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**Considerations in the Assessment  
of the Consequences  
of Effluents from Mixed Oxide  
Fuel Fabrication Plants**

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June 1975

Prepared for the U.S. Nuclear  
Regulatory Commission

 **Battelle**  
Pacific Northwest Laboratories

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CONSIDERATIONS IN THE ASSESSMENT OF THE CONSEQUENCES OF EFFLUENTS  
FROM MIXED OXIDE FUEL FABRICATION PLANTS

by

J. M. Selby	L. D. Williams
E. C. Watson	R. J. Hall
J. P. Corley	L. C. Schwendiman
D. A. Waite	J. Mishima
L. A. Carter	R. K. Woodruff
J. G. Droppo	T. I. McSweeney
R. G. Clark	J. B. Burnham
C. L. Brown	

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BATTELLE  
PACIFIC NORTHWEST LABORATORIES  
RICHLAND, WASHINGTON 99352

## FOREWORD

The Atomic Energy Commission, through Fuels and Materials, Directorate of Licensing, established a contract with the Battelle, Pacific Northwest Laboratory to conduct a study on "Considerations in the Assessment of the Consequences of Effluents from Mixed-Oxide Fuel Fabrication Plants." As a result of the great interest expressed by the nuclear community in the results of the basic study, documented in BNWL-1697, an extension for further work in the area was granted. This report contains the original information in an expanded and more detailed form plus new materials derived from the study extension.

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I. INTRODUCTION

The purpose of this study was to provide information and identify parameters relevant to assessing the consequences to man and his environment of large scale mixed plutonium-uranium oxide fuel fabrication plants which will be needed in the next 10 to 15 years. The report identifies the pertinent parameters, values, factors and methods which may be used in evaluating the environmental consequences of routine plant operation as well as postulated accidents. This study provides a base for the development of siting criteria and safety analyses for mixed oxide fuel fabrication facilities.



## II. SUMMARY

In response to instructions from AEC Regulatory (now Nuclear Regulatory Commission), a 1 metric ton (MT) per day plutonium fuels fabrication plant 10 to 15 years in the future was characterized and assumed as the base case. The "reference facility" manufactures  $UO_2$ - $PuO_2$  fuel which contains 2 to 4 wt%  $PuO_2$  in natural or depleted  $UO_2$ .

Safety considerations generically defined and evaluated can be scaled as necessary for plants with varying capacity. Scaling factors include: production capacity and rate, criticality control limitations, personnel exposure, equipment limitation, plant design, filter loading, fabrication techniques, and risk (consequences of the release times the probability of such a release).

Source terms for the reference plant were developed for normal operation and for five postulated accidents. Normal operation of the 1 MT/day facility is expected to result in a source term or annual plutonium emission rate of 15  $\mu$ g. For the accidents analyzed, a maximum of  $10^{-2}$  g of respirable plutonium was assumed to be emitted from the two-stage high-efficiency filter systems. Both soluble and insoluble plutonium of this magnitude were deemed credible. Since the accidents examined were not inclusive of all of the credible events leading to releases from the facility, additional information germane to estimating source terms resulting from accidents is included in the text and in Appendix A.

Probabilities of releasing radioactive material from the reference plant were developed by propagating accident occurrence and equipment failure rate statistics from fuel fabrication facilities and from related industries. The difference was carefully discerned between the probability of events leading directly to releases and releases which would require a chain of events occurring simultaneously.

The main chemical contaminants from the facility would be compounds of fluorine and nitrogen. The anticipated releases of these chemicals represent little concern to the environs; they are well below recommended standard and threshold limit values.

Material released from the facility is assumed to be dispersed to the environment in either of two ways: 1) continuous releases (normal operation) based on meteorological data from 26 nuclear facility sites or 2) short duration release (accidents) according to accident description guidelines in USAEC Regulatory Guide 1.3.

Uranium is not specifically considered, except incidentally when in combination with plutonium, because the radiological considerations of uranium are overshadowed by those of plutonium. The plutonium to be processed in the plant has an isotopic composition typified by a Connecticut Yankee Reactor type<sup>(98)</sup> fuel assembly discharged at about 35,000 MWd/MT. This is a conservative estimate of the average isotopic mixture which will be experienced during fabrication in the next decade and beyond. The higher exposure plutonium represents the greatest radiological hazard; the "reference mixture" maximizes the consequences of the postulated plutonium source terms.

Inhalation of plutonium aerosols and subsequent deposition in the critical organs is the most important exposure pathway to man. The critical organs for plutonium are the bone for soluble compounds and the lung for insoluble compounds.

Dose calculations for material deposited in the critical organs are made using parameters recommended by International Commission on Radiological Protection Publication 2 and Publication 19. Total dose commitments for the lung and 50-year dose commitment for the bone are given for acute and continuous releases of the individual isotopes as well as the reference mixture. Comparisons are made between the ICRP Publication 2 lung model and the ICRP Publication 19 model (Task Group Lung Model, TGLM).

In the final analysis, an attempt was made to quantify the risks a plutonium fuel fabrication facility imposes on the environment. Because of limited information on occurrences and failure rates, the results developed in this study should be viewed as preliminary.

The 50-year dose to the critical organs of an individual at 1000 meters from the facility was estimated for the 5  $\mu$ g annual release rate of the mixture of plutonium and americium. Using the ICRP Publication 2 lung model

and Calculated Annual Maximum Sector (CAMS) atmospheric dispersion model, the 50-year dose to the lung (insoluble) and to the bone (soluble) from inhalation of the mixture continuously released at ground level is 0.06 mrem and 4 mrem, respectively. These values are about 0.006% and 0.4%, respectively, of the limits proposed in this report for routine plant emissions.

Exposures that could result from acute inhalation following accidents were estimated for an individual at 1000 meters from the facility. The basic data used was from the ICRP Publication 2 lung Model and the USAEC Regulatory Guide 1.3 for atmospheric dispersion (8 hr curve), dose commitment to the lung (insoluble) and the 50-year dose commitment to the bone (soluble) from acute inhalation following a ground level release of  $10^{-2}$  g of the reference mixture is 0.06 rem and 9.4 rem, respectively. Since the probability of releasing this amount of material is estimated to be  $10^{-2}$ /year (insoluble) and  $10^{-3}$ /year (soluble), the risk for this type of accident is about 0.6 mrem to the lung and 9.4 mrem to the bone. The total risk that the facility imposes on the environs is expressed as a summation of the product of the consequences and the probability of the release for each credible accident and for normal operation. For the accidents and normal operation discussed in this report, the annual "dose commitment" risk to an individual at 1000 meters from the facility is estimated to be less than 16 mrem to the bone, 1.5 mrem to the lung, 1.7 mrem to the thyroid, and 0.4 mrem to the whole body. It is expected that the remaining spectrum of accidents will not significantly increase these values. The analysis clearly indicates that the dose to the bone is the major consideration in evaluating the environmental impact of a plutonium fuels fabrication facility.

Limits are proposed for plutonium surface contamination and for the annual dose rate to the general population from the continuous release of plutonium from a fabrication plant. An area was judged to be "contaminated" with plutonium if the activity exceeded  $10 \text{ nCi/m}^2$ . It is proposed that the 50-year dose commitment to the lung and bone of the average individual in the population should not exceed 0.85 rem and 1 rem, respectively, for routine plant emissions.

