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THE ENVIRONMENTAL AND BIOLOGICAL BEHAVIOUR OF PLUTCNIUM AND SOME OTHER TRANSURANIUM ELEMENTS

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The Environmental and Biological Behaviour of Plutonium and Some Other Transuranium Elements

C. R. Richmond Oak Ridge National Laboratory

Good morning ladies and gentlemen. In the time allotted this morning I would like to discuss with you a report prepared by a group of experts for the Organisation for Economic Co-Operation and Development's (OECD) Nuclear Energy Agency, on the subject of Environmental and Biological Behaviour of Plutonium and Some Other Transuranium Elements.[1] This report has been available for slightly over a year. It has not been widely distributed throughout the relevant scientific community in the United States. Many individuals in the radiation protection community are unaware of its existence. It has been my experience that many reports of this kind published by European organizations such as the OECD or the Commission of the European Communities (CEC) are not widely read within the United States. An example of the latter would be a report of the CEC entitled "The Toxicity of Plutonium, Americium, and Curium," by J. C. Nenot and J. W. Stather.[2]

Table 1 shows the composition of the NEA Group that prepared the report. Representatives of seven countries participated in the preparation of the report.

The objectives of this review are threefold.

- To summarize our knowledge of the way in which plutonium and some other transuranium elements migrate through ecosystems.
- To consider how the physiochemical state of these elements and the biological systems through which they pass may influence this movement.
- To put into perspective the risks of serious illness in man resulting from his exposure to these elements in the environment.

Table 2 shows the Table of Contents and the major sections of the report. The report considered the transuranium elements from the standpoint of sources and inventories, release through testing of nuclear weapons or from the nuclear fuel cycle, behavior in the environment, and transfer to man. Also discussed is the biology of the transuranium elements including a section on human exposure to transuranium elements and the risk coefficients for several human organs and tissues. Of interest in an annex which includes examples of environmental contamination from various locations around the world.

The summary is of special interest. It appears at the beginning of the report, and in ten pages covers a succinct summary of the entire 116-page report. The summary is written in relatively simplified language so that it is useful to the non-scientist and those who do not specialize in the area of plutonium and the transuranium elements.

Table 3 shows a major source of plutonium contamination; that is, levels in man resulting from the testing of nuclear weapons in the atmosphere, most of which occurred prior to the limited test ban treaty of 1963. Table 3 shows the computed levels of plutonium and various organs of man based on inhalation intake data for New York City.[3] Peak levels of 0.146 Bq in the total body were observed in 1964.

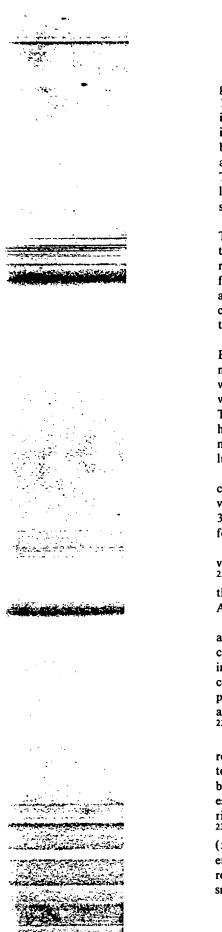


Table 4 shows the tissue levels of plutonium for members of the general public throughout the United States for the period 1959-1976.[4] For tissues such as lung and liver, which are of major interest as regards potential biological effects, over 700 samples were included in the 1979 report. The last column shows the comparison between the measured organ burdens derived from the McInroy study and the burdens estimated from the Bennett data, shown in Table 3. The data are in reasonable agreement for organs such as lung and liver, whereas the measured and estimated levels for vertebrae are considerably different.

Table 5 lists the risk coefficients for death from cancer for 239 Pu. These values are theoretical calculated cancer deaths as an estimate of the total expression in a population of both sexes and all ages. The risk coefficients for lung cancer, bone cancer, and leukemia are taken from ICRP 26,[5] and the values for liver and gut cancer are treated as two of the five "remaining tissues" as defined by ICRP in its publication 26. These latter two risk coefficients contribute toward the total risk coefficient of 5 x 10⁻³ per Sv.

Table 6 provides an estimate of the calculated cancer deaths in Europe which could hypothetically result from $^{239-240}$ Pu in fallout from nuclear weapons tests. Three organs--namely, lung, bone, and liver--were considered. Individual organ doses for the period 1954-1974 were applied to the European population of 0.45 x 10⁹ persons. The risk coefficients are those shown in Table 5. Resulting hypothetical cancer deaths totaled 187 for a population of approximately 0.5 billion individuals. The major predicted health effect was lung cancer.

Table 7 gives the annual limits on intake (ALI) and derived air concentrations (DAC) for workers inhaling 1 μ m AMAD aerosols of various transuranium elements. These data are taken from ICRP 30.[6] For ²³⁹Pu the class Y give an ALI of 500 Bq. A similar value for class W compounds is 200 Bq.

