Use where large volume sampling is desirable, and where a high sampling rate will provide good collection efficiency.

2. WHATMAN #44

Use where a low ash paper of good collection efficiency is needed, especially at low flow rates. This should be the general purpose paper of the air-assay laboratory.

3. MEMBRANE FILTER (Example: Millipore Types AA and HA)

a. For the quantitative collection of the very finest particles (submicron).

b. For collection of particles that are to be viewed, counted, or measured on the filter directly under the microscope.

c. Where the particles must be collected wholly at the surface (as in α counting); here HA is preferred.

d. When quantitative collection of very small particles is coupled with the need to ash the filter during analysis.

4. HV #70 PAPER

a. For monitoring devices requiring a high efficiency paper, and where ash content is of no concern.

b. In continuous monitoring stations requiring low resistance, high efficiency paper in roll form, where color or ash content of the paper is not important.

5. GLASS FIBER PAPERS

To be used where high collection efficiency is required and where the use of cellulose is precluded,

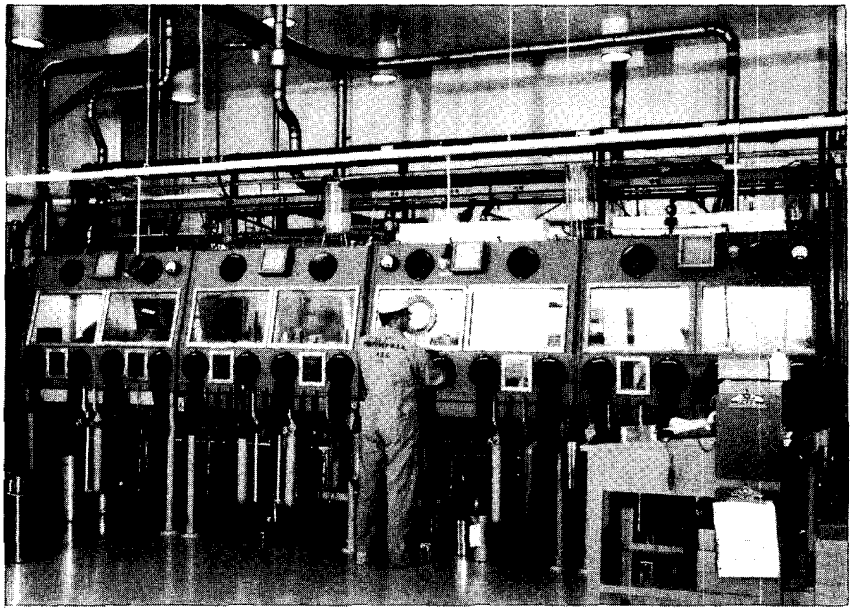
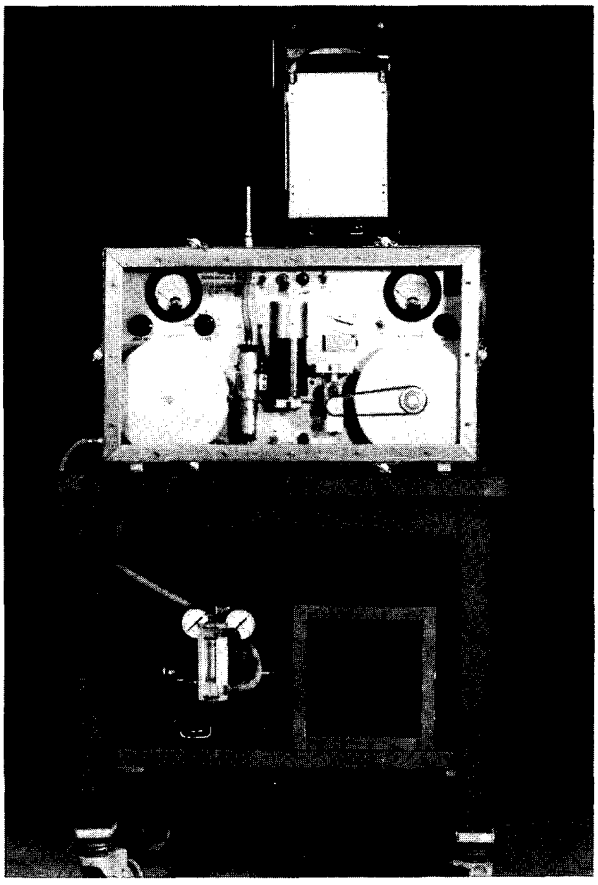


Figure 7. Central air sampling system on line of dry boxes. A plastic sampling head is shown $\approx$10 in. above the workman's head. The 2-in. white air sampling line above the dry boxes is one of many such lines operating from one central multistage air pump.



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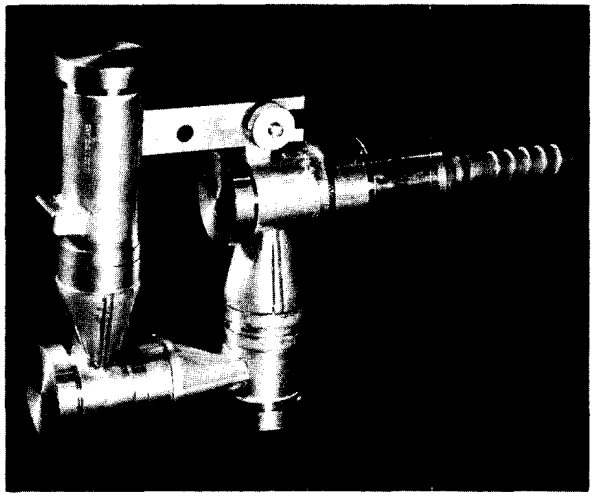

Figure 9. Los Alamos modification of the Casella cascade impactor.

e.g., sampling of high temperature stack gases. MSA Paper No. 1106B (Mine Safety Appliances Company), or any equivalent paper, is recommended. The AEC all-glass air filter medium also may be used successfully for assay purposes.

SUMMARY

At present efforts are being made to improve air sampling procedures to distinguish between safe exposure levels and minimal hazard levels. Concentrations are no longer expressed in multiples of the maximum permissible levels as they sometimes were in the early days of the uranium industry. As the quality of dust control improves, the requirements of air sampling will change. It is apparent that some have lost sight of the ultimate purpose of air sampling. We believe this is best summarized in the following quotation by Professor Theodore Hatch.[8] "The ultimate purpose of sampling and analysis of air-borne dust is to obtain the necessary information concerning the nature and magnitude of dust exposure from which to predict the kind and extent of health hazard that might result from inhalation of the dust by exposed workers. The adequacy of the method of sampling and of the analytical technique used to study the collected dust must be judged, therefore, in terms of biological rather than physical criteria."

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Air Sampling Procedures in Evaluating Exposures

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The evaluation of the degree of contamination of a plant atmosphere as well as of occupational exposures to the contaminant may be accomplished by means of carefully selected air samples. In general, five basic methods are most widely used to determine exposures to radioactive or toxic materials, all dependent on obtaining some type of sample of the atmosphere normally breathed by the worker. These are (1) fixed location general air sampling, (2) multiple spot general air sampling at locations dictated by experience, (3) fixed location semi-breathing zone sampling, (4) multiple sample time weighted average exposure procedure, and (5) sampling in which the operator is used as a sample collecting device. Each can be defended on some basis: one may prove more applicable to certain types of operation, a second better for the organizational arrangement of the sampling group, another more economical, and still another more accurate. It is worth while to examine the variables inherent in these different methods in order to indicate which procedure is best suited to the needs of any particular case.

1. Fixed Location General Air Sample

A sample of the workroom atmosphere is taken from a single fixed location with the following qualifications:

a. Reasonably representative of the average air concentration.
b. Easily accessible for sample changing.
c. Easily accessible to power (electric or vacuum).
d. Unobtrusive.

The basic advantages of this kind of sampling are:

a. The sample may be taken throughout the entire operation period or even for 24 hr/day.
b. Personnel costs are minimal.
c. Long-term sampling tends to average exposure peaks and represent a leveled-off concentration.
d. The psychological effect on the operators is that, although not embarrassed by its presence, they are aware of the constant monitoring.
e. This sampling is useful in emergency situations where sampling of personnel might not be possible.

The disadvantages include:

a. The concentration gradient in a space can be very large, and the likelihood of the sampler being in an area of average concentration is small.
b. Although this sampling method recognizes concentration peaks, it does not evaluate them.
c. Except in areas where the air turbulence is extremely high, the sample does not represent true exposure and can vary from it by orders of magnitude.
d. Equipment cost is generally high.
e. Such sampling provides poor guidance for engineering modification.

2. Multiple Spot General Air Samples Taken at Locations Dictated by Experience

This type of sample is generally taken for up to about 10 min, the equipment being placed and handled by a sampler in locations predetermined to be representative of average concentrations. The advantages of this type of sampling include:

a. In the hands of an experienced person it provides a simple, rapid method for evaluating average exposures.
b. Equipment cost is small.
c. Personnel time is small.

The disadvantages include:

a. The sample is taken during a relatively small portion of the operating time, and thus unusual conditions may be missed.
b. A high degree of skill is required on the part of the sampler.
c. At best, this procedure does not truly evaluate occupational exposures, but provides a guide to an understanding of the situation.
d. This type of sampling provides a relatively inadequate basis for engineering controls.

3. Fixed Location Semi-Breathing Zone Samples

This type of sampling is used at fixed location operations. The sample is taken by a device whose intake may be positioned at or very near the operator's breathing zone. The advantages of this procedure are:

a. For applicable operations it can provide useful information on exposure to operators during routine procedures.
b. Operator time requirement is small.
c. Useful information is obtained about worker performance and for engineering controls.
d. This method can cover whole shift operations and provide averaged concentration values.

The disadvantages include:

a. High equipment cost.
b. Inflexibility.
c. Psychological pressure on the operator.
d. High sensitivity to placement.
e. The method is inadequate to define peak concentrations.
f. Time spent away from the operation by the operator is excluded from sampling.

4. Multiple Sample Time Weighted Average Exposure Procedure

The basic principle is to obtain a large number of samples which in the judgement of the sampler truly represent each increment of the operator's exposure during the working day. The advantages of this system include:

a. Depending on the judgement of the sampler, this method can provide a representative average of the daily exposure under the conditions observed.
b. Equipment cost is low.
c. Degree of replicability is high.
d. Accurate evaluation is provided for engineering control requirements.
e. The sampler gains a deep understanding of the operation.
f. Normal peak concentrations are recognized.

The disadvantages are:

a. High manpower cost.
b. Only the conditions at the time of sampling are recognized.
c. The method places a significant psychological restriction on the operator, which is not permanent.

5A. Respiratory Sampling

In this procedure the operator is provided with a respirator for all or a part of the workday, and analysis of the respirator filter provides a measure of his exposure. A variant is to equip the operator with a sampling device which he carries with the intake near his nose. The advantages of this system are:

a. Such a procedure will probably most accurately reflect the average exposure of the operator.
b. Personnel cost is low.
c. Equipment cost is generally low.

The disadvantages are:

a. Strong psychological pressure on the operator, although the presence of the equipment throughout the workday would probably reduce this significantly.
b. This system is usable only on an occasional basis, and thus might miss unusual circumstances.
c. It is apt to be considered cumbersome by the operator.
d. The respiratory unit may be inaccurate because of poor fit.

5B. Biological Sampling

As generally used this procedure involves obtaining a biological sample from the operator for analysis for the material of interest. The advantages are:

a. It indicates the true biological accumulation more closely than other methods.
b. No psychological stress is placed on the operator.
c. Equipment cost is low.
d. Operator cost is low.