### III. SCOPE

Although siting guides are needed for all of the facilities connected with the fuel cycle, this report deals only with the parameters germane to the siting of mixed oxide fuel fabrication plants. Extension of this study to uranium and thorium fuel fabrication plants or fuel reprocessing facilities would be relatively simple since much of the effort would overlap.

The effects of effluent releases resulting from in-plant accidents as well as normal operation are considered. Both radioactive and nonradioactive effluents are reviewed, with the emphasis placed on plutonium as the most limiting consideration in plant siting.

Proposed plutonium-containing fuels include: oxides, carbides, nitrides, carbonitrides, borides, sulfides, metal and various alloys. Each of these involves different preparation techniques and different safety characteristics. The fuel fabrication plant selected for this study is a production facility manufacturing power reactor fuel 10 to 15 years in the future. Existing fuel fabrication facilities of the "job shop" type should be able to handle the relatively minimal demand for the experimental assemblies for power reactors and the special fuel for R&D reactors. However, portions of this study will be of value in siting considerations for any type of plutonium fuel fabrication plant.

Fuel management experts predict that plutonium fuels for LWR's and FBR's will, for the next decade, be essentially in the form of mixed plutonium-uranium oxides. Routine use of carbide fuels will occur at some later time. Plutonium fuel requirements for HTGR's in the form of  $\text{PuO}_2\text{-ThO}_2$  or  $\text{PuC-ThC}$ , will be minimal for at least the next decade. Credibility for these predictions of plutonium usage is illustrated in Table 1 by the number of United States commercial fuel fabricators capable of fabricating plutonium fuels.

Based on the projected utilization of plutonium in the form of mixed oxide, the focus of this study has been on facilities and processes which

TABLE 1. U.S. Commercial Fuel Fabricators  
with Plutonium Capabilities

1. Atomics International
2. Babcock & Wilcox
3. Exxon Nuclear
4. General Electric
5. Gulf United Nuclear
6. Kerr-McGee
7. Nuclear Fuel Services
8. NUMEC<sup>(a)</sup>
9. Westinghouse

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a. Owned by Babcock & Wilcox.

are designed to fabricate mixed oxide fuels for LWR's and FBR's. Because of the highly competitive nature of the fuel fabrication industry, innovations are continually being made in mixed oxide fuel fabrication. There are no "standard" techniques as such. However, certain techniques do predominate so that alternative methods currently employed have minimal effect on the source terms.

#### IV. FACILITY AND OPERATIONAL PARAMETERS AFFECTING THE POTENTIAL MAGNITUDE OF RELEASES

##### A. MATERIAL AND PROCESS PARAMETERS

Fuel fabrication license applications, federal regulations, and the open literature were reviewed to characterize the mixed oxide fuel fabrication facility. With the assistance of personnel experienced in fuels research and operations, the process information was projected to identify the quantities and characteristics of fuel material expected for plants of the future. Uranium was not specifically considered in the study, except incidentally when in combination with plutonium, because the hazards of natural or depleted uranium are overshadowed by those of plutonium.

##### 1. Plant Product and Design Capacity

The postulated plant manufactures  $\text{PuO}_2\text{-UO}_2$  fuel for light water reactors (LWR's). The plant design capacity is 1 MT of fuel per day.

##### 2. Content of Plutonium in the Fuel

The LWR fuel is assumed to contain 2 to 4 wt%  $\text{PuO}_2$  in natural or depleted  $\text{UO}_2$ . Since the LWR fuel process line, with some modifications, can also be used for fabrication of fast breeder reactor (FBR) fuels, the character of these fuels are also examined. FBR is assumed to contain 8 to 30 wt%  $\text{PuO}_2$  in natural or depleted  $\text{UO}_2$ .

##### 3. Plutonium Isotopic Composition

The majority of the plutonium to be used in fuel fabrication in the next 10 to 15 years will come from processing of spent uranium fuel from LWR's. Isotopic compositions will range from approximately 95 wt%  $^{239}\text{Pu}$ , 5 wt%  $^{240}\text{Pu}$  up to that resulting from 30,000 to 35,000 MWd/MT exposure in an LWR.

Plutonium obtained from a processing plant will have a range of isotopic compositions, a function of the specific power production of the fuel material and the neutronic characteristics of the reactor system to

which the fuel was exposed. As the fuel exposure increases, the percentage of the higher isotopes of plutonium increase. Because the relative quantity of  $^{241}\text{Pu}$  increases with fuel exposure, the specific activity and therefore the radiological hazard of a unit quantity of plutonium increase with fuel exposure.

For the dose calculation, a specific isotopic distribution of plutonium, chosen as the "reference mixture," was obtained from a Connecticut Yankee Reactor type fuel sample discharged at about 35,000 MWD/MT.<sup>(1)</sup> This is above the isotopic mixture for the plutonium presently being processed and thus is a conservative estimate of the isotopic mixture which will be experienced during fabrication in the next decade and beyond.

Bone and lung dose curves have been included (in Appendix B) for specific unit releases of plutonium isotopes, i.e., bone dose per gram of  $^{238}\text{Pu}$  released, in addition to the curves for the reference mixture (in Section VI), i.e., bone dose per gram of Pu released. These isotopic curves were included for those who wish to determine doses for other isotopic compositions of plutonium. Contributions of  $^{236}\text{Pu}$  to the total dose were not included in these calculations.

The isotopic composition of the reference mixture of plutonium is shown in Table 2.

TABLE 2. Reference Mixture of Plutonium

<u>Isotope</u>	<u>wt%</u>
$^{236}\text{Pu}$	$7.3 \times 10^{-8}$
$^{238}\text{Pu}$	1.9
$^{239}\text{Pu}$	63.3
$^{240}\text{Pu}$	19.0
$^{241}\text{Pu}$	12.0
$^{242}\text{Pu}$	3.8

The total quantities of plutonium and its isotopic composition that will be available for fabrication into reactor fuel, including recycle, has been calculated for the years 1972 through 1985 inclusive for the LWR industry (Appendix C). The plutonium will also contain some  $^{241}\text{Am}$  (from the

A third possibility would be to employ totally remote, heavily shielded fabrication operations. If this method is used, personnel exposure considerations would have little effect on process design.

2) Equipment Limitations (Capacity and Reliability). Equipment capacity limitations could limit the quantity of fuel processed in a particular enclosure (e.g., if the largest sintering furnace available has a capacity of 1/6 ton of fuel per day, then six furnaces would be required). This would reduce the amount of material which is available for release in a furnace accident by a factor of six over what it would be with a 1 ton/day furnace.

Equipment reliability considerations affect the quantity of plutonium available for release in an accident. A fabricator may wish to protect his plant from complete production stoppage in the event of equipment failure by using, for example, four 1/4 ton/day capacity pellet presses in four different gloveboxes rather than one 1 ton/day press. In the event of a press failure, output would only drop to 3/4 ton/day rather than to zero.