Table 8 gives the ALI values for workers following ingestion of various transuranium elements as taken from ICRP Publication 30 for ²³⁹Pu. The f_1 value of 10^{-5} results in an ALI of 2 x 10^6 Bq. For those compounds of ²³⁹Pu with an f_1 value of 10^{-4} , the corresponding ALI value is 2 x 10^5 Bq.

Table 9 gives the risk of radiation-induced cancer deaths per ALI as calculated from ICRP Committee 2 dosimetric models and risk coefficients derived for various tissues. For class Y compounds of inhaled 239 Pu the risk of developing lung cancer is 3.8 x 10⁻⁴ and a corresponding value for class W compounds is 7.2 x 10⁻⁶ or about 7 per million. Similarly the risk of developing bone marrow cancer is about 1 in 10,000 following inhalation or ingestion of 1 ALI for 239 Pu.

Table 10 summarizes some of the information contained in the report for globally dispersed plutonium rising from the atmospheric testing of nuclear weapons. Of the 12 PBq of ²³⁹Pu and ²⁴⁰Pu globally distributed, the amount in the upper several centimeters of soil is estimated to be about 0.4% (4 x 10⁻³) of the total naturally occurring alpha radioactivity in that soil layer. In addition, the amount of ²³⁹⁻²⁴⁰Pu measured and estimated for people is about 0.5% (5 x 10⁻³) of the total body content of naturally occurring alpha emitters such as ²²⁶Ra and ²¹⁰Po. It is apparent that for plutonium released through the atmospheric testing of nuclear weapons, relatively small quantities find their way through the environment into human

subjects.

Table 11 shows the individual locations covered in the report that represent special cases of plutonium and actinide contaminations of the environment.

I thank you for your attention.

References

- 1. Report by an NEA Group of Experts. The Environmental and Biological Behaviour of Plutonium and Some Other Transuranium Elements, Nuclear Energy Agency Organisation for Economic Co-Operation and Development. (1981)
- 2. Nenot, J. C. and Stather, J. W. The Toxicity of Plutonium, Americium and Curium, A Report Prepared Under Contract for the Commission of the European Communities within its Research and Development Programme on "Plutonium Recycling in Light Water Reactors." (1979)
- 3. Bennett, B.G. Transuranic Element Pathways to Man, in Transuranium Nuclides in the Environment, 1AEA, Vienna, 367. (1976a)
- McInroy, J. F., Campbell, E. E., Moos, W. D., Tietjen, G. L., Eutsler, B. C. and Boyd, H. A. Plutonium in Autopsy Tissues: A Revision and Updating of Data Reported in LA-4875, Health Phys. 37, 1. (1979)
- Recommendations of the International Commission on Radiological Protection, ICRP Publication 26, Annals of the ICRP, Vol. 1 (3). (1977)
- 6. Limits for Intakes of Radionuclides by Workers, ICRP Publication 30, Part 1, Annals of the ICRP, Vol. 2 (3/4). (1979)

THE COMPOSITION OF THE NEA GROUP OF EXPERTS WAS AS FOLLOWS:

Mr. Lucien Fitoussi, France (Chairman)

Mr. Asker Aarkrog, Denmark

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Dr. Gian Felice Clemente, Italy

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Fallout ^{239,240}Pu Inhalation Intake and Body Content in Man New York, USA (Bennett, 1976a)

		Computed burden in man Bq x 10 ⁻³					
Year	Inhalation intake Bq x 10 ⁻³	Lung	Lymph nodas	Kidney	Liver	Вопе	Total body
1954	38	5.5	0.4	0.0	0.0	0.0	6
1955	50	11	i.1	0.0	0.4	0.4	13
•							
•							
1962 .	170	37	12	0.0	7.8	8.1	65
1963	453	91	17	0.4	11	11	130
1964	246	92	24	0.4	14	15	146
•							
•							
1972	7.4	7.0	18	0.7	33	35	94
1973	3.5	4.8	16	0.7	33	36	91
1974	11	4.4	15	0.7	34	37	91

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Concentration of Plutonium in Tissues of Non-Occupational Exposed Persons in the United States, 1959-1976 (Data from McInroy et al., 1979)

	Number of samples analyzed	239,240 Pu concentration Bq x 10 ⁻³ kg ⁻¹ Median	239,240 Pu burden of standard issues		
Tissue			Weight kg	Measured Bq x 10 ⁻³	Estimated* Bq x 10 ⁻³
Lung	· 705	7.0	1.0	7.0	7
Liver	701	26.0	1.8	46.0	33
Lymph nodes (tracheobronchial)	360	96.0	0.015	1.4	18
Kidney	631	1.8	0.31	0.56	-
Gonads (testes)	264	5.3	0.035	0.19	-
Vertebrae (skeleton)	325	11.0	10.0	110.9	35
Rib .	95	9.7		97.0	
Spleen	325	2.7	0.18	0.49	-
Thyroid	184	11.0	0.020	0.22	-

*Estimated from the data by Bennett, 1976a, for the year 1972.