The disadvantages are:

a. Peak concentrations are disregarded.
b. The method is highly dependent on the physiological handling of the material.
c. The results are highly dependent on the relationship between sampling time and exposure time.
d. The sample represents a combination of exposure immediately preceding the sample and chronic accumulation that is difficult to interpret.
e. Sensitivity varies with chemical composition of material, and its biological fate.

Of all these methods, the HASL has found from experience that the multiple sample time weighted average exposure procedure (No. 4 above) is the most accurate and desirable. A large number of

Table 1

Analysis of Variance for Three-Way Classification With Replication

Source of variation	Sum of squares	Degrees of freedom	Mean square	*F* ratio
Time	776,127,082	2	388,063,541	1.28
Site (area or location)	1,811,467,319	3	603,822,439.6	1.99
Character of sample (GA or BZ)	746,756,658	1	746,756,658	2.47
Time×site	1,833,586,544	6	305,597,590.6	1.01
Time×character of sample	720,553,491	2	360,276,745.5	1.19
Site×character of sample	1,819,853,141	3	606,308,856.8	2.0
Time×site×character of sample	1,816,783,426	6	302,797,237	*
Within replicates	1,845,840,405	48	38,455,088	–
Total	11,370,968,066	71		

*T×S×C/W.R. = 7.87; therefore, T×S×C is used as the divisor in testing for significance.

carefully selected samples are obtained, from both the general air (GA) and the breathing zone (BZ). The BZ samples are taken at least in triplicate wherever possible, and average concentration values are computed from the replicates. The time study data necessary to compute the average exposure are obtained as a corollary of the BZ sampling and from discussion with supervisory personnel.

To determine the merits of obtaining either GA or BZ samples versus obtaining both for exposure evaluation, data obtained from several plant surveys were statistically analyzed. Three surveys conducted at one plant over a 3-year period were subjected to a three-way classification with replication analysis of variance. In each survey multiple sampling was performed at comparable locations and at comparable operations within these locations. The variables consisted of main effects, interactions, and within replicate groups. The main effect variables were (1) type of sample (GA or BZ), (2) location or area of samples, and (3) time (i.e., which survey). The interaction variables were: (1) type of sample and time, (2) type of sample and area, (3) time and area, and (4) time and area and type of sample.

From the analysis of variance table for three-way classification with replication (Table 1), all three of the main effects (time, type of sample, and area) are not significant. The two-way interactions are likewise not significant; but the three-way interaction of time, type of sample, and area is very highly significant. In other words, the time of sampling, the type of sample (whether GA or BZ), and the location sampled, when considered individually or in any combination of two (i.e., time and type of sample, time and area, and type of sample and area) are not important and have no effect. However, when the interaction of all three variables is considered together, the results are important. In addition, the replication or within sample factor is not significant, which indicates close agreement among repetitive samples.

It may be concluded from the analysis of variance that, in plants where many types of operations are performed, some continuously and others intermittently, and exposure times and contamination levels are variable, exposures analyzed as time weighted average exposures most nearly represent the true exposures of the workers. These exposures should be derived from both types of samples, BZ and GA, with replication at different representative times.

Another analysis of variance, shown in Table 2, was performed on data obtained in a rolling mill where the operation was continuous and essentially the same from day to day. Several locations were selected, and repetitive GA samples were taken at 30-min intervals over a period of 2 to 2½ hr. From the analysis of variance table it can be seen that the main effect of time was not significant, however, the location factor was very highly significant. It can be concluded from this study that the important consideration, when sampling a continuous operation with little change in air turbulence, is the selection of the sampling location. This is extremely important for proper

Table 2

Analysis of Variance Table

Source of variation	Sum of squares	Degrees of freedom	Mean square	*F* ratio
Time	35,975.46	3	11,991.81	0.76
Location	7,741,590.46	23	336,590.89	21.2
Residual	1,093,454.04	69	15,847.16	–
Total	8,871,019.96	95		

Table 3

Analysis of Variance for Two-Way Classification With Replication

Source of variation	Sum of squares	Degrees of freedom	Mean square	*F* ratio
Among sites	1664.96	4	416.24	2.42
1 vs 2-3-4-5	1311.75	1	1311.75	7.61
2 and 3 vs 4 and 5	0.02	1	0.02	–
3 and 4 vs 2 and 5	346.18	1	346.18	2.0
2 and 4 vs 3 and 5	7.01	1	7.01	–
Among times	97.56	2	48.78	–
Interaction	1419.35	8	179.89	1.04
Subtotal	3181.87	14	–	
Within treatments	2585.03	15	172.34	–
Total	5766.90	29	–	

evaluation of exposure when relying solely on GA samples. The samples obtained can also be considered truly representative of the locations sampled, since the concentration does not change significantly from hour to hour or from day to day if the absence of sudden bursts caused by operational failure is assumed.

An analysis of variance for a two-way classification with replication was performed on data from BZ samples collected on various operations by two samplers operated simultaneously side by side during the collection time and replicated at different times. The analysis of variance shown in Table 3 indicated a very highly significant effect of the operation site but no significance in regard to time. The operation site variable was subjected to further treatment by the method of orthogonal partitioning of individual degrees of freedom. The results indicated that the first site of the five observed was very highly significant and was the major contributor to the significance of the sites in the analysis of variance. The conclusion from this test is that individual operation sites can be very highly significant, while simultaneous sampling of the different sites enhances the reliability of the results.

In summary, statistical treatment of the available data shows that in most cases no one type of sample will suffice to evaluate a variable occupational exposure properly. Multiple GA samples obtained at carefully selected locations by an experienced sampler in a continuous process area may be employed to evaluate occupational exposure with reasonable confidence. This is possible in a continuous operation, with little change in air turbulence, where the change in concentration within a location is small compared to the difference in concentrations between locations over long periods of time.

In a plant where various operations are performed, some intermittently, repetitive GA samples alone will not represent true exposure and can vary from it by many orders of magnitude. The use of BZ samples alone, taken at different time intervals, will also lead to an erroneous occupational exposure, much greater than the true one. A close appraisal of true exposure may be achieved by obtaining multiple GA samples in all areas normally occupied by plant personnel as well as repetitive BZ samples at all operations conducive to high concentrations. The GA sample normally will tend to underestimate an operator's exposure and the BZ sample to overestimate it, but by time weighting the average concentrations for both types of samples an operator's exposure may be closely evaluated. The data indicate that this type of treatment yields reasonably reproducible results.

Discussion of Apparent Anomalies in Lung Retention of Uranium

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There have been a number of references at this symposium to the relatively high concentrations of uranium dust that existed for some years during and shortly after the war in certain manufacturing plants. During that time two workers who died of natural causes were autopsied, and I would like to review the results.

These men were exposed to uranium in the form of the fluoride and the oxide, one man for 24 months and the other for 12. The postexposure period prior to death was 15 months in one case and 12 months in the other. The first man was exposed to an average of 17 mg/m^3, the other to an average of 5 mg/m^3. These are estimated weighted average exposures for the whole period of employment. I might add that these levels of exposure are substantiated by the high levels of urinary excretion during the employment period.

Based on the assumptions customarily used to estimate the uranium burden of the human lung, in the first case there should have been 600 μg U/g lung tissue, and in the second $\approx$56 μg/g. To our surprise, autopsy showed the lungs from the first case to contain only 0.34 μg/g, and from the second only 0.24 μg/g. This is on the one hand very curious and on the other very reassuring; it also must be explained.

I think one possible explanation is available. This discrepancy is too large to be assigned to overestimates either in the level of exposure or in the method of calculating the tissue burden. Something is operating that has not previously been taken into account, and I believe that in this instance it may be the density of the dust. A search of the older literature reveals that Findeisen performed operational analysis studies to predict the

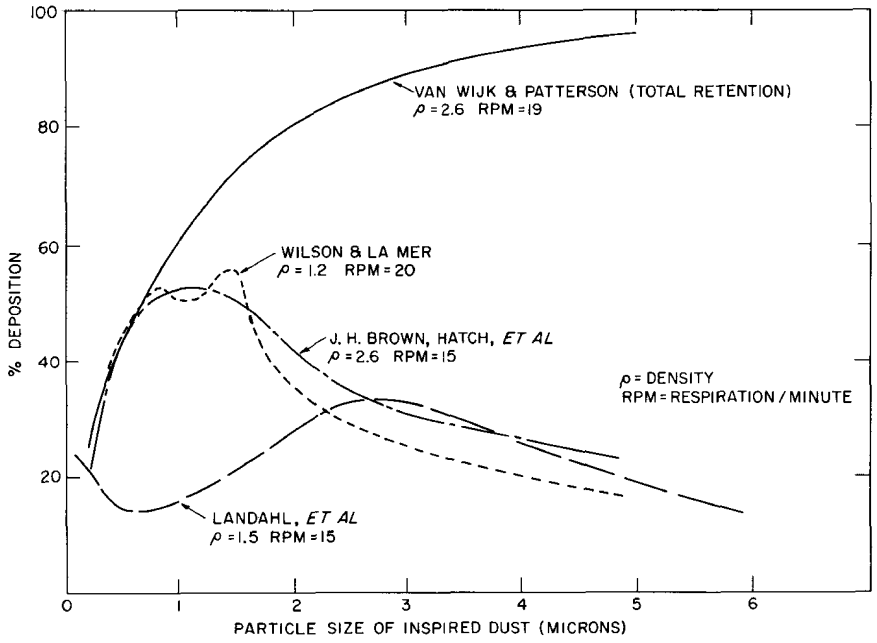

Figure 1. Pulmonary deposition of inhaled particulates as a function of particle size. The solid curve is for over-all deposition; the others are for alveolar deposition.

fate of dust entering the lung, with the calculations based on unit density dusts. More recently his analysis has been confirmed by experimental work with liquid aerosols having a density of about 1.

Examination of the mathematics of particle deposition in the lung, considering both sedimentation and initial deposition in the respiratory tract, shows that both the former, which takes place in the alveolus, and the latter are dependent on the density of the dust. This dependence is such that one can normalize for density with what Landahl called the effective diameter, which he defined as the actual diameter times the square root of the density. What is meant by the effective diameter, and how can it be used in this instance? According to the literature, plotting particle size against alveolar deposition gives the curve shown in Figure 1. Note that the peak for alveolar retention is in the region of 1 to 2 μ, and that this curve is for dust of unit density. If this curve is displaced so that it is applicable to uranium oxide dust, which has a density of about 9 or 10, the peak is displaced in the direction of the smaller particles. A 1 or 2-μ particle of uranium oxide will behave as though it were a 3 to 6-μ particle of unit density.

This being the case, alveolar retention would be very much lower than if calculated on the basis of the above curve. Actually, it has been shown that in these plants the mass median diameter was about 2 μ. Because of the dense character of the dust, it behaves as though it were 6-μ unit density material. In other words, it may not have been getting down into the alveoli at all. Perhaps this explains the findings in the cases autopsied.

A Test of Response to Exposure

H.E. Stokinger
U.S. Public Health Service, Cincinnati, Ohio

It would be very useful in the medical control of workers to have some individual test of response to exposure to a toxic material. I want to emphasize that I am talking about the *response* to an exposure, not about the degree of exposure. In the past few years we have spent a considerable amount of time in developing sensitive indicators of response to exposure, and have developed five or six. I would like to discuss one that I think may have some applicability to radioactive materials, the quantitative measurement of the amount of cystine in the fingernail. This test was developed with the help of Dr. Holaday in the vanadium workers in the Colorado Plateau, and we have had 5 years' experience with it.