3) Product Uniformity Considerations. The need to produce large quantities of fuel (LWR cores contain 22 to 44 MT of fuel) with uniform characteristics makes handling large quantities of fuel in certain fabrication operations desirable. Such operations include homogenization of plutonium nitrate solution to obtain isotopic uniformity and blending of  $UO_2$ - $PuO_2$  to obtain large batches with uniform plutonium content and powder characteristics. Such operations would probably be conducted with the maximum amount of material permitted by criticality safety, radiation exposure, and equipment design considerations.

b. Process and Product Variations

An alternate process to mechanically blending  $PuO_2$  and  $UO_2$  powder is coprecipitation. In the coprecipitation process, plutonium nitrate and uranium nitrate solutions are blended and ammonium hydroxide is added to coprecipitate plutonium hydroxide and ammonium diuranate. The precipitate is then dried and reduced and calcined in an atmosphere containing 6 to 15



vol%  $H_2$  in  $N_2$  to yield  $UO_2$ - $PuO_2$ . The remainder of the fuel fabrication process is identical to the model process steps that follow powder blending. Use of the coprecipitation process rather than the mechanical blending process should have little effect on the accident source terms. However, it does eliminate  $PuO_2$  powder treatment, identified as the one step representing the greatest potential source of releasable plutonium. Elimination of this step would negate this threat and could reduce the normal operation source term.

An alternative to loading elements with pelletized fuel is vibratory compaction. In this process,  $UO_2$ - $PuO_2$  powder is poured into the fuel tubes and compacted by vibration. (Typically, less than 10% of the  $PuO_2$  feed particles used in this process are less than  $10 \mu$ .) Other vibrational compaction processing steps are similar to those of the model process which precede pellet pressing and follow fuel rod loading. It appears that the vibrational compaction process will have minimal effect on the magnitude of routine or accident source terms; however, more detailed analysis could reveal effects not currently envisioned.

### c. FBR Fuel Fabrication

Because of the similarities of the fuel materials, a fabrication plant for LWR plutonium recycle fuel can, with minimal changes, be converted for fabrication of FBR fuels (possibly desirable as the FBR fuel market develops in the 1980's and 1990's). The principal difference in the fuel materials is that the FBR plutonium enrichment would be higher ( $\sim 8$  to 30 wt%  $PuO_2$  in natural or depleted  $UO_2$ ) and the FBR fuel pellets would be much smaller ( $\sim 0.25$  in. diam).

If the LWR  $UO_2$ - $PuO_2$  fuel fabrication plant were converted for 1/6 MT/day FBR (75%  $UO_2$  - 25%  $PuO_2$ ) fuel fabrication, the plant plutonium inventory would be approximately the same, but criticality considerations would force reduction in the amount of plutonium handled in one batch. Therefore, assuming the same facility design features, the accident and routine source terms for 1/6 MT/day FBR fuel fabrication should be no greater and, in some mixed oxide processing steps, less than for 1 MT/day LWR  $UO_2$ - $PuO_2$  fuel fabrication.

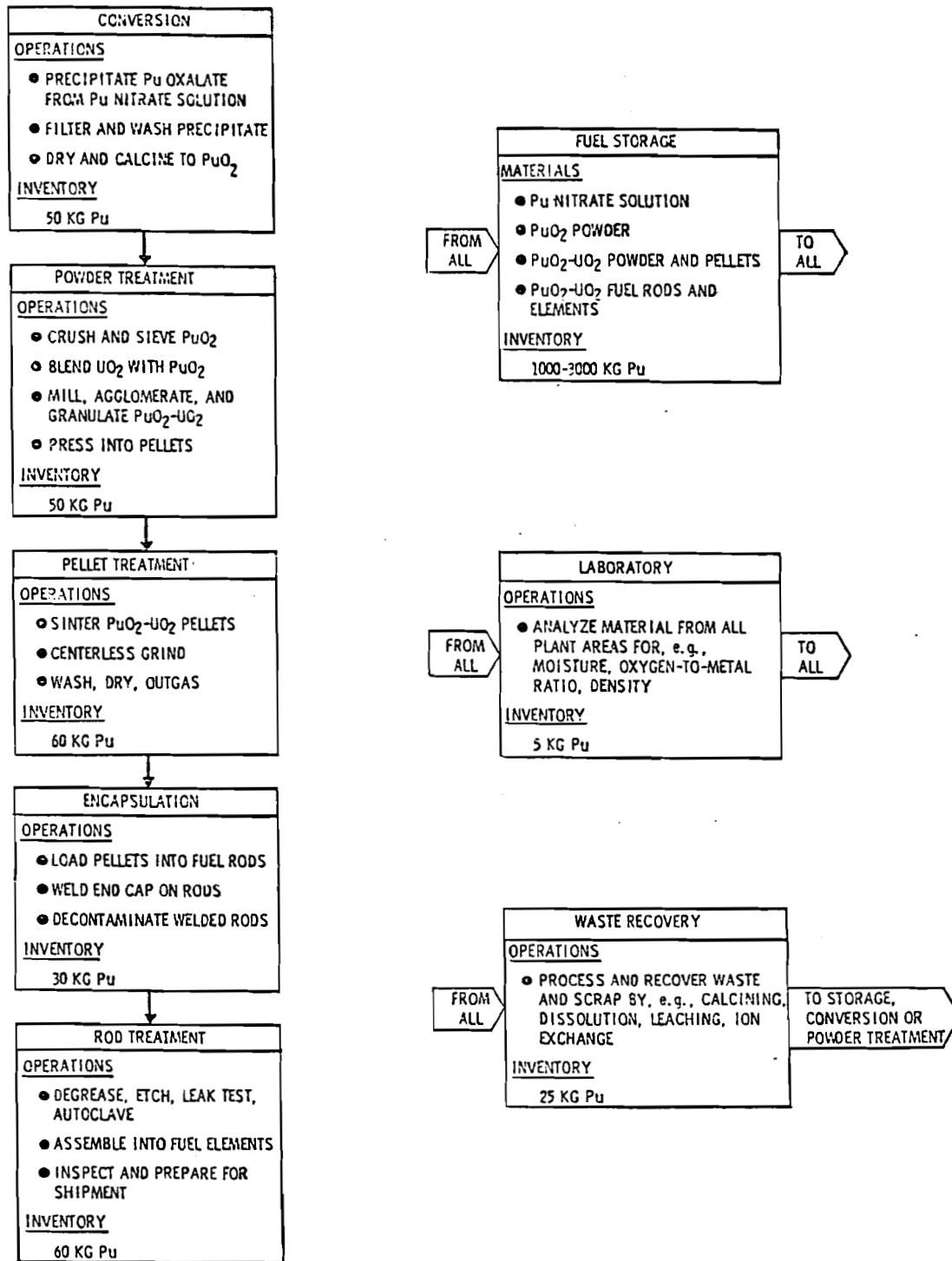


FIGURE 1. Area Flow Diagram for Model Process

processing line is presented in Appendix A. The chemical form, physical form, and mass of plutonium in the various areas of the plant have been included in the description since they are important in determining release parameters.

## 7. Additional Considerations

### a. Variations in Potential Release Inventory

For in-plant accidents (e.g., fire, explosion) the potential release inventory is dependent on the inventory in the component (e.g., furnace, glovebox, process area) in which the accident occurs. Several additional considerations are given in the following sections that could result in an in-process inventory size and distribution different from that assumed in the reference plant. As an example of the potential effect of these considerations, a conceptual study by Merker et al.,<sup>(4)</sup> of a 1 MT/day PuO<sub>2</sub>-UO<sub>2</sub> fuel fabrication plant describes a line that operates with less than 2 kg of plutonium at each processing station. The low station inventory is achieved by use of multiple, identical stations and semi-continuous inflow outflow of material on conveyors.