Table	5
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Risk Coefficients	for	Deaths	From	Cancer ^a
NISK COEfficients	101	Deatins	1.1.0m	Cancer

Tissue at risk	Effect	Risk coefficient Sv
Lung	Lung cancer	2×10^{-3}
Liver ^b	Liver cancer	1×10^{-3}
Cells near bone surfaces	Bone cancer	5×10^{-4}
Bone marrow	Leukaemia	2×10^{-3}
Lower large intestine ^b	Gut cancer	1×10^{-3}

^aCancer deaths are an estimate of total expression in a population of both sexes and all ages.

^bTwo of the five "remaining tissues" defined by ICRP (1977) which contribute towards a risk coefficient of 5 x 10^{-3} Sv⁻¹.

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Estimated Cancer Deaths in Europe Due to 239,240 Pu in Fallout

Tissue cancer	Individual organ doses* (1954-1974) Sv x 10 ⁻³	Collective doses** Sv x 10 ³	Risk coefficient Sv	Cancer deaths
Lung	0.16	72	2×10^{-3}	144
Bone	0.09	40.5	5 x 10 ⁻⁴	20
Liver	0.05	22.5	1×10^{-3}	23
				187

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*Richmond (1974) **European population 0.45 x 10⁹

Annual Limits on Intake (ALI) for Workers and Derived Air Concentrations (DAC) for Inhalation of 1 µm AMAD Aerosols of Some Transuranium Elements (ICRP, 1979)

		ALI	DAC ^a -3	
Isotope	Class	Bq	-3 Bq m	
238 Pu	Y	6 x 10 ²	3 x 10 ⁻¹	
	w	$\frac{2 \times 10^2}{2 \times 10^2}$	9 x 10 ⁻²	
239 Pu	Y	5×10^{2}	-1 2 x 10	
	w	2×10^2	⁻² 8 x 10	
241 Am	w	2 x 10 ²	8 x 10 ⁻²	
244 Cm	w	4×10^2	-1 2 x 10	

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a -1 40 h wk .

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Annual Limits on Intake for Workers for Ingestion of Some Transuranium Elements (ICRP, 1979)

lsotope	a f 1	ALI (Bq)
238 Pu	-5 10 -4 10	3 x 10 5 3 x 10
239 Pu	10 ⁻⁵ -4 10	2×10^{6} 2 x 10^{5} 2 x 10
24] Am	5 x 10 ⁻⁴	5 x 10 ⁴
244 Cm	5 x 10 ⁻⁴	9 x 10 ⁴

a f is fraction absorbed from C.1. tract. 1

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Risk of Radiation Induced Cancer Death Per ALI Calculated From the Radiation Dose to Body Organs Derived From ICRP Committee 2 Dosimetric Models (ICRP, 1979) and the Risk Coefficients for the Different Tissues

	²³⁹ Pu	²³⁹ Pu		
Tissue at Risk	Class Y	Class W	- Pu (ingested)	
Lungs	3.8 x 10 ⁻⁴	7.2 x 10 ⁻⁶	-	
Liver	1.2×10^{-4}	1.1 x 10 ⁻⁴	1.3 x 10 ⁻⁴	
Cells near bone surfaces	2.9 x 10 ⁻⁴	2.6 x 10 ⁻⁴	3.0×10^{-4}	
Bone marrow	1.0 x 10 ⁻⁴	8.0 x 10 ⁻⁵	1.0×10^{-4}	
Lower large intestine	3.0 x 10 ⁻⁸	8.8 x 10 ⁻⁹	2.5 x 10 ⁻⁴	

FOR GLOBALLY DISPERSED PLUTONIUM FROM ATMOSPHERIC NUCLEAR WEAPONS TESTING ONE CAN DEMONSTRATE THAT

• About 12 PBq ^{239,240}Pu, 4.2 tonnes, were distributed.

• The amount in the upper 2 cm of soil is about 0.4% of the total natural alpha radioactivity.

• The amount in people is about 0.5% of the total body content of natural alpha emitters such as ²²⁶Ra and ²¹⁰Po.

• About 10⁻⁸ of the amount released is available to man.

ANNEX I. PLUTONIUM IN THE **ENVIRONMENT: SPECIAL CASES**

Trinity Site, NM, USA
Nevada Test Site, NV, USA
Hanford, WA, USA
Savannah River Plant, SC, USA
Los Alamos, NM, USA
Rocky Flats, CO, USA
Maxey Flats, KY, USA
Oak Ridge, TN, USA
Thule, Greenland
Palomares, Spain
Pacific Proving Ground, Micronesia82
Farallon Islands, USA
Windscale, United Kingdom
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