The interesting thing in relation to this symposium is that the changes in the fingernail cystine in these workers correlated with urinary vanadium levels of about the same order of magnitude as the urinary uranium levels that have been discussed here. That is, in America, the changes, i.e., a lowering of fingernail cystine content, occurred at about 30 μg vanadium per liter of urine. In Peru and in Venezuela, however, in similar individuals working with similar materials, this change occurred at about 10 μg vanadium per liter of urine. Thus, this test is a fairly sensitive indicator of change when body burden has been ascertained, and I would like to suggest that industrial physicians in a position to study this test apply it to workers in other industries. Such a study would be wholly exploratory, as the test has not been applied to workers handling uranium or other internal emitters.

The test has been tried under a large variety of conditions. We know the effect on it of a number of chronic diseases such as diabetes, heart disease, etc. Studies are now in progress in different parts of the United States. We have a pretty good idea of the conditions that might affect this test.

The test has never been applied in cases of radiation exposure. It might be interesting to develop it in the atomic energy field.

Uranium in Tissues, a Case History

DUNCAN A. HOLADAY
U.S. Public Health Service, Salt Lake City, Utah

In connection with the discussion on lung burdens of uranium, I would like to describe the one autopsy we have had on a miner, which we did in 1951. It is difficult to obtain autopsies on uranium miners, but in this case we were able to get sufficient tissue samples for analysis of almost every organ.

This particular man worked underground in a uranium mine between 1927 and 1932. This was his only mining experience with anything like a significant exposure to air-borne dust. He ended his career as a foreman of the same mine, which involved being underground for only a few minutes a day; this covered a period of several years. In between he had worked as a shop foreman at a uranium refinery, which inevitably meant some exposure to dust when he was in the crushing section supervising maintenance work. He died of a combination of things, including third-stage silicosis, pyelonephritis, acute alcoholism, and a touch of virus pneumonia. According to the autopsy report, the lungs showed innumerable silicotic nodules, and the lymph nodes were completely hardened, with innumerable granulomas.

The point of interest here is that the uranium deposition in the whole lung was 0.08 μg/g. The lymph nodes contained 0.01 μg/g; the liver and spleen, $\approx$0.005. The bone burden was similar to that of the lymph nodes, $\approx$0.01.

We started to be concerned about miners when the data were reported on deposition of uranium in the human body versus that found in experimental animals. The question arose as to what happens to an individual who is inhaling a low concentration of uranium (0.3% in the ore) together with material containing a high concentration of free silica (75%). Will the lung clearance of uranium be anything like that of a person working with a relatively inert dust such as uranium oxide? Apparently, in this case at least, something had happened. The pathologist's report showed that the lymph nodes were not normal. Perhaps the physical condition of a man will have a very marked effect upon the distribution and metabolism of uranium in the body.

Urinalysis Summary

BIRNEY R. FISH
Oak Ridge National Laboratory, Oak Ridge, Tennessee*

We have been admonished to recognize the facts and to relinquish our illusions in this symposium. Some of the illusions a few of us had at the beginning may not have been completely relinquished, but they have been somewhat battered. If anyone thought there was even a moderate unanimity in the approach to urinalysis, they surely do not have that same view now. Furthermore, if anyone was of the opinion that urinary uranium might be used to measure air-borne uranium, that idea should be dispelled also; and if anyone believed that a single sample would give a good estimate of body burden, that impression too should be shaken.

In spite of the fact that the interpretation of urinalysis data is not simple, some method for monitoring individual exposure is considered necessary by a large number of operating groups. In six installations that have reported on their monitoring programs at this symposium, more than 4000 individuals are sampled by urinalysis. This represents over 1200 samples, or well in excess of 75 gal urine, each week. Samples may be obtained on any day of the week, Friday afternoon, Monday morning, etc., and either singly or in pairs, either weekly, monthly, quarterly, yearly, or at autopsy. It has been suggested also that samples be obtained after vacation. There is no clear pattern for maximum allowable excretion levels among the reporting groups. Instead, various organizations place emphasis upon different actions to be taken when an employee is found to be excreting more uranium in 24 hr than some specified amount. Action levels that have been mentioned range from 12 to 400 dis/min/24-hr sample, and at least five different radiochemical analysis procedures have been described.

Advantages and disadvantages of urinalysis as a means of measuring body burden have been mentioned. One of the disadvantages is the lack of cooperation by some employees. This could certainly become a problem, although some installations have indicated they have very good cooperation. Despite the fact that there are always a few individuals who would be very happy to confound any well outlined program, still, with good communication and adequate motivation, most people will cooperate.

Attempts have been made to correlate urinary uranium with body burden. This has been accomplished with varying degrees of success. It has been shown that it is possible to get good correlation between urinary uranium and lung burden for a case of acute inhalation of U_3O_8. On the other hand, an attempt was made to correlate *in vivo* counting estimates of body burden with estimates assuming body burden to be a fixed multiple of the amount of uranium excreted per day. The results, for a number of industrial workers, show that in many cases the estimates based upon the urine level seem to be low. This may be because deposited uranium is being released very slowly, or not at all, from certain body organs; consequently, any estimate based only upon urinalysis would tend to be low. If this is the case, it serves to emphasize the point, at which we have been hinting, that probably no one method alone is entirely adequate for estimating internal deposits of uranium.

The correlation of urinalysis data with levels of air-borne uranium met with limited success. It should be suggested that there are better means of measuring air-borne uranium than the method of urinalysis.

Snyder pointed out that there appears to be some question concerning the relationship between the animal and the human data. An uncritical appraisal of some reported observations of abnormal urine and the related uranium excretion levels might lead to the conclusion that man is several orders of magnitude more resistant to chemical damage from uranium than animals. Many of us are reluctant to accept this hypothesis blindly.

*Operated by Union Carbide Nuclear Corporation for the United States Atomic Energy Commission.

The main idea brought out with respect to urinalysis is the need for uniformity – not regimentation in the approach to the problem but, at least, uniformity in reporting to one another. Methods for estimating body burden are not at all clearly understood. Consequently, increased efforts should be directed toward establishing a realistic framework for evaluating uranium body burden.

An exchange of information on single high exposures, autopsy results, and experimental programs coordinated through a small working group has been suggested. Although the results of exchanging ideas may be expected to produce some beneficial effect upon health protection criteria, it is not suggested that the group be constituted for the purpose of recommending any specific changes in action levels. The major tasks of such a committee would include the promotion of uniformity in reporting uranium exposures and serving to facilitate the flow of valuable information.

DISCUSSION

HARRIS: I would like to ask a loaded question. In view of everything we have heard, does Mr. Fish believe that a urinalysis program in a plant processing uranium has any validity whatsoever; if so, approximately what should the frequency be; if not, can't the money be better spent for something else?

FISH: The validity of the entire program, as it now exists, depends on what you are trying to find out. If, by exposure, you mean air activity, I consider that the present urinalysis program is not measuring it. If you are interested in body burden, the urinalysis program again is not adequate. This is not to say that it should be dropped but rather that it should be changed. For detecting acute exposures, overexposures during nonroutine operations, I guess urinalysis is far better than air sampling alone; it is worth while to use it in conjunction with other methods. The measurement of body burden can certainly be improved. Whether urinalysis is better than another method or not, one cannot say; certainly the data presented at this meeting are not adequate to answer this. In my opinion, with better information we may be able in a short time (or a long time) to choose one or two procedures to estimate body burden, but at present no one procedure is adequate.

BERNARD: I subscribe to the view that uranium just doesn't belong in the body; and at this meeting we have heard that urinalysis gives a low estimate of body burden. Dr. Neuman corroborated this, but pointed out the convenience of this method for estimating body burden. Mr. Burkhart also confirmed the low estimate of body burden. I believe that urinalysis is a good and convenient method to estimate body burden, and that it should be maintained. As to frequency, I favor two samples a day, because these would show the uranium coming out of the body and can be analyzed accurately, and I believe with this sampling frequency, urinary uranium can be related to body burden.

HARRIS: We have heard two opinions: one, that urinalysis gives a reasonable estimate of body burden; the other, that it is of value to estimate acute exposure. On this basis, two samples a day, as suggested by Mr. Bernard, should be of value; but I wonder whether in fact they would. It has been definitely shown that the amount of uranium in the urine drops very rapidly after acute exposure, so rapidly, in fact, that after an exposure at 9 A.M. the amount in a 4-P.M. specimen would have dropped by at least one order of magnitude compared to that in an immediate (9-A.M.) specimen, which would show a very high uranium content. How, then, without continuous sampling of all workers, can one establish what the exposure was at the time of overexposure? It does not seem to me that urinalysis is giving us very much useful information.

Also, it has been shown that no single sample is really valid because it may be contaminated, or there may be an analytical error, or there may be a high result due to a recent overexposure. It is not known whether any single high result means that the man had a high exposure or whether the sample was contaminated; therefore, another sample is taken a day or two later, but by then the uranium content is lower by three orders of magnitude.

As far as estimation of body burden is concerned, I think it is true that some correlation can be found under certain circumstances. On the other hand, if a man is exposed continuously at his job, is it not necessary to wait a significant period of time before taking a sample to estimate body burden?

BERNARD: In the Rochester patients there was no significant decrease in the excretion rate in a

period of 10 hr. In 24 hr it probably drops from 10%/hr down to about 2%/hr; maybe by a factor of 5. I think the sample taken in the morning will indicate the contamination level, whereas the sample taken at the end of the day will indicate a recent exposure. That is why there should be two samples a day.

BROBST: Perhaps we should define two different types of urinalysis programs. One would be for the purpose of learning more about the metabolism of uranium in the body in the field of scientific research. The other would be an entirely different type of program with entirely different requirements, for an operating plant where the urinalysis program is primarily another means of estimating the effectiveness of control procedures in the handling of uranium and the control of concentrations. These are two different things and should not be lumped together in trying to set up standards.

Air Sampling Summary

W.B. Harris
Health and Safety Laboratory, US AEC, New York, New York

It is fairly obvious that the type of operation will strongly influence the type of air sampling, if air sampling is to be done at all. With a totally enclosed operation going at a uniform rate, such as a gaseous diffusion plant, the only requirement is that the general air be sampled, because there is no such thing as a breathing zone. General air samples, by and large, will give a measure of the atmosphere being breathed by the men. In the same type of plant there are unusual operations connected with maintenance, taking down equipment, etc., so variable that each must be evaluated as it is done. The general air sample in such a situation, according to the information presented, would not be adequate to measure the exposure; and some sort of localized sample must be taken at a point where the atmosphere is representative of what the men are breathing, in addition to any other samples taken.

For an enclosed operation that is not uniform, but varies from day to day, the general air sample, if taken at the proper place, can probably give some measure of what the men might be exposed to. However, it is very difficult to get proper placement, so that general air sampling in totally enclosed operations is at best of doubtful value.

It may be that to evaluate the air concentration for operations that are likely to produce sudden extremely high levels, air sampling equipment must be operated around the clock. On the other hand, it is not certain from what we have heard here that it is necessary to analyze the atmosphere in the case of a single acute exposure; probably interesting data can be obtained from urine samples. We have often done this, and we have some good curves that duplicate each other fairly well. I think we can now say that after a high exposure uranium will be excreted in the urine, and we can predict fairly well how much there will be at the end of any given time.

A sudden acute incident is almost always obvious when normal uranium is being handled, but this is not true for enriched material. An acute incident involves exposures of many mg U/m^3; this is the range for significance. I don't know of any incidents at levels approaching this that were not fairly obvious to the plant management. After these exposures a series of urinalyses is invariably done, and invariably it gives good curves; but we cannot now say what the value of these curves is.