Considerations which could affect the in-process inventory include:

1) Chronic Radiation Exposure of Operating Personnel. The relatively high neutron and gamma radiation levels from high exposure plutonium could influence process changes in order to reduce the chronic radiation exposure of operating personnel.<sup>(5)</sup> These changes could take several forms. One would be shielding of the gloveboxes and reduction of the quantity of plutonium at the station. This would require an increase in the number of stations and/or a continuous inflow-outflow of material at the station in order to prevent the accumulation of large quantities of plutonium. If these stations are isolated from one another by some barrier, the potential release inventory for glovebox scale accidents could be reduced.

Another possibility would be to use the above strategy for some operations, but to (heavily) shield others such that the criticality considerations, rather than personnel exposure, limit the quantity of plutonium used in those select operations.

beta decay of the 13 year half-life  $^{241}\text{Pu}$ ). Approximately 5% of the  $^{241}\text{Pu}$  will decay to  $^{241}\text{Am}$  in 1 year. One year was chosen as representative of the elapsed time between fuel processing and use in a fuel fabrication plant.

#### 4. Plutonium Inventory

For a plant producing 1 MT of LWR fuel per day, the total plant plutonium inventory will be of the order of 1000 to 3000 kg.

#### 5. Design Limitations Imposed by Criticality Safety Considerations

One consideration that will limit the amount of plutonium in a process area is criticality. Criticality safety considerations will either limit the plutonium to a safe mass under specified conditions or the mass will be effectively unlimited (e.g., if a plutonium solution is contained in a cylinder whose diameter is less than the minimum critical diameter, then the cylinder length is not limited and the cylinder can contain an infinite amount of material). Safe masses of plutonium and mixed plutonium-uranium are given in Table 3. Because of its hygroscopic nature and the possibility of the addition of binders in processing, the reduction of the safe masses of  $\text{PuO}_2$  is shown for water uniformly distributed in the powder and the pellets. Values for 1 wt% and 5 wt% water are given.

#### 6. Fuel Fabrication Process

##### a. General Description

(1) Fuel Preparation. The plutonium is assumed to be received in the form of plutonium nitrate solutions that must be converted to  $\text{PuO}_2$ . However, the plutonium may be received in the form of  $\text{PuO}_2$  which would allow elimination of the conversion steps.

The standard techniques for plutonium fuel preparation are: 1) conversion of  $\text{Pu}(\text{NO}_3)_4$  to  $\text{PuO}_2$ , followed by mechanical mixing with  $\text{UO}_2$  to obtain  $\text{PuO}_2\text{-UO}_2$  powder and 2) coprecipitation of U and Pu from the nitrate, followed by reduction to  $\text{PuO}_2\text{-UO}_2$  powder. The Sol Gel technique, which is a special case of precipitation, shows promise but has been demonstrated only for small batch fuel preparation. Since to scale up this process involves unknown difficulty, it has not been considered in this analysis.

TABLE 3. Safe Masses in Plutonium Fuel Fabrication(a,b,c)

	PuO <sub>2</sub>		FBR Fuel <sup>(d)</sup>		LWR Recycle Fuel <sup>(e)</sup>	
	Pu Mass	UO <sub>2</sub> -PuO <sub>2</sub> Mass	Pu Mass	UO <sub>2</sub> -PuO <sub>2</sub> Mass	Pu Mass	UO <sub>2</sub> -PuO <sub>2</sub> Mass
Dry Powder	11.3 kg	115 kg	31.5 kg	>3600 kg	>126 kg	>3600 kg
Dry Pellets	4.86 kg	43 kg	11.7 kg	>3600 kg	>126 kg	>3600 kg
Powder with 1 wt% water		102 kg	28 kg	>2300 kg	>81 kg	>2300 kg
Powder with 5 wt% water		66 kg	18 kg	>338 kg	>11.9 kg	>338 kg
Pellets with 1 wt% water		39.5 kg	10.8 kg	2300 kg	81 kg	2300 kg
Pellets with 5 wt% water		32 kg	8.8 kg	338 kg	11.9 kg	338 kg
Optimumly Moderated Systems	230 g	1.02 kg	279 g	11.75 kg	413 g	11.75 kg

- a. A safe mass or batch is <45% of a fully water reflected critical mass.
- b. Plutonium is assumed to be 100% <sup>239</sup>Pu. The presence of <sup>240</sup>Pu will increase the safe masses (e.g., 20 wt% <sup>240</sup>Pu will more than double the safe mass of optimumly moderated LWR recycle fuel).
- c. Powder density = 5.6 g/cm<sup>3</sup>, pellets at theoretical density.
- d. Assumed to be 31 wt% PuO<sub>2</sub> - 69 wt% U<sub>(nat)</sub>O<sub>2</sub>.
- e. Assumed to be 4 wt% PuO<sub>2</sub> - 96 wt% U<sub>(nat)</sub>O<sub>2</sub>.

(2) Fuel Shape Fabrication. Pelletization is the fuel shape fabrication technique currently used almost exclusively for power reactor fuel. Although vibratory compaction, a developed fuel loading technique, is currently receiving only limited use, it does have advantages that may result in more extensive use in the future.<sup>(2,3)</sup>

(3) Scrap Recovery. Whenever possible, oxide scrap will be recycled through the process without chemical processing. For scrap and waste where this is not possible, a typical recovery process is leaching, or dissolution, followed by reduction and ion exchange yielding Pu(NO<sub>3</sub>)<sub>4</sub>.

b. Reference Process

In order to allow detailed investigation in the study, a specific process, (called the "reference process,") was chosen for examination. It includes conversion of plutonium nitrate to PuO<sub>2</sub>, followed by pelletizing of the mechanically mixed PuO<sub>2</sub>-UO<sub>2</sub>. A flow diagram for the reference process is given in Figure 1. A tabular description of the characterized

For 1 MT/day FBR fuel fabrication, the routine source term could be up to a factor of approximately six higher than for equivalent capacity LWR  $UO_2$ - $PuO_2$  fuel fabrication. Likewise, the source term from a major facility could be higher due to the larger plutonium inventory required. However, the quantity released could be limited by physical phenomena such as quantity of particles which can be supported by a volume of air. For an accident involving only one batch of fuel material, the FBR fuel fabrication source term should be no greater than for LWR  $UO_2$ - $PuO_2$  fuel fabrication.

## B. FACILITY

There are several facility design and operational features including the process confinement system, control of process inventory, and building integrity that influence or limit the potential for release of plutonium from a fuel fabrication plant. The process confinement system, designated as the primary barrier in this study, includes tanks and piping in the wet process areas, the gloveboxes and associated exhaust systems in the powder steps of the process, and the cladding after encapsulation of the sintered fuel pellets. Any plutonium facility requires an effective and reliable primary confinement system, if only for the protection of operating personnel. This required system plays an important role in minimizing release of material to the environment.

A second facility design feature (designated as the final barrier) which has significant bearing on the potential release of material is the integrity of the building structure and the associated building exhaust system.

Minimizing the fuel inventory which is at risk is another design feature of this facility. It includes decoupling various steps in the process relative to a given accident mechanism either by isolation by distance or barriers or by employing parallel production lines to minimize the normal process inventory. In some facilities, similar processes are confined within separate rooms or areas which provide an additional factor in controlling the release of radioactive material. This design feature is referred to as a secondary barrier.

Elaboration of some of the primary and final design features follow.

### 1. The Primary Barrier

For purposes of this study, the tankage and piping system is assumed to have sufficient integrity to assure that the system is capable of withstanding moderate stress above normal requirements without rupture. Further, all points of potential leakage (e.g., flanges and valves), are contained within enclosures to prevent spread of contamination to the room in the event of minor leaks that may be expected to occur periodically. While the contents of the system may represent a large inventory for dispersal in a severe accident involving catastrophic failure of the vessels or piping, its contribution to the normal operations source term is expected to be very small.

All powder operations are assumed to be carried out in discrete gloveboxes or glovebox systems that have sufficient strength to withstand operational pressure without structural failure. The gloves represent the principal weakness in the primary confinement barrier since they are subject to attack by chemical agents used in the glovebox, to physical damage from abrasion or cutting or puncture, and since they will likely fail in case of an explosion or pressurization within the glovebox. Hood gloves will periodically fail; however, only trace contamination will normally be observed outside the hood. In the event of a fire or explosion within the glovebox system, substantial quantities of plutonium could be released from this barrier.

The powder operations normally are carried out in air within a glovebox that has either a single pass or recirculating air system. Because of the possibility of propagating fumes throughout the system, it is assumed that air in a glovebox is received through a HEPA filter and exhausted through another HEPA filter to an exhaust system. The HEPA filters at each glovebox exhaust are highly desirable in that they minimize the accumulation of plutonium in the exhaust duct system.

Once filtered glovebox air may be routed through additional filters prior to release from the building, or may be combined with the general

room exhaust stream prior to release through the final building filters. Glovebox exhaust air typically is filtered through two or three HEPA filters before release to the atmosphere. The integrity of the glovebox system(s) and associated exhaust filter system(s) throughout the powder processing stages is clearly a major consideration in limiting the release from normal operation.

## 2. The Final Barrier

The final barrier is assumed to be a structure of sufficient strength to withstand severe stress (such as earthquake, tornado, intense fire or process explosion). It is further assumed that all building exhaust air is released to the environs through a building final filter system capable of withstanding the same severe stresses as the building.

The number of HEPA filters in series required in the building final filter system is not generally agreed upon; some facilities operate with one stage, some with two, and in one case five stages were deemed necessary.<sup>(6)</sup> This apparent disagreement stems largely from the fact that while the capability of a single stage is well known, the benefit to be derived from additional stages of filtration has not been well established. One extreme position would be that if one HEPA filter is demonstrated to be 99.95% effective by cold DOP test<sup>(7)</sup> and therefore has a transmission factor of  $5 \times 10^{-4}$ , then two HEPA filters in series will have a transmission factor of  $(5 \times 10^{-4}) \times (5 \times 10^{-4}) = 2.5 \times 10^{-7}$ . The alternate extreme view would be that the particulates that can pass through the first filter have demonstrated their capability for passing through a HEPA filter and therefore will continue to do so, thus a two filter combined transmission factor is  $5 \times 10^{-4}$ . The truth probably lies somewhere between. Based on AEC HEPA filter guide lines as referenced in Preliminary Safety Analysis Report for the Plutonium Recovery and Waste Treatment Facility at the Rocky Flats,<sup>(8)</sup> the first stage is assumed to be 99.9% effective and all successive stages 99.8% effective. Thus the combined transmission factor for two stages would be  $(1 \times 10^{-3}) \times (2 \times 10^{-3}) = 2 \times 10^{-6}$ .



In the practical case, the second stage at least filters that portion of the exhaust stream that bypassed the first filter because of seal leaks that may periodically develop or during changing of the first filter. Two stages of building exhaust stream filtration are frequently provided for this reason alone. During approximately 25 years of operation of the Hanford plant, no in-service HEPA failures have been identified. Filter changes are routinely required because of filter loading.

One engineering study of building filter capabilities determined, on the basis of actual measurements made on four stages of HEPA filters, that four stages are required to achieve a transmission factor of  $1 \times 10^{-7}$ .<sup>(9)</sup> Based largely on this engineering study, it will be assumed that, in practice, the following benefit may be expected to be achieved by multiple filter banks.

	Removal Efficiency	Transmission Factors	
		Specific	Aggregate
First Stage	99.9%	$1 \times 10^{-3}$	$1 \times 10^{-3}$
Second Stage	99%	$1 \times 10^{-2}$	$1 \times 10^{-5}$
Third Stage	94%	$6 \times 10^{-2}$	$6 \times 10^{-7}$
Fourth Stage	83%	$1.7 \times 10^{-1}$	$1.0 \times 10^{-7}$

As will be discussed further in Section VII, the material present in the intact gloveboxes or within vented piping systems was assumed to be separated from the plant environs by three stages of HEPA filters, and material elsewhere within the building, by two stages of HEPA filters as follows:

The glovebox air filtration system:

Overall Efficiency (3 HEPA)	99.99994%
Aggregate Transmission Factor*	$6 \times 10^{-7}$

The room air filtration system:

Overall Efficiency (2 HEPA)	99.999%
Aggregate Transmission Factor*	$1 \times 10^{-5}$

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\*Aggregate Transmission Factor is the ratio of the grams of material transmitted to the grams impinging on the first stage.

The foregoing values have been shown to be conservative by more recent data obtained under laboratory conditions. Although each filter reduces the average particle size, efficiency of the next filter remains above 99.9%. The minimum aggregate transmission factor obtained under laboratory conditions with  $\text{PuO}_2$  aerosols was  $1.0 \times 10^{-9}$  for two steps of HEPA filtration. A facility which has a properly installed and tested HEPA filtration system and can maintain integrity of the final confinement system will release only minor amounts of particulate materials to the environment.

A characteristic of filter media which makes the  $6.0 \times 10^{-7}$  and  $1.0 \times 10^{-5}$  transmission factors more conservative (higher than actual) is the improved efficiency with loading. Although little, if any, data are available on the increased efficiency due to dust loading, one observation showed that approximately 250 g of dust with an aerodynamic equivalent diameter (AED) of  $2.6 \mu\text{m}$  resulted in a 3 in. pressure drop for a 12 in. by 12 in. by 6 in. deep filter.<sup>(10)</sup> Although a pressure drop of 3 in. is not excessive in some installations, when exceeded it would call for a filter change. The efficiency is probably significantly higher at this point in the life of the filter than for the newly installed filter.

In an installation requiring 100 HEPA filters (100,000 cfm) each of which were loaded with plutonium to reach a pressure drop of 3 in. of water, the total loading would be of the order of 200 kg of Pu in the filter bank, a rather ridiculously large amount. Even with this unrealistic assumption, the amount of plutonium which would have penetrated the filter up to that point would have been no more than about 2 g ( $2000 \times 1.0 \times 10^{-5} \times 100$  filters). Evidently there is a realistic upper boundary to the amount of fuel material that can pass a HEPA filter. Although without total support, this example plus experience apparently justify an assumption that about 1 to 5 g is the maximum release which can be postulated from the most severe in-plant accident that does not compromise either the building or the two stages of HEPA filtration.

Evidence that such a self-regulating mechanism is at work is obtained from examination of the releases measured from a wide variety of plutonium

handling facilities, characteristically equipped with two or three stages of HEPA filters between the gloveboxes and the environs. Reported releases at AEC installations for 1971<sup>(11)</sup> (with one notable exception which is not a mixed oxide plant) are in the range of 1 to 75  $\mu\text{Ci}/\text{year}$ .