Air samples are useless in such situations, because the statistical likelihood of an air sampler being in operation at the right place at the right time is relatively small. (Again, I am excluding enriched material.) On the other hand, if operations are out in the open (in accordance with normal chemical industry practice) and are uniform from day to day, general air samples are usually adequate to indicate the exposure. If a worker is required to be relatively close to the operation, so that his proximity has a strong influence on the concentration he breathes, the only useful air sample must be a localized breathing sample. Where operations vary considerably with time, as in ordinary chemical practice, no estimate of exposure can be made from any one kind of sample, and a rather extensive program of air sampling must be undertaken.

Any program of air sampling must necessarily be spotty; the sampling will not all be uniform, and all exposures will not be indicated with uniform reliability. Chances are that sampling will not be in progress during an unusual happening, particularly if the air is contaminated only in the immediate breathing zone of the operator. We believe that we can approach reasonable estimates with a combination of samples, but they are no more than reasonable estimates.

As far as establishing uniformity of procedure is concerned, one must take into consideration the fact that different situations present different problems. In gaseous diffusion plants the problem is very different from that in chemical refineries, and both differ from the problems in small experimental and developmental laboratories, so that there is bound to be a significant variation in the

over-all procedures. Another point to consider is that each organization in this field has some vested interest in maintaining its current procedure, whether this be in trained personnel, in the type of equipment on hand, or whatever. Even if we could determine optimum procedures, these would have to be varied from plant to plant. However, I think much of the discussion at this symposium indicates that there are areas in which uniformity would be desirable. It may be that as a result more effort will be put into elutriating the sample in some manner to remove the particles thought to be nonrespirable, so that only the material most likely to deposit in the lung will be sampled. I would like to see this, and we will work on it in our laboratory.

The next step is to set up procedures for evaluating air concentrations, and I am not sure this is possible. We may be able to decide that for a general air sampling program to be useful, the concentrations should be in a certain range, or that a program of determining average exposures can be carried out only in certain circumstances; but we must recognize that different types of establishments have different problems, and each must have a program useful under the existing conditions.

In closing, I believe air sampling gives a good estimate of exposure. I think an air sampling program can provide a reasonable estimate of what the usual person – not each person – in an operation is exposed to. Whether he breathes the air; and whether, if he breathes it, the suspended material gets into his lung; and whether, if it gets into his lung, it does him any harm; I cannot say.

DISCUSSION

HEATHERTON: Mr. Harris mentioned that air sampling is a measure of the atmosphere being breathed, or what a man may encounter, but does not necessarily show what the man is actually taking into his body and into the lung. In our air sampling procedures, we have found pretty fair correlation in some cases and not in others. We wonder where the fault lies, in the air sampling or in the urine sampling. We think urine sampling is probably a better measure of exposure than air sampling, but we are not sure.

HARRIS: You should ask Mr. Fish. I think both methods are defective. Air sampling gives a measure of the atmosphere to which a man is exposed at the time the air samples are taken. We can make our analysis methods as accurate as we like, but there will still be periods when a man actually inhales greater or lesser amounts than shown by air sampling. A urine sample indicates what a man is excreting at the moment, but it does not indicate anything else. Part of this excretion may come from long-term storage in various organs, from which material is being removed at various rates; part of it comes from soluble material in the lung, which is being given out at varying rates depending on the individual's metabolism and on the time and degree of exposure; etc.

Thus, neither air sampling nor urinalysis can be expected to measure exposure accurately in all cases. When these two inaccurate measures are compared, they will not always give a good correlation, although they may, depending on the type of operation. If a person is exposed continually every working day at a relatively constant rate, some correlation would be expected between this rate and the urinary excretion at a particular time after exposure, but that is all.

ROSS, D.M.: It seemed to me that J.E. Ross' experimental approach to the correlation of urinalysis results with air sampling results was a very good one, and that he took into account more of the factors that affect the entry of particles into the body than did any of the other speakers. However, there was one flaw in the data, namely, that the exposures were so low that they can hardly be considered exposures. I would like to see similar studies performed in plants that admittedly have higher uranium concentrations to see whether a more positive correlation can be found.

HYATT: We are trying to justify air sampling on the basis of correlation with urinalysis; but Mr. Harris, in his report on health protection practices, pointed out that the criterion for industrial hazards is the maximum permissible air level, and this is the area of disagreement. I think that the real role of air sampling is to determine whether or not the maximum permissible level is being exceeded, and correlation with urinary excretion is secondary.

HEATHERTON: I do not advocate the discontinuation of air sampling; I think it should be further investigated. But there are situations where it is very difficult to evaluate the air exposure. Mention has been made of peak exposures due to accidents when no one is present to take air samples. Also exposures during maintenance, moving of equipment, and repair work are practically impos-

sible to establish. We think that urine sampling complements air sampling and vice versa, and we advocate continuing both.

MASON, M.: In listening to these discussions I have noted a fundamental point that needs clarification, with respect to both air and urine sampling. There are two schools of thought regarding the maximum allowable concentration. One believes that, whatever the maximum allowable concentration is, no exposure should reach this level, and has devised air sampling and urine sampling techniques in accordance with this philosophy. The other school believes that the maximum allowable concentration may be reached if it is expedient to do so, and has developed techniques to fit this attitude. I think the primary role of this symposium must be to furnish some guidance in this area; then perhaps we can develop techniques of air and urine sampling that will allow us to evaluate our findings.

SCHOEN: Mr. Mason's point is well taken. The discussion this morning has created the impression that we are pitting urine sampling against air sampling. As Dr. Neuman has said, there is sufficient justification for the use of both. We all agree in general on the reasons for taking air samples, and accept air samples as necessary to assay the environment for potential exposure.

Once we assess the environment, however, none of us to any great extent separates the portion of the air-borne uranium that is of real critical concern. We get from the air samples an assessment of potential exposure; however, it seems to me we do not go beyond the general control this gives and use air samples to evaluate individual exposure in this environment. To do this would require much more sampling than anyone is doing, and much more than it is reasonable to expect: it would require hovering over every employee continuously, creating all sorts of problems.

Therefore, I don't think we can assess individual exposure on the basis of air samples at all. This is where urine sampling has a place. Air sampling is useful for general control purposes; but, to find out whether some individual, as a result of certain idiosyncrasies – the way he works, laxity in using respirators, etc. – is being exposed not to the general plant levels indicated by air sampling but to a much higher one, urine sampling is needed.

The urine sampling program should serve more than one purpose. One is to see that the personnel in general are not being overexposed. The second is, in cases of acute exposure, to follow the excretion, by two samples a day if necessary, to determine the net effect of this acute exposure in terms of actual deposition. But to consider sampling everybody in the plant twice a day for urinary uranium doesn't make much sense.

I don't think we ought to be trying to justify either air or urine sampling; we have ample justification for using both, the first in attempting to control the industrial environment and the second in attempting to control individual exposure as an adjunct to general controls.

Medical Findings Summary

T.S. ELY
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The main point brought out at this symposium, from a medical standpoint, is that uranium is a toxic material to which many people have been exposed over a fairly long period of time, and yet, according to one viewpoint, we have yet to see the first case of injury from normal uranium. There is a fringe consideration here, namely, that of abnormal kidney function; there are perhaps a few cases of albuminuria, however temporary, that could be ascribed to uranium toxicity. This is really quite a striking observation, particularly in view of the fact that in the early days exposures were many times what we consider permissible today, not twice or ten times but up to 100 to 200 times the maximum permissible air-borne concentration. In spite of this, there has been no great wave of uranium illness.

The other viewpoint is that uranium is a highly toxic material, and this has been borne out by laboratory experiments; however, these were carried out with laboratory animals, and one wonders whether there isn't some remarkable difference between humans and laboratory animals in regard to uranium toxicity.

We have had discussions on tissue analyses from autopsy material, presented by Dr. Quigley and Dr. Butterworth, and these showed no definite injury that could be ascribed to uranium, and, in addition, showed smaller concentrations of uranium in the tissues than expected.

The Rochester experiments are quite significant, and the final outcome of the 5-year insoluble uranium inhalation study will be of great interest. It is quite striking, I think, that no definite histopathologic changes have been seen in the animals examined so far, in spite of the fact that concentrations of uranium in pulmonary lymph nodes of up to 15% have been found.

DISCUSSION

CHAPMAN: I have two questions. There are at least three groups of people at this symposium, those from service organizations (like myself), the research group, and some who are seeking information; and I am sure they are all confused. I consider that air sampling and urine sampling are both necessary, but the results cannot be carried out to many significant figures. Maybe it is only necessary to determine whether milligrams or micrograms are involved; or maybe one order of magnitude is not enough.

Several plants, such as Harshaw, Linde, and others (Mallinckrodt is the only one remaining), had some experience in the old days when some people were probably overexposed, and we need some clinical history on these people.

I think the thing that we have to determine is the point that has been raised several times, and about which Dr. Neuman said that we must be patient. I admire his courage, but my first question still is: Is there any way that uranium can injure a man other than his dropping a 100-lb pellet on his foot?

The second question involves beta exposure: Is 2.3-Mev beta penetrating radiation? How do we interpret an employee's exposure to uranium? Can we write it down on a piece of paper and give it to the man, so that he can give it to his next employer for the latter to calculate how much exposure he has left in life according to the recent formula? What is the maximum permissible body burden? And what about skin dose: can it be disregarded or should it be included?

The new exposure levels are based on genetic effects. Can the 2.3-Mev beta reach the gonads? I consider it academic whether we measure air exposure or urine exposure when the thing we are interested in is the clinical picture. We have been quibbling over numbers that to the practical man don't mean very much. What we are really interested in is whether money should be spent for ventilation or for shields or for a urinalysis program. Some of us have to answer to stockholders, some to taxpayers, and some to unions. This is a serious practical problem which the research

people do not consider when they are quibbling over these numbers out to three or four significant figures. The International Committee on Radiation Protection, which does not have to answer to stockholders, unions, or taxpayers, but only to a group like this, has set up these numbers, and they require practical interpretation.

BERNARD: Some of us believe that the damage is due to the amount of uranium in the body; that is why we argue about how to estimate the amount of uranium in the body.

MASON, M.: I think the question concerns what the damage is.

NEUMAN: I have been accused of being courageous; I think it would be courageous to say the MPL is too low; it is not courageous to defend it. As I understand the MPL, it is the maximum exposure that is considered not to harm anyone, further reduced by a factor of 10 because of our great uncertainties. Therefore, an exposure below the MPL should not injure anyone, particularly in a group as small as 4000. The MPL for the population as a whole is reduced by another factor of 10 because of the variations in the reactions of the individuals in so large a group. At this level there should be no injury to anyone in the population as a whole. I cannot understand why everyone is surprised that people are not showing bad effects from uranium exposure, since that is what we have been trying to prevent.

Also, do you want to say to your stockholders that you are running an economical plant because you are not spending money for these precautions, and are consequently injuring so many individuals each day – or each month or each year? Is that proof of an economical operation? This decision between human lives and damage, and dollars and cents, is difficult. I, for one, will help you defend your precautionary actions against any stockholder who chooses to object.

QUESTION: It seems to me that several types of injury have been mentioned. Dr. Snyder spoke of kidney injury, and it seemed that the injurious concentration in terms of μg U/g kidney for different species might be the same, but that perhaps some species have to take in much more uranium to get that concentration in the kidneys. Maybe this depends on the solubility of the uranium. This brings up the practical question of whether one MAC can be set for both soluble and insoluble uranium.

What were the symptoms in the individuals on whom information was collected? It has been said that there were no symptoms found in certain individuals exposed under certain conditions in the past; but symptoms have been found under certain experimental laboratory conditions. Is it possible that these laboratory symptoms might be found under actual conditions?