For a nominal flow rate of  $10^5$  cfm, the 75  $\mu\text{Ci}$  value is equivalent to an average release concentration of  $5.0 \times 10^{-14}$   $\mu\text{Ci}/\text{cm}^3$ . The reporting of "less than" release values of  $1.0 \times 10^{-15}$   $\mu\text{Ci}/\text{cm}^3$ , because of insufficient analytical sensitivity, is often misleading in analyzing plant emissions. Similar "less than" emission rates are reported for the commercial mixed oxide fuel fabrication plants. Therefore, within the present plutonium analytical capabilities of the industry, the apparent concentrations and annual release rates are similar for a large number of plutonium installations involving a wide variety of chemical and physical processes and significant differences in throughput.

### 3. Facility Model

It is assumed that prudent design considerations as well as regulatory requirements will result in the design of a facility such that the structure and the final filter system will maintain their integrity against the action of fires and natural phenomenon (earthquakes, tornadoes, floods, etc.). For the purpose of this study it is assumed that the facility design criteria will be comparable to criteria used in the design of reactors and for certain new AEC plutonium facilities.<sup>(12)</sup> In characterizing the reference facility, the design features which could have significant effect on the normal operation and/or accident source terms were examined. The following assumptions were made about these features for the plant "model."

#### a. Earthquake

The facility will be designed such that shutdown systems of the facility will remain operational during the maximum possible acceleration that could occur at the site. Additionally, the facility is designed such that those elements remain functional that are required for continued safe operation during the maximum probable acceleration that could occur at the site.

b. Tornado

Sections of the facility as defined in the Minimum Design Criteria for New Plutonium Facilities<sup>(12)</sup> will be hardened to withstand the effects of the AEC Regulatory Model Tornado.<sup>(13)</sup>

c. Flood

The facility either is located such that water from the Probable Maximum Flood (PMF,) which is the most severe flood reasonably possible at the facility site, would not reach the facility, or the facility is afforded the necessary protection in the form of dikes, diversion channels, etc., to remain unaffected by the water from the flood.<sup>(14)</sup>

d. Fire Protection

The facility design includes the necessary alarms and/or equipment to prevent, suppress, or contain a fire. In at least one facility<sup>(15)</sup> the filters are required to withstand 180°F continuously and 700°F for 5 min. Several tests have been conducted on the effects of elevated temperatures on high efficiency filters.<sup>(15,16,17,18)</sup> These tests indicate that filters made according to AEC minimum specifications will withstand temperatures of 700°F for 5 minutes. In the same facility the glovebox system final exhaust filter bank is to be protected by a minimum of a scrubber which acts as a large heat sink, a cooling chamber which cools by deluge spray and a demister together with the necessary heat detectors and alarms. The room air system final exhaust filter bank will be protected by a minimum of a spark arrester, a cooling chamber and a demister together with the necessary heat detectors and alarms.

The design of the process and the process equipment are extremely important in defining the protection necessary to assure the integrity of the building and the final filter.

e. Criticality

The facility is designed to remain subcritical under all operating circumstances. This is assured by either mass control of the fissile material (double batch principle) or by engineered safety features. Safe

geometry, backflow protection, poisoned systems, etc., are included in the design whenever feasible to assure safety with a minimum of administrative controls. Rapid pressurization of a hood or enclosure as a result of a criticality accident could be sufficient to breach the primary barrier. It is not credible that this force would compromise the final barrier, the building structure or the final filter system.

The generation of a fire as a result of the energy released during a criticality event in the reference facility is not considered credible. Criticality accidents are most likely to be initiated in solutions. All of the criticality accidents that have occurred in plutonium recovery systems have involved solution.

f. Liquid Waste

This facility is assumed to have three separate liquid waste systems considered in facility design. These include systems for contaminated wastes, clean process wastes, and sanitary wastes. Potentially contaminated wastes normally are collected at the point where they are generated or in a hold-up tank. It is assumed that there is no continuous liquid effluent flow from areas of the plant where there is potential for liquid effluents becoming contaminated with plutonium. Thus, with this type of passive system, accidental or routine release of plutonium is extremely low.

The sanitary sewer system is maintained separate from the process system with automatic sampling provided for the process system. Thus, this system should not contribute to environmental contamination.

Areas where water from fire fighting could become contaminated are assumed to be equipped so that the potentially contaminated water is prevented from reaching the environs.

## V. ENVIRONMENTAL CONSIDERATIONS AFFECTING CONSEQUENCES OF RELEASES

### A. ATMOSPHERIC DISPERSION

#### 1. Normal Operation

The Atomic Energy Commission has provided guidance for estimating the atmospheric dispersion of radionuclides accidentally released from nuclear power plants.<sup>(19,20)</sup> Similar guidance is not currently available from routine releases. A method is proposed for the purpose of evaluating long-term routine releases from normal plant operations. Two conditions were imposed on the method: (1) it should yield conservative estimates, i.e. overestimate the average ground level air concentrations, and (2) it should reflect realistic meteorological and diffusion conditions.

The long-term sector average air concentration at ground level,  $\bar{\chi}$ , of radioactive material diffused from a continuous point source is given by:<sup>(21)</sup>

$$\bar{\chi} = \left(\frac{2}{\pi}\right)^{1/2} \frac{0.01fQ'}{\sigma_z \bar{u}_h w} \exp(-h^2/2\sigma_z^2)$$

where:

$\bar{\chi}$  = long-term sector average air concentration at ground level,  
 $\mu\text{Ci}/\text{cm}^3$

f = percent of frequency with which wind blows in a sector during a specified stability at a given speed, %

Q' = average rate of release from source, Ci/sec

$\sigma_z$  = standard deviation of crosswind vertical dispersion of air concentration in cloud, m

$\bar{u}_h$  = average wind speed at height of release, m/sec

h = height of release, m

w = arc length of sector, m  $w = 2\pi x/n$

x = downwind distance, m

n = number of sectors contained in 360 degrees

Note: values for f, Q',  $\sigma_z$  and  $u_h$  are long-term average values.

In this study, a version of RACER<sup>(22)</sup> modified for annual releases,<sup>(23)</sup> was used in the calculation of annual average air concentrations for a 22.5° sector, normalized to an average annual release. On-site micro-meteorological data from nuclear power plant environmental reports for 26 sites were selected to calculate normalized ground level average air concentrations for both ground level and 100 meter release heights.

Calculations of the  $\bar{x}/Q'$  values with the data from each site were performed in a manner to obtain the maximum possible values. The calculational method went further than just using the sector with the greatest  $\bar{x}/Q'$  value, it maximized  $\bar{x}/Q'$  in each step by using the joint frequency of occurrence of wind speed and direction data, separated into stability classes. Table 4 is representative of data from one of the sites for the stable class. The maximum wind occurrence values for each wind speed group were found as shown by the circled numbers in Table 4. These maximum occurrence values were combined to create a hypothetical maximum sector for this stability class. The identical process of selecting the maximum occurrence values was used for each of the other stability classes for the site. Then, an average  $\bar{x}/Q'$  value was calculated as a function of distance based on the hypothetical maximum sector using RACER. This complete process was repeated for each of the sites. After the  $\bar{x}/Q'$  values for all of the sites were plotted as a function of distance, the maximum values were fitted to form an upper limit for both the ground level and 100 meter release heights. These curves are referred to as the calculated annual maximum sector curves (CAMS). (Appendix D)

The effect of a change in the distribution of stability classes resulting from this procedure was investigated and found to lead to a slight tendency to emphasize the effects of stable conditions. As a result, the CAMS curves tend to be slightly more conservative at greater distances.

For comparison, the on-site meteorological data from the reactor sites were also used to calculate mean values of sector average annual air concentrations. In this instance, the frequency distributions within each stability class and for each wind speed class were summed and then divided by sixteen. This results in a mean frequency distribution for a 22.5°

**TABLE 4. Sample Data Sheet for Maximum Sector Derivation**

CONNECTICUT YANKEE

100 FT WIND DATA

ANNUAL (3/4/63-3/6/64)

STABLE  
DELTA T LESS THAN +1.5 BUT GREATER THAN OR EQUAL TO -0.5 DEG C/100 M

WIND FROM	WIND SPEED (MPH)								ALL
	1-3	4-7	8-12	13-18	19-24	25-31	32-38	39+	
NNE	8	3	0	0	0	0	0	0	11
(1)	3.3	1.2	0.0	0.0	0.0	0.0	0.0	0.0	4.5
(2)	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.2
NE	7	5	0	0	0	0	0	0	12
(1)	2.9	2.1	0.0	0.0	0.0	0.0	0.0	0.0	4.9
(2)	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.2
ENE	19	20	0	0	0	0	0	0	39
(1)	7.0	7.4	0.0	0.0	0.0	0.0	0.0	0.0	14.4
(2)	0.3	0.4	0.0	0.0	0.0	0.0	0.0	0.0	0.7
E	33	45	5	0	0	0	0	0	83
(1)	10.5	14.3	1.6	0.0	0.0	0.0	0.0	0.0	26.4
(2)	0.6	0.8	0.1	0.0	0.0	0.0	0.0	0.0	1.5
ESE	47	70	13	0	0	0	0	0	130
(1)	13.0	19.4	3.6	0.0	0.0	0.0	0.0	0.0	36.0
(2)	0.9	1.3	0.2	0.0	0.0	0.0	0.0	0.0	2.4
SE	25	35	9	5	0	0	0	0	74
(1)	8.2	11.5	3.0	1.6	0.0	0.0	0.0	0.0	24.3
(2)	0.5	0.6	0.2	0.1	0.0	0.0	0.0	0.0	1.3
SSE	7	21	9	2	3	2	0	0	44
(1)	2.5	7.6	3.3	0.7	1.1	0.7	0.0	0.0	16.0
(2)	0.1	0.4	0.2	0.0	0.1	0.0	0.0	0.0	0.8
S	6	18	18	10	2	0	0	0	54
(1)	2.1	6.3	6.3	3.5	0.7	0.0	0.0	0.0	18.9
(2)	0.1	0.3	0.3	0.2	0.0	0.0	0.0	0.0	1.0
SSW	4	12	5	2	1	0	0	0	24
(1)	1.6	4.7	2.0	0.8	0.4	0.0	0.0	0.0	9.4
(2)	0.1	0.2	0.1	0.0	0.0	0.0	0.0	0.0	0.4
SW	8	16	11	2	0	0	0	0	37
(1)	3.0	6.0	4.1	0.7	0.0	0.0	0.0	0.0	13.8
(2)	0.1	0.3	0.2	0.0	0.0	0.0	0.0	0.0	0.7
WSW	5	8	9	5	2	0	0	0	29
(1)	1.9	3.1	3.5	1.9	0.8	0.0	0.0	0.0	11.2
(2)	0.1	0.1	0.2	0.1	0.0	0.0	0.0	0.0	0.5
W	9	7	22	26	12	2	0	0	78
(1)	2.9	2.3	7.1	8.4	3.9	0.6	0.0	0.0	25.2
(2)	0.2	0.1	0.4	0.5	0.2	0.0	0.0	0.0	1.4
WNW	44	63	80	49	19	1	0	0	256
(1)	9.0	12.9	16.4	10.1	3.9	0.2	0.0	0.0	52.6
(2)	0.8	1.1	1.5	0.9	0.3	0.0	0.0	0.0	4.7
NW	82	98	59	30	2	0	0	0	271
(1)	16.3	19.5	11.8	6.0	0.4	0.0	0.0	0.0	54.0
(2)	1.5	1.8	1.1	0.5	0.0	0.0	0.0	0.0	4.9
NNW	26	18	0	0	0	0	0	0	44
(1)	9.5	6.5	0.0	0.0	0.0	0.0	0.0	0.0	16.0
(2)	0.5	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.3
N	16	6	0	0	0	0	0	0	22
(1)	6.3	2.4	0.0	0.0	0.0	0.0	0.0	0.0	8.7
(2)	0.3	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.4
VRBL	14	2	0	0	0	0	0	0	16
(1)	5.7	0.8	0.0	0.0	0.0	0.0	0.0	0.0	6.5
(2)	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.3
ALL	360	447	240	131	41	5	0	0	1224
(1)	24.7	30.7	16.5	9.0	2.8	0.3	0.0	0.0	84.1
(2)	6.5	8.1	4.4	2.4	0.7	0.1	0.0	0.0	22.3

(1) = PERCENT FOR THIS STABILITY AND WIND DIRECTION  
(2) = PERCENT OF ALL GOOD OBS

8568 TOTAL PSBL HRS  
231 HRS OF CALM (4.2 PCT)

1455 HRS THIS PAGE (26.4 PCT)  
3047 HRS OF MSG WINDS/DELTA T



sector. The mean meteorological condition for all of the sectors around a facility is represented by the mean sector value. Normalized average air concentrations were then calculated as a function of distance. The averages of these mean curves for all sites are referred to as the mean sector curves. Figures 2 and 3 contain the CAMS and mean sector curves of  $\bar{X}/Q'$  as a function of distance for all sites for the ground level and 100 meter releases, respectively.

The CAMS curves were developed to be used as initial estimates of the annual average  $\bar{X}/Q'$  values that might occur at any site. Comparisons with independently calculated  $\bar{X}/Q'$  values<sup>(24)</sup> has shown that the CAMS curves may be used as realistic but conservative estimates of  $\bar{X}/Q'$  in the absence of actual site climatological data.

It is necessary to consider the applicability of meteorological data derived from nuclear power plant sites to a site for a plutonium fuel fabrication plant. The results can only be as good as the similarity of the sites. Moreover, plutonium fuel plants may be located under different siting criteria, and may well have different local topographical features. As noted earlier, the attempt to divide diffusion regimes by apparent local topographical differences showed that similar variations occurred in all groups. There were nearly equivalent extremes in all groups, suggesting that whatever makes a site "good" or "bad" cannot be expressed simply in terms of apparent topographical differences.

Although the most conservative sector (CAMS) has been used in this study, it is desirable to give credit for local site characteristics in specific applications. Conversely, it should be recognized that even though CAMS has been shown to be conservative in this study, it is not expected, in a statistical sense, that the CAMS curves will enclose the population of  $\bar{X}/Q'$  values.