I would particularly like a résumé of the type of damage found in the individuals in the Boston hospital who were intentionally exposed to uranium. Since these were brain cancer patients, certainly not in good health, what conclusions can be drawn from such cases?

SNYDER: I am not a medical man, and I cannot make a definitive evaluation of any of these medical data; rather, I am seeking guidance in interpreting them. The data that I presented, except for a very few of the numbers at the end, were animal data. I am not competent to tell you exactly what the symptoms were. I tried to pick out exposure levels that were most representative. There were higher levels of exposure where severe effects were found. There were levels lower than those I discussed, where it was very difficult to decide that there was no evidence of damage. At the intermediate levels, damage was fractional. I did this realizing my incompetence to assess the medical data and in the hope of getting competent people to do so in order to give us a more precise estimate of the various factors that enter into a determination of the permissible level. These factors are numerous, and interpreting them will not be easy. We need to know at what level of kidney damage we are willing to set the permissible level, or by what factor we should stay below the level of any kidney damage.

At this symposium two entirely different kinds of damage have been discussed. On the one hand, no worker has been killed or had symptoms so severe as to be apparent on casual inspection. I feel that an exposure level is too high if it is based on the absence of severe symptoms or the appearance only sporadically of any symptoms; to consider such a level safe is to take a considerable and unwarranted risk. This is my personal opinion, and it is true that there are other risks of greater magnitude in everyday living, but this is a question of values outside the present discussion. But I cannot be convinced, without detailed follow-up of the individuals exposed at high levels, that no severe injury will ever be found, and its absence up to

now is not a valid reason for considering such levels permissible. I think that would be a dangerous attitude, even though I cannot evaluate the expectation of such symptoms appearing. On the other hand, in the laboratory experiments detailed examinations are made of tissues on quite a different level, and an entirely different sort of damage is being studied.

Someone must decide what degree of damage we are really trying to prevent, the very slight appearance of a kidney defect or severe damage that is clinically quite apparent. The underlying philosophy of the NCRP and the ICRP tends towards the former. These groups try to set limits below the level at which any detectable damage will occur. The exact determination of this level is a matter for careful consideration by medical people. The same remarks apply to practically every other parameter with which we are concerned. More precise information should be obtained about the effects of such things as particle size, chemical form, manner of exposure, and dose rate. The data indicate that the body burden is not directly proportional to the exposure level. Further study is needed before a maximum permissible level can be definitively determined.

As it stands now, in view of the many uncertainties, I think that Mr. Bailey's philosophy is that the maximum permissible level is set at a level where we hope that no severe effects will turn up later to embarrass us. The level cannot be given in precise numerical terms. On the other hand, many of the questions about the specific parameters can be answered much more precisely than they have been by both experimenters and practical men. Then we will be in a position to say what limit we are willing to accept.

A question was also raised about the Boston patients. I have been told that every one of them showed severe kidney damage, though I do not know the exact details. Of course they were brain tumor patients, and this must be taken into account in interpreting the data. Most of the human data will be subject to such uncertainties, probably as long as we operate.

EISENBUD: The people who have to discuss economics with stockholders are not callous, and they do not want to subject the industrial workers to any risk. We are not challenging the maximum permissible concentration, *per se*, but I think it would be presumptuous, after 15 years, not to permit a re-examination of the assumptions that went into the calculations of the maximum permissible dose. The figure used to govern the maximum permissible air concentration is based on experiments with lower animals, and is based on assumptions concerning the amount of inhaled dust deposited in the alveolar tissues and the rate at which it is cleared from these tissues. These assumptions should be examined in the light of the new data. I think the deposition of dust in the lungs is lower in the experimental animals, and there is evidence that the lung clearance is somewhat more rapid than was assumed. The fact that autopsies on men exposed to uranium show only a very small fraction (perhaps $<1\%$) of the amount which calculation says should be present in the tissues necessitates a re-examination of the calculations and the factors introduced many years ago, which have not been changed on the basis of data accumulated since.

At present there are not sufficient human data for us to be able to say that, because some men exposed to 78,000 $\mu g/m^3$ did not get sick, the maximum permissible dose can be raised by a factor of 100. No one is saying that. But, considering that the present limit is 50 $\mu g/m^3$, the accumulation of new evidence changing some of the factors that went into the calculations furnishes a good reason for these calculations to be re-examined.

HOLADAY: Our primary mission is to protect the workers, but, if by so doing we make a process inoperable, that is no solution to the problem. I agree with Mr. Eisenbud that the question concerns not so much the level of the MPC as its validity. On what assumptions were these levels based? And how do they stand up with experience? As I pointed out earlier, the only way to answer some of these questions is by an epidemiological study of people exposed 10 to 15 years ago. This is a long time in the history of a toxic exposure, and, considering that the exposure levels ranged from fairly low to rather high, these few hundred people should show us something. I think such a study is necessary.

As Mr. Eisenbud said, these permissible levels have been set on the basis of the calculated radiation dose to an organ, e. g., the lung. There is probably some correlation between the calculated and the true dose, but I do not know what it is. There is certainly not an exact equivalence because of the various assumptions used. Therefore, the difference between levels of, e.g., 50 $\mu g/m^3$ and 250 $\mu g/m^3$ is for all practical purposes insignifi-

cant considering the various errors in the assumptions. People experienced in this field are aware of this low degree of exactitude. It is the inexperienced person, following the procedure of a factory inspector, who says that 55 or 75 $\mu g/m^3$ is bad, not realizing that it is no better and no worse than a figure of 40 $\mu g/m^3$, or for that matter 100, as far as estimating the true situation is concerned.

I would like to see the threshold level for uranium treated in the same way as threshold levels for other toxic materials, as a guide to an estimation of the situation with other factors also taken into account. An understanding of the fuzziness of the figure will allow it to be properly used for the present, until such time as epidemiological data become available. I think there is good reason to believe that no one will be hurt by the use of the present MPC as a standard threshold level, rather than as a fixed "'good" or "bad" reference point.

SESSION VI

Panel Discussion

M. EISENBUD, *Chairman*,
AND
E.C. BARNES, T.S. ELY, H.F. HENRY, E.C. HYATT,
W.F. NEUMAN, J.A. QUIGLEY, AND E.C. WATSON

EINSENBUD: This is not the first time an attempt has been made to dispel the mysteries surrounding the hazards of using uranium. About two years ago Professor Drinker assembled enough material to fill the July 1956 issue of the *A.M.A. Archives of Industrial Health,* most of which was reproduced from papers presented in Geneva in 1955. I would like to refer to two papers in particular. One was entitled "Industrial Hygiene of Uranium Processing," by Dr. Quigley and myself, in which the work of many investigators was reviewed. In the summary we stated:

"There has been sufficient experience with severe exposures to both soluble and insoluble compounds of uranium to permit the conclusion that uranium has a low order of chemical toxicity in man. Many of the nonradioactive heavy metals, such as lead, arsenic, and mercury, would produce very severe, perhaps fatal, injury in the lungs at the levels of exposures reported here."

The second paper was by Dr. Hodge, "The Mechanism of Uranium Poisoning," in which he stated:

"Uranium is one of the most toxic elements chemically, more toxic than arsenic or mercury."

This was two years ago, and it was, I believe, the first attempt to bring together scientific views on uranium toxicity. These two articles side by side must have been a source of confusion to the readers. This is a good point from which to begin the present discussion, because this conflict between the findings on humans over a 15-year period and on experimental animals over a somewhat longer time continues to pervade this symposium. I would like to begin the discussion by asking Dr. Neuman to comment on these inconsistencies.

NEUMAN: I am reminded of the saying that the same cup may look half-full to one person and half-empty to another. I think a large part of this seemingly big difference in opinion is a matter of viewpoint. These meetings seem to be focusing on certain aspects. The experience of people in the field indicates no evidence of injury in exposed persons, and this is taken to mean that animal data probably should not be extrapolated to man. I take the perverse viewpoint that the maximum permissible concentrations were derived from animal experimentation, and, although they may be too stringent, they have proven reasonably safe; and I wonder if at present we have enough evidence to raise or lower them. I think the maximum permissible concentration should be kept under constant surveillance, and our assumptions should be re-examined from time to time. I think we do have some evidence that may indicate that the present regulation may be too strict. On the other hand, in the animal data the appearance of tremendous accumulations in the lungs of dogs raises the question of whether, on a radiological basis, the limit is low enough. We do not find, at least in the evidence available so far, any real accumulation of uranium in human lungs. This is a problem that must be resolved.

This symposium has brought out the need for epidemiological study of those persons who have been exposed. People did not think radium was very toxic until they examined the patients, and the more they examined the patients, the more toxic it was shown to be. This would not be readily apparent at an early date.

It must be remembered that there are several kinds of damage. Uranium has a nephrotoxic effect, but the kidney has a tremendous power to make new cells and repair itself, particularly without loss of renal function. I think the sensitivity of humans to injection was found to be approximately of the same order as that of animals, with injury beginning at about the level expected from the animal data. There may be many differences in terms of lung damage, but I think, on the basis of what has been said about nephrotoxic effects, delayed effects, etc., that we should be aware of our

uncertainty: the present limit may be very safe or may not be safe at all.

EISENBUD: I think it might help solve the problem to consider the nephrotoxic effect on one hand and the radiologic effect on the other. Can we agree that the nephrotoxic effects of uranium are not of as great concern in man as they seem to be in the experimental animal?

QUIGLEY: I do not feel that it is nearly as severe a problem as we have been told. I consider a uranium plant one of the safest places to work; I would not say that of radium. It seems that the toxicity of uranium is not as serious in man with respect to the nephrotoxic effect.

EISENBUD: In the case of kidney poisons, can one rule out the latent effects? If a man worked for 10 years in the uranium industry and left with healthy kidneys, are his kidneys likely to show damage from uranium exposure 10 to 20 years later?

QUIGLEY: I would think not.

NEUMAN: I would be surprised, but I certainly would not exclude the possibility. Up to the present we have not had sufficient kidney examinations to know whether such people have been damaged.

EISENBUD: I would like to return briefly to the question of radiological hazard to see whether we can find any area of agreement on that. To me the significant thing is that we are not challenging the validity of the original calculation in the sense of saying that an exposure limit of 300 mrem/week is too safe and should be raised. What we are saying is that uranium is not being found in the tissues in the amounts originally anticipated, and therefore a given air concentration corresponds to a much lower tissue dose than we previously thought. Would Dr. Ely comment on the significance of the low autopsy findings reported in humans?

ELY: I think that is the crux of the matter. I don't think we are endeavoring to or are in a position to argue about the radiological levels set by the various committees. The only possible point of discussion concerns the other parameters such as the lung retention, and there are others here more qualified to speak on that matter.

I would like to say, however, that I don't think we have settled or even come close to establishing an agreement on the nephrotoxicity of uranium or mercury or lead.

Would Dr. Neuman comment on the difference between the two articles previously mentioned? I gather from what I have read and seen that, according to Quigley and Eisenbud, mercury, for instance, is considerably more toxic than uranium in equivalent concentrations.

NEUMAN: One must separate the theoretical from the practical in this case. On theoretical grounds I am not sure I would be justified in making any such statement. But in terms of plant experience such a statement is perfectly safe; and if these gentlemen find in terms of plant practice that uranium is easier to handle, with less danger of renal effects, then this is a practical observation with which I would not argue. But then, I cannot say that uranium is not a nephrotoxic agent, or that when injected into animals it is not more toxic than mercury, because we have seen that in animals it leads to nephrotoxic effects.

EISENBUD: The plant population Dr. Quigley studied was, for a considerable period of time, excreting uranium in the range of 1 to 5 mg/l. According to my experience, people excreting mercury at this rate would have been ill. Mercury is excreted at these levels only in cases of severe chemical nephrosis. It was actually quite astonishing to see this group excreting uranium at 1 to 5 mg/l with otherwise negative urine findings.

BERNARD: I saw the paper by Quigley and Eisenbud and the cases reported on. With respect to the first Geneva paper mentioned earlier, we made some calculations from the Boston data to determine what the kidney damage would be. Using the parameter of the Boston patients, we calculated that they had 2 to 3 or 4 μg/g in the kidney, and that this should cause essentially no kidney damage. Dr. Quigley did see some abnormal findings. My point is that it took more than 2 or 3 μg/g, based on the Boston patients, to produce this small amount of damage.

QUIGLEY: Since the collection of the data presented in that paper, we have operated a plant where for quite some time the exposure was to uranium hexafluoride, and we had many high exposures. In spite of this, from data obtained "after-the-fact," we did not find any evidence of kidney damage.

BERNARD: Were the exposures determined from air measurements rather than urine samples?

QUIGLEY: No, these were urines.

BERNARD: High urines?

QUIGLEY: Yes. This is the only measurement we could make, since normally the air is not being sampled when an accident occurs. We did do air surveys, but in the type of hex operation we had they were not very meaningful because nobody was present during most of the episodes. We had as much as 27 lb hex gas released into the air near a group of workers. We have had >1 mg U in the urine in more than one case, and we have not found any clinical kidney abnormality.

BERNARD: Were these data published?

QUIGLEY: No.

HARRIS: I don't quite understand the point Mr. Bernard was making. The data I have seen on the Boston patients indicate that the one injected with 97 μg/kg (≈5½ mg U) was excreting uranium at the rate of 280 μg/hr after 2 hr, which comes to ≈6 mg/day, and apparently showed no kidney pathology. Another one, injected at about twice that level, showed some positive findings, i.e., a trace of albumin, but everything else was negative; and a third at an intermediate level showed no symptoms whatsoever. A fourth and a fifth patient, at 170 and 280 μg/kg (11.5 mg total each), excreting uranium at the rates of 1 and 1.1 mg/hr, respectively, showed positive symptoms. I don't understand the implication of 2 to 3 μg/g.

BERNARD: I think you are quoting from Dr. Luessenhop's paper. One tenth mg uranium per kg body weight is supposed to produce a definite nephrotoxic effect. With reference to the Boston patients: if you can show that ⅓ of the body uranium is in the kidney, then ⅓ × ⅒ mg U × 70 kg body weight divided by 300 g kidney tissue gives ≈6 μg per g kidney. Note that this is based upon the tolerance value of ⅒ μg/kg body weight.

BUTTERWORTH: I think in talking about nephrotoxic effects some of us are trying to draw conclusions from small numbers of samples. No two kidneys are alike and no kidneys are necessarily normal: the kidneys that have responded to uranium could very well have been injured previously by some other nonoccupational cause. These are some of the factors that tend to cause variations in response to uranium in the kidney.

Over the years I have come to the conclusion that it does not appear to be so much the single large exposure resulting in a large uranium excretion that affects the kidney (and the only effect I have seen is transient albuminuria) but rather a prolonged exposure, even at such low levels as to cause excretion of a few hundred μg/l urine (in contrast to several mg/l from a single exposure), that causes albumin to appear in large concentrations. We have evidence to support this view.

With reference to the radiologic effect of stored uranium on the lung, I think we should not be misled when trying to draw conclusions from our findings of the amount of uranium present in post-mortem material. If the uranium is turned over (i.e., removed and replaced) rapidly, only a small amount will be present at the time of post mortem although the total amount that has gone through the lungs could be very considerable.

BAKER: I would like to ask Dr. Neuman at what level of air exposure he could see effects in experimental animals without sacrificing them. In other words, did he look for symptoms as we do in plant employees?

NEUMAN: There are several theoretical points under discussion simultaneously. We are assuming that there is a nephrotoxic effect. I admit that there might be, but I am not certain. The kidney has tremendous reserves: one kidney can be removed (this is done routinely) and the other can handle renal function adequately. This organ also has a tremendous recuperative power.

I was always very much impressed by the results of histologic examinations of animals under chronic exposure. They initially get very sick, and about the seventh day they look very bad; yet under the continued assault of the same level of exposure, they gradually perk up, and by the end of the year they seem quite well. The histologic pattern shows the same thing. At the end of the first week there is serious renal injury, easily demonstrable, but there are also signs of repair, and, as the year progresses, there is a pattern of simultaneous injury and repair. These animals were not sick after the second or third week; they would not present a clinical problem; yet, they were undergoing damage.

The criterion for any kind of limit in terms of nephrotoxic effects is to choose air values that would not result in easily demonstrable histologic changes in the kidney. These limits are far below those at which functional loss of renal capacity could be seen.

QUESTION: Was this a tubular effect or glomerular?

NEUMAN: The glomerulus seemed relatively unharmed. If tubular function is interfered with, glomerulose involvements are often found, but the initial attack and initial effect is to the distal part

of the proximal tubule, where we suspect the change of *p*H occurs.

SNYDER: The calculations for the limits in *Handbook 52* are supposedly for a 50-year period of exposure, and formerly were for a 70-year period. Would Dr. Neuman comment on the effects of such long exposure periods as compared with those of a year or 15 months, which I believe was the duration of most of the experiments. I realize we don't have the evidence to give a definite answer, but I would be interested in an opinion as to whether the effects might be more severe under full lifetime exposure.

NEUMAN: You said it was impossible to answer, and I agree. I think it is safe to say that we all hope the long-term record will be clean. That is the reason I said the other day that we must have patience. We have made good progress; it seems we have set levels that are not inoperable. It might be less expensive to be dirtier, but I think we owe it to our people, since we have not had a generation of experience, to keep the levels on the safe side. Maybe there are effects we have not observed yet.

EISENBUD: I am certain that many times as much money has been spent on the study of the toxicology of uranium as on that of all other toxic metals. When the experiments become so elaborate and so well controlled as in this instance, one begins to question, as, for example, Dr. Neuman does, whether or not there is a threshold. We could follow customary toxicological practice and conduct experiments that could only pick out effects on more than a few percent of the population, as is the case when we label most things as "safe." On the other hand, I don't think we can set two standards.

QUIGLEY: I don't feel I can summarize, but I can briefly describe the changing concept of industrial medicine. In the early days industrial physicians were faced with reparative problems such as cut off hands or severe lead poisoning, and they were interested in trying to cut down a very definite human loss that was easy to measure. The second stage was trying to make early diagnoses so that workers could be removed from exposure before they received a fatal dose of toxic material. We are now in the third stage, in which we have not done too well, that of prevention.

I am not saying the present levels should be changed; they have been so well established that we wonder whether they are too low. We have not seen any death or disease that can be attributed to uranium. It is only natural for the health officers at the plants to be concerned about this, especially since they also have to consider costs. I am not suggesting that the levels should be lowered. I think we should accumulate information that can be analyzed, better than what we have now, with the thought in mind that in a few years it might be possible to change the standards.

CHAPMAN: There is also a question of semantics. Back in the days of the Manhattan District we talked about tolerances, and then the industrial hygienists changed the term to maximum allowable concentrations. In the field of organized labor we are running into another problem. We consider the limit as a value below which we are safe, but organized labor considers it as a value above which they are unsafe, and, rather than seeking correction of an above-limit condition, they seek an economic benefit every time it occurs temporarily.

EISENBUD: I think it goes without saying that the big plants are already built, the equipment is installed, the procedures are established, and the industry is meeting the present levels; and I don't think anybody really expects any major change in the levels until we have had another 10 years of experience at least. I think another 10 years will probably be adequate to decide whether any change should be made.

With respect to the radiological hazards, there are a great many safety factors that we might review briefly. First there is the over-all safety factor of 10 which Dr. Neuman indicated is applied as a general procedure, and there are others included in the calculation which I think should be brought up for consideration. With reference to this factor of 10, some biologists consider it much too large and think a factor of 2 or less would be adequate. Also, we are using as a basic criterion that the lung should only be exposed to 300 mrem/week. This is derived from the maximum permissible whole-body dose, which is based on the most sensitive tissues in the body, e.g., blood vessels and genetic tissue. When the dose is lowered to protect the genetic tissues, the question comes up of whether or not one ought to lower the dose to, say, the lung tissue. One could, for example, apply the same values for bone, which are based on radium. Thus another factor of 10 is introduced. The rbe brings in a factor of about 3, in that 300 mrem/week is used although 1000

mrem/week could be justified by analogy with radium. This brings the factor up to 300; and it can be brought up to 1500 in this way. The safety factors are very large. I have not mentioned them all, but we should not lose sight of the fact that they are built into the calculation.

QUESTION: Is it true that the factories are all well equipped? How about the mining and milling industry that have to follow Section 20?

EISENBUD: The mining and milling industry is obviously not as well equipped, and many practical problems will have to be faced.

Would Mr. Barnes summarize the small area of agreement with respect to our present evaluation of the hazard of uranium compared to that of lead, mercury, etc.?

BARNES: I don't feel particularly qualified to answer that question, but my experience in the industry over a number of years shows that at comparable concentrations lead and mercury present a greater problem than uranium.

Returning to the subject of safety factors, I think another point should be made: in many instances people really misinterpret the significance of the maximum permissible concentration by taking it as a literal maximum rather than as a maximum *average* permissible concentration. I am sure that in many cases another factor of 5 or so is thrown in because of this interpretation of the numbers actually used. The limits are based on averages over long periods of time, not on instantaneous concentrations that may occasionally be present, and the actual average is perhaps ⅕ of what these temporary maximums may be.

EISENBUD: I think we would all agree that we are more relaxed about the toxicity of uranium than we were 10 or 12 years ago, and that is important. As long as we are not too worried about it, I think we can maintain the *status quo* for another 10 years.

Concerning urine sampling, I had the feeling this morning that too many people are trying to make a science out of this; I think it is more of an art. Mr. Hyatt, what would your recommendation be after hearing the discussion this morning?

HYATT: I am really not qualified to answer.

EISENBUD: I purposely asked you because I don't think this question should be answered by a specialist, but by someone who has to use the results and be guided in his work by the laboratory reports.

HYATT: It has been suggested that perhaps urinalysis has no value. I can't agree. We wouldn't think of operating a plant without a urinalysis program. Whether we will make it into a science is another matter. First, I think we should consider normal uranium and enriched uranium separately because we are more concerned with the chemical toxicity of the former and the radioactive effects of the latter. I don't think anyone can argue that urinalysis is a very useful tool to indicate exposure. We are not attempting to estimate body burden. I was interested in Mr. Burkhart's methods for enriched uranium. We tried them but did not find anything. Normally service groups like ours make no attempt to estimate body burden accurately because at this point I don't think we know enough about it.

BERNARD: You said you didn't get anything from applying these methods. What were you looking for?

HYATT: I think that the main reasons we didn't get useful results are that we have not followed the program as closely as you have, and the Research and Development Laboratory exposures are not as consistent as yours because our men are removed from exposure for several months at a time.

BERNARD: The only useful result is the identification of "high" and "low" people, so that the "high" ones can be temporarily removed from exposure.

EISENBUD: It would be useful to discuss this problem quantitatively. If urinalysis is being used as a criterion and everyone in the plant is excreting <10 μg/l, this is worth knowing; and certainly the frequency of survey at such a plant would not be the same as at one where the results are scattered from 5 to 100 μg/l. It has been my observation in industrial hygiene practice that the air sampling, the ventilation system, and the urinalysis program are all related, through a kind of feed-back, and that the well-managed program is balanced. It is useless to analyze the urine once a week, year after year, if only 10 μg/l is found consistently; in such a case monthly or bimonthly sampling would be sufficient.

HENRY: I have listened with some amazement to the descriptions of some of these programs. Since I was originally a physicist, I lack the industrial hygiene background and the familiarity with lead and mercury hazards, but I do know that in the field of physics the main search is for

truth. What are the facts? No one has said that raising the permissible limit will kill everybody; however, I think we need some definitive information.

Urinalysis is probably one of the things about which we will never have really definitive information, and without this it cannot be related to body burden. How is body burden to be defined? One could say it is what somebody has when he quits his job. The urinalysis results presented seemed to indicate that uranium is excreted by humans at a much faster rate than expected. The amounts of uranium found in the tissues after death have also been lower than expected. The only categorical statement that can be made about urinalysis is that a man who is excreting no uranium has none in his body.

Perhaps statistical analysis can give us a safe upper excretion limit, but the data I have seen do not seem to be leading to a good statistical figure that can be applied to a large plant population, if it is not known what each worker is doing at all times and what exposure he is receiving, i.e., how much, where, when, and in what form. As far as quantitative results are concerned, up to now the range of error in the data has been too large.

BARNES: We should separate in our thinking the two elements that seem to be continually intermeshed in our discussion, namely, the use of urinalysis for evaluating plant exposure, and its use for measuring body burden, i.e., the amount of uranium that has been absorbed and stored in the body. If urinalysis is to be used for evaluation and control of plant exposures, a plan of sampling and limits of accuracy must be set up. For the determination of body burden many tools are available, of which urinalysis is only one. In fact the use of urinalysis is the only link between these two problems, and they should not continually be confused on this basis.

HARRIS: Two of the previous three speakers seemed to indicate that the use of urinalysis has very little merit in establishing body burden of uranium. One of them said that it was useful in establishing plant exposures. If this is the case, what would be the best urinalysis program for evaluating plant exposures?

QUIGLEY: I think that would depend on the type of plant and the length of time it had been in operation. In a new plant, where the equipment is being tried out and changed on a day-to-day basis, urinalyses are probably much less meaningful than in a plant with established procedures. During the early days, when the exposures would tend to vary a great deal, more frequent urinalysis would be advisable to indicate how things were going. As the operation becomes stabilized, fewer urinalyses (and fewer air analyses) would be satisfactory.

HARRIS: Could you be somewhat more quantitative?

QUIGLEY: In our plant we did four complete air surveys a year when operation started, but we did not do as many urinalyses as I feel we should have. We now do a complete air survey in each part of the plant once a year, and resurvey areas showing high results. This year we plan to get four urine samples a year from the production men, and possibly a fifth at the time of vacation, rather than weekly samples as we were doing at one time.

HENRY: This seems to mean that the frequency of the urinalysis program depends on how it fits in with the over-all control program. One of the basic criteria for determining the condition of the workers is that they will not be excreting more than some average amount; if a large number are excreting close to this amount and they are also subject to reasonably varied exposures, then it would seem to me that a very frequent urinalysis program is needed – maybe once a week would not be too often in that case. It appears that the sampling interval could very well be considerably longer, on the other hand, if conditions are less variable and the employees do not regularly excrete more than some average amount. This is also true if they have been sampled over a sufficiently long period that their excretion pattern is known (this is a particular condition) and their average excretion is well below some arbitrary figure (this being the point of concern). Also, if there is a possibility that an employee can be seriously overexposed unknowingly, then a frequent urinalysis program is needed, whereas if this is practically impossible, then it can be cut down tremendously.

One must consider how urinalysis fits into the program, what the unknowns are, and why exposures are not detected in other ways; checks and balances must be applied. At one of the plants in Oak Ridge the use of film badges is supplemented by the use of pocket chambers that are looked at every day, so that there is little chance that anyone will get an extremely high exposure without it becoming promptly known. Without the pocket

chambers as a check, I think the use of film badges alone would be highly dangerous.

WATSON: At Hanford we certainly don't sample every person in the plant with the before-and-after exposure sampling frequency described in Mr. Wilson's paper. Several hundred people work in plant areas where we consider the exposure to be sufficiently irregular to put them on this type of sampling frequency. Other people who work in the same buildings as supervisors and people who occasionally go into these buildings are sampled annually.

We don't try to correlate air sampling results with bio-assay results. Someone expressed this adequately this morning by saying that taking air samples gives an idea of the potential exposure, whereas the urine specimen shows whether or not a person was exposed.

As to the relationship of urinalysis to body burden, when we find a person continually excreting >25 μg/l in the after-weekend sample, we frequently remove him from exposure and take a series of special samples in order to attempt to evaluate the body burden. In the case of before-weekend samples, we give special consideration only to unusually high ones.

HYATT: It has been pointed out that when a new process is introduced more urine samples are needed. We have sampled on a daily, weekly, and monthly basis, but the question remains of how often to sample routinely. Dr. Quigley advocates once every 3 months. I would like to determine the minimum frequency, once control is established and air sampling indicates levels below tolerance so that urinalysis has become routine; is the minimum every three months, or every year? Also, would it be reasonable to collect only an after-weekend sample? This is the type we prefer.

EISENBUD: The question of minimum frequency has considerable economic implications, because each analysis must cost between $2 and $5, and for, say, 4000 employees this could mean a large expenditure.

HYATT: The cost is about $5.

EISENBUD: That is $20,000 to do a set of 4000. Whether they are done every week or every month or every year could make a considerable difference in cost.

PATTERSON: With respect to the best sampling frequency, we use statistical analysis to a considerable extent in our film badge program, our air sampling program, and our urinalysis program for both enriched and normal uranium. We have reduced the frequency of sampling in both our air programs and our urinalysis programs on the basis of the statistical evaluation of the individual samples and the long-term data collected. In the normal uranium urinalysis program, there are very few if any persons in our routine program from whom we request samples more than once a quarter. In the enriched uranium urinalysis program, past experience has shown that we must take samples much more frequently. However, there are many people ($\approx$5% of the 1100 persons working with enriched uranium) whom we sample only once a quarter routinely. This is a result not only of our past experience but also of the efficiency or precision of urinalysis.

All of us realize that urinalysis is no science. But we are being pressured by people outside our own field of industrial hygiene or health physics: by the workers, who are the subjects in our program; by their supervisors, who want maximum use of employees and maximum production; by management, who want the optimum program for the least expense; and by the AEC itself, which frequently demands that we submit scientifically the unit dose that people have received. They all want a scientific answer. The AEC requires a scientific unit of dose in each of several different ranges, estimated by the best method of analysis, and we are left to decide the best method. And at present – at least until a method such as *in vivo* counting becomes available for use with large groups – urinalysis is our only method for making this sort of evaluation, leaving the question of frequency unanswered. As an indication of the problems connected with *in vivo* counting of large groups, we have estimated that it may take 3 years with our single counter, at the rate of 6 persons per day, to test the people in our enriched uranium exposure program, not including controls or special cases.

WATSON: As time goes on and we get more information from physicians (case histories, autopsies, etc.), urinalysis data will be valuable in relation to these findings.

QUESTION: I wonder if there isn't a point about urinalysis that we have overlooked. By analogy with lead, for example, what is being excreted is an index of the uranium currently mobile in the body, and not of the total amount in the body. In the case of lead the urine value can be low

while there is still lead in the body. I don't think any attempt has been made to estimate a body burden of lead.

NEUMAN: That is true. I think lead is more complicated than uranium in this respect. All of us are agreed that it is very difficult and only rarely possible to estimate body burden from urine data even roughly. But it is a means and in certain instances a fairly good one. We are also agreed that air data have severe limitations and also are very limited in their usefulness in determining body burden. We are not at all sure about body burden being the only parameter because there are both nephrotoxic and radiologic effects. We all agree that air data and urine data do not measure the same thing, but both are useful and should be used in assessing possible hazards in the plant. Whether the measurement of body burden will ever become as routine as urinalysis is a serious question, but, particularly in difficult cases, we should get all the data we can.

With respect to frequency of urinalysis, in a plant with few personnel changes and shifts from one job to another, even less than 4 times a year should be adequate. This seems to me a good figure for safety and not too expensive.

BARNES: I don't like to talk in terms of a plant. There are many different processes and functions carried out in any one plant, and the analysis should be in terms of jobs performed rather than on a plant basis.

NEUMAN: Rather than argue, I would like to emphasize that.

BERNARD: Do you think 4 months between urine samples is adequate to determine body burden without a direct body burden count, as Mr. Burkhart reported this morning? What about uranium in the bone?

NEUMAN: I have no illusion that a quarterly sample (this would be every 3 months) would furnish any real basis for estimating body burden, although trouble spots would be indicated by such a survey. If the AEC wants to know body burdens, they are demanding more than quarterly samples.

RUNDO: In principle I am opposed to routine monitored whole-body counting mainly because of the tremendous difficulty (if not impossibility in most cases) of differentiating between internal contamination and surface contamination that has not yet been washed off but has gone unnoticed because the usual hand monitors are insufficiently sensitive. Hands can appear clean when checked in this equipment and still have a fairly high activity in low background counting, which is usually impossible to distinguish from internal contamination. In fact I am rather alarmed to hear that they plan to measure 6 people a day for the next 3 years.

HARRIS: The question of sampling frequency seems to have been answered, in that quarterly urine sampling appears adequate with routine operations.

The next question is, what is to be considered an action level?

SCHOEN: The subject of action levels is extremely complicated. If we accept the fact that a certain urinary excretion rate is permissible, and a figure generally used seems to be 50 μg/24 hr, then excretion at this rate is taken as an indication that the amount of uranium in the body is approaching the permissible body burden. If we accept this figure (or any other figure), then, as Mr. Barnes stated with reference to air levels, we ought to accept the indicated burden as permissible. What has actually happened throughout the industry (as determined by a survey of health practices) is that the action points used are set at 50% to 10% or less of this permissible level. In most instances the action point meant stopping the operation or transfer of the people to another job, or some overt action other than simple evaluation of the job or the particular location. We should discuss at what levels to act and what sort of action is indicated at any particular level. The point of economics comes into the question too, because obviously as action levels are lowered, costs will rise.

WATSON: I would like to correct something I may have implied, that we permanently transfer a person who excretes at a certain level. This is not the case at all. We do not think in terms of action levels, but consider a high result only as a signal to instigate special sampling on the employee to try to estimate his body burden. I think you will all agree that once the body burden is above the permissible level, it is almost too late to do anything about it. If you want to keep body burdens below the maximum permissible level, then you have to take action at a lower level. At Hanford we work with many isotopes, and our general policy is that, once the body burden is estimated to be above 20% of permissible for any isotope, then the man is restricted from work with that isotope. This is deemed necessary to keep him

from accumulating more than the body burden in case of an accident.

HENRY: I don't think you can define action points and action to be taken. For example, restriction of work in a certain zone should not occur until there is some indication that, first, the urine result is due to a relatively fixed form of uranium exposure, not just a recent inhalation, and, second, that it reflects a level above what you call the body burden. There may be economic reasons to act otherwise that could be entirely justified: i.e., it may be cheaper to restrict a worker at a lower level so that he can return to work sooner. The action taken must depend on the type of operation, the possibilities of transfer, etc. The action point depends on the action taken; for example, one action is to investigate the operation, and this can be done at any level. For definite, drastic action, I think the two criteria I mentioned above should be met. Furthermore, the body burden cannot be defined because in 3 months it may have disappeared.

BARNES: I don't like the term action point because I think it implies some things that are not necessary. We prefer to call it a check point, some value at which we think that the person is getting reasonably close to a permissible body burden. The action that results from this check point is to find out why the man is receiving a higher level of exposure than he has in the past. Rather than an elaborate program involving the man, it is a program involving the job. Why has the exposure increased? Has something gone wrong? Is the ventilating system not working? Are the filters plugged? Has there been a change in the process? Is there some factor we didn't realize existed? We feel that this system gives good returns for the amount of time and effort put in.

QUIGLEY: I think that there is a great American custom that we can use to advantage in such situations and that is, set up a committee. We used to become concerned if any urine showed >50 μg/l and very concerned if it showed >100, but it was very difficult to get any action, so we set up a committee. It includes one industrial physician who is primarily responsible, who is the head of our analytical laboratory so that he can defend the methods of analysis, and an industrial hygienist who is responsible for conditions in the production area. Now if we see a urine result >50 μg/l the committee is concerned, and if we see one >100 we investigate immediately. The Medical Department's responsibility is to call in the employee for immediate recheck. The original samples are Monday morning samples, so that a recheck on Tuesday morning is not an attempt to keep sampling until we get a low result. If the second result is also high, the industrial hygienist has to find out why. Incidentally, the committee reduced the level from 100 to 50 μg/l, simply because they were not getting any results >100.

EISENBUD: I think that is a very valuable suggestion. This problem cannot be made so simple that all one has to do is prepare a list of actions and put them into effect as the results roll out of the lab. The air program and the urinalysis program and the conditions in the plant have to be combined as a basis for making a decision. Many factors must be evaluated before the decision is made to take a man off a job or to shut down an operation.

BUTTERWORTH: When we talk about action levels, is it not first of all necessary to specify that they must vary according to the frequency of some occurrence? When we have a new plant, a new operation, repair work, maintenance work, and so on, so that we know a particular job involves a hazard, we take spot samples at the end of the working day, not less than once every 3 days. On the other hand, for routine operations whose safety has been reasonably well established, if we want information for qualitative purposes or for comparison of different plants, then we sample not less than once every 14 days.

We don't sample once every 3 months because for this one has to have a definite purpose in mind. Such a schedule might be used to compare qualitatively the operations at one plant with those at another, or to indicate a person's exposure over the past week or two, but it would not reflect the exposure over the past 3 months. We occasionally sample employees on their return from long-term sickness absence, hoping, perhaps, to define body burden. After a long sick leave we also keep the employee away from uranium for a time, perhaps a month, after he is back on the payroll.

As far as action levels are concerned, we use two. The first is the investigation level. At this point we inform the plant management that all may not be well. We also obtain further samples, which are checked not only for uranium but also for protein, because protein in the urine is the only indication that the kidney has been injured. The second level, the real action level, is three times

the investigation level. This requires further urine samples and a full-scale inquiry by Health Physics personnel and, if necessary, by management. At the levels we use now, we are really not required to take any action because we very rarely reach them. The investigation level on spot samples is 100 μg/l, and the action level is 300 μg/l; these were formerly 40 and 100, respectively.

BERNARD: I suggested years ago that more attention should be given to past history. You suggested your limit, Dr. Butterworth, but you are not using all the data but only one piece. You say that if the urine result is >300 μg/l then you will take some action. It seems that no one asks what has happened to an employee in the past. I suggested at Y-12 that all these results be taken into consideration in arriving at an active rem dose.

HARRIS: I asked for an action level and I have gotten reasons why action levels may or may not be important, but no numbers, except from Dr. Butterworth. It is important that we come to some agreement on numbers. I think both Dr. Quigley and Mr. Barnes have presented useful information on what kind of action might be taken if a urine result exceeds a certain limit, but this is not universal; many different actions are taken at many various levels. I am perfectly willing to accept an investigation level or a complete investigation level (which I think are the two Dr. Butterworth mentioned), but what kind of numbers should we use? Can this panel specify a urine level below which the results are unimportant and beyond which it is worth while to make an investigation?

EISENBUD: You have probably done as much thinking about this as anybody; what figures would you propose?

HARRIS: I have very little faith in urine sampling as a program. I think urine sampling can show certain things, and that, for a specific purpose, a urine sample can be taken from an individual or a group at a time such that it will give useful information. Since I consider routine urine sampling to have relatively little value, I cannot suggest an action level because I don't think there is any level that would be very meaningful. On the other hand, if quarterly urine sampling is done, then one must choose some value at which to act, rather than merely taking the data and filing them away. I would be willing to accept 100 μg/l as a recheck level, to find out whether or not there had been contamination, applied to a Monday-morning sample. It makes no difference whether the uranium is soluble or insoluble; the urinalysis tells us what is being excreted, and the only thing being excreted is what is solubilized.

EISENBUD: A urinalysis program also indicates whether there has been a change. The industrial hygienist knows what the air samples and urinalyses have been showing. He uses his judgement as to the level at which conditions need not be better but should not get worse. Any upward shift in the level would be a basis for action.

HARRIS: This would be true if the samples were not influenced by daily changes in exposures. However, if a quarterly urine sampling program is being used and a sample shows more than a certain value (I would be willing to accept 100 μg/l) then, if the recheck verifies this value, I would consider instigation of a field investigation as an acceptable action. Often in practice, when the action level (whatever it be) is exceeded, an employee is immediately transferred and/or put into a respirator; I don't think the facts warrant this.

QUIGLEY: In our program we are not anxious to transfer men unless we are sure it is necessary. If we find a man excreting above the action level, we recheck. If the result is still high, we take samples from other men in the same group. It was by this method that we uncovered a condition we had failed to recognize by our air survey, and we were able to go into the plant and find the cause. It is also useful to know what is being processed in the plant and the changes in the processes. We find it valuable to sit in on engineering meetings at which changes are discussed before they are made.

EISENBUD: Let us turn to the discussion of air sampling procedures.

HYATT: Many methods of air sampling are being used, but I think we could find certain areas of agreement. First I would like to mention filter papers and to suggest that they be standardized. Besides the five recommended in the Little report, a few others are adequate. With regard to other sampling equipment, there seem to be no problems. The biggest differences are in the methods of sample collection. I think we agree that breathing zone samples are the only ones that truly represent a man's exposure on open procedures.

One thing on which there is disagreement is again an action level, a certain air concentration on a fixed operation. I think the only action that should be taken if one sample result is near or slightly above tolerance is engineering control, or an investigation of plant conditions.

EISENBUD: I assume that you are asking for an action level in the context of a reasonably repetitive operation that is expected to continue, so that by the time the investigation was finished the operation would not be discontinued. This must be taken into consideration, because for a short-term operation conditions can be permitted that would not be acceptable on a long-term basis.

QUIGLEY: We have always used weighted averages established in the manner discussed this morning by a speaker from the AEC. In the early days our action level was 10 times the MAC on a weighted basis; an employee getting this exposure was required to wear a respirator for the portion of the job that was the major contributor. We now take such action at 3 times the MAC. But the important thing is that the portions of the job responsible for the greatest part of the exposure are put in for engineering changes as soon as possible.

HENRY: With respect to sampling, the only true sample collected is inside the man who breathes the air. It can be assumed that the breathing zone sample is the best one collected. However, there are two things to consider: First, to find a change in conditions may not require a breathing zone sample, since a general area sample may show it. Second, it may be sufficient to relate the sample to what the man breathes; for example, if it were known that the sample is always 10% of what the man breathes, it would not matter whether it was a breathing zone sample or something else, as it would simply be multiplied by a factor of 10.

We did a few experiments in which we placed samplers with the nozzles about even with the nose and at the same time put some respirators on the men. In some experiments the men were relatively stationary and in others we followed them around. We found an interesting thing: the respirators indicated air concentrations higher by factors of 4 to 10 than did the breathing zone samples.

With respect to general air samplers, we mentioned that we had put some continuous ones into operation, and we obtained some rather interesting results. They were counted 5 hr later in order to permit the radon and thoron to die out. In the early days we had found high activity in a certain area where material had been released near the end of a shift (so that 5 hr had not elapsed when the next shift came on) and the second shift had been working for a while before the counter started chattering. In this way we found that general samplers picked up highly localized changes in air activity. It seems not unreasonable to assume that a general air sampler could be used pretty generally if it were possible to calibrate the meaning of the readings.

CHRISTOFANO: In the light of our present information, is it at all possible to suggest a more stringent allowable level in terms of air exposure, with the understanding that it might be exceeded for a half or a quarter of the time?

HENRY: All the limits used are based on a continuous average. If the level is twice as high half the time, then to avoid harm it must be zero the rest of the time.

NEUMAN: On the other hand, I think it would be wrong to give the impression that all the contractors are obliged to fill everyone up with the maximum allowable body burden.

EISENBUD: There is a rumor that the ICRP is about to recommend a very much higher permissible level for uranium exposure, something like 250 $\mu g/m^3$. I don't know the basis of this decision, but it may well have been the judgement of the rbe.

SNYDER: I happen to be connected with the preparation of both these handbooks on internal dose, and I really can't tell you what those numbers will be. Ballots have been sent to the committee members for their opinions, and they are slowly coming in. Among the changes being considered is whether the half-time in the kidney should be lower. The members may express their own opinions, but the figure 15 days is one of those suggested on the ballot for consideration. The toxic level in the kidney is also under consideration, and there is no basis at present to say whether the figure will be higher or lower. I don't think the change will be great.

On the matter of rbe, many of the committee members consider that the rbe for alpha is high for acute conditions. On the other hand, many of them feel, and there is some experimental evidence, that for chronic conditions 10 is not at all an extreme figure; in fact, there are some experiments dealing with some of the chronic effects we are supposed to be concerned with that indicate even higher values.

BERNARD: Our Boston data suggest that the half-time in the kidney should be of the order of 300 days, but others think this is too long. The data indicating 300 days are from only one patient. The point is that, if the 300-day half-time

can be used, then the occupational exposure can be divided by the factor of 10 to give 9 $\mu g/m^3$.

HYATT: Regarding future activity along the lines of this symposium, some of us have wondered whether the AEC Division of Biology and Medicine might not appoint a committee, say, a Health Protection Committee for Uranium Processing, the main purpose being to indicate areas of agreement and to attempt to standardize certain procedures, such as air sampling, urinalysis, etc., as well as to point out the limitations of these. Would this be feasible?

EISENBUD: I think I speak for everybody when I say it is feasible, and seems to be desirable. Perhaps Dr. Ely might take this suggestion back to Washington to see whether it would be possible to produce a document providing a concensus, with some flexibility, of the views with regard to air sampling, urinalysis methods and frequency, target levels, etc., on which people apparently would like to have some – not standardization – guidance. Is that what you had in mind?

HYATT: That is right.

PATTERSON: It has been agreed by all of us that the study of the people who have been exposed in the past would be very valuable to the industry as a whole and to the country as a whole. A plant such as ours at Y-12, with a long experience in various types of uranium processes, would be a very fertile field for such a study. We have some people who have been there for a long time, some who years ago, according to our criteria, had to be restricted from further exposure. The support, funds, and cooperation for such a project will have to come from some central agency. The individual plants, unless they are provided with the funds and the go-ahead will never be able to do it themselves.

EISENBUD: In closing, I would like to express my personal thanks, and thanks on behalf of all of us, to Dr. Norton Nelson and General Armstrong and the others here who made this auditorium available to us. I know how much effort Mr. Harris and his staff have put into the preparation of this symposium, and I think we owe them a vote of thanks. Also Dr. Ross and the people from the AEC Division of Biology and Medicine have been active, as well as Dr. Ely.

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*Symposium paper.
†Panel discussion.