In order to properly interpret the  $\bar{X}/Q'$  values calculated for the extreme distances presented herein, it must be understood that an upper limit has not been placed on vertical diffusion. Such a limit does naturally occur and is discussed by Holzworth.<sup>(25)</sup> For the United States

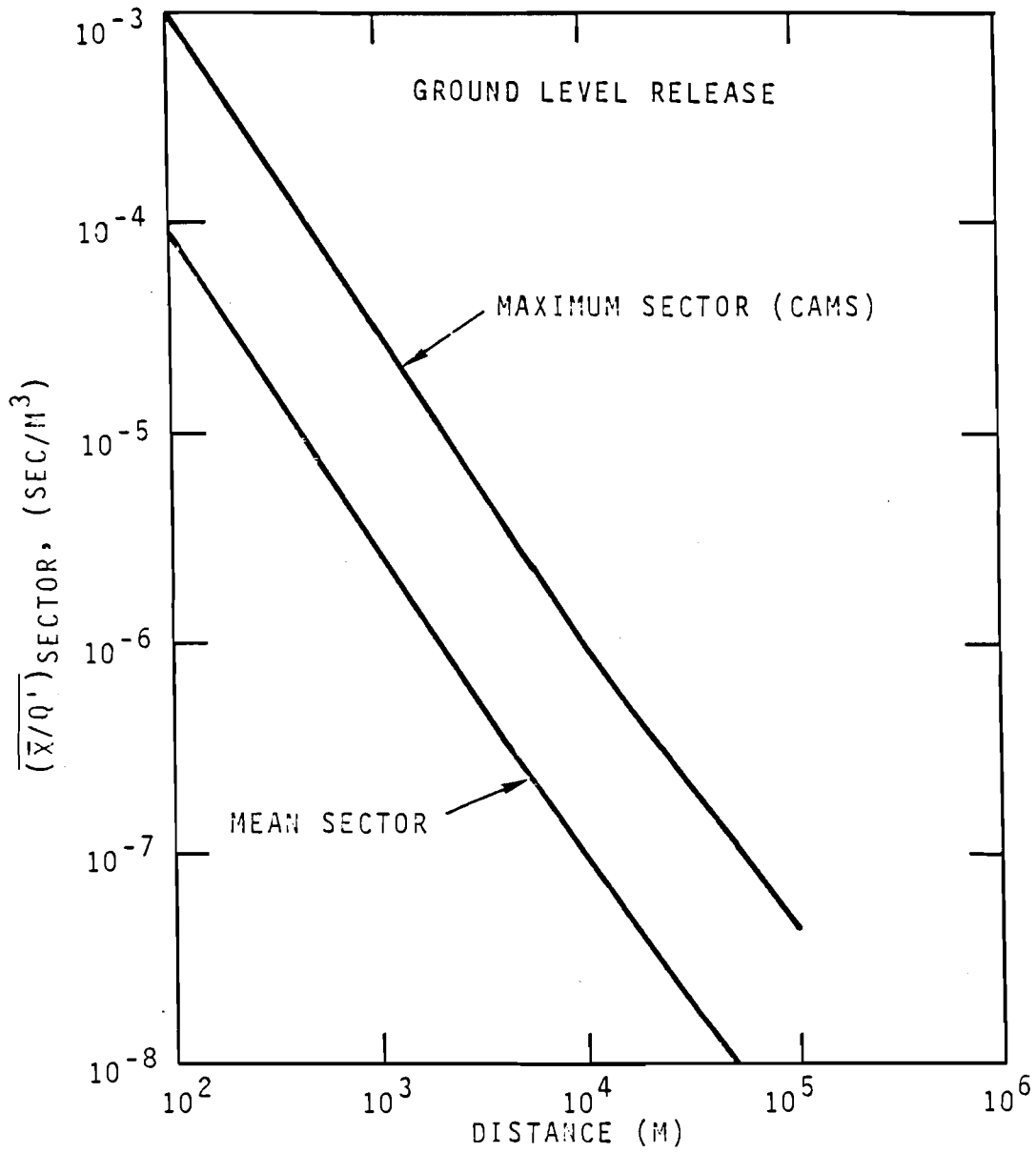


FIGURE 2. Maximum (CAMS) and Mean  $\bar{x}/Q'$  Curves for Ground Level Releases

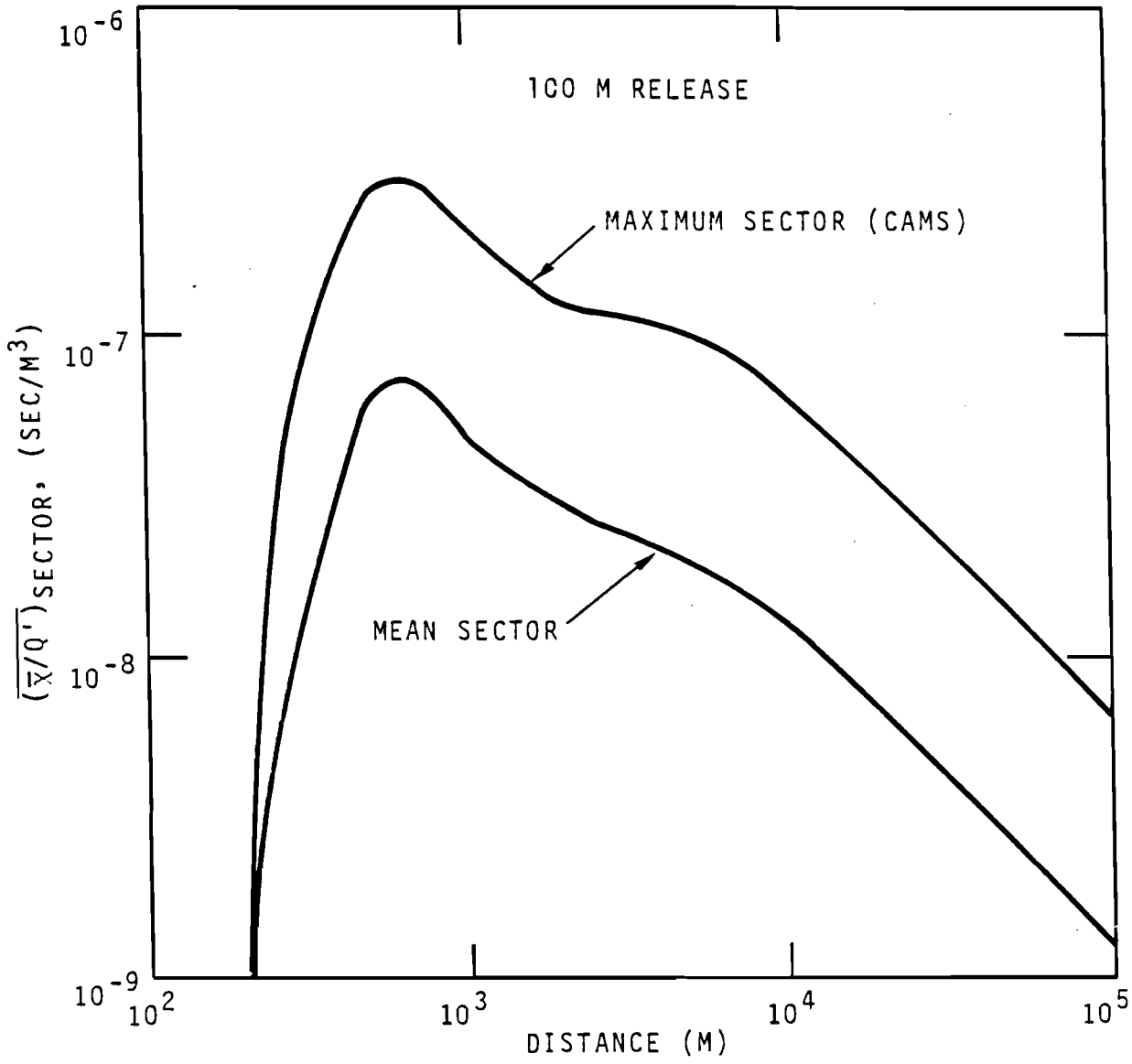


FIGURE 3. Maximum (CAMS) and Mean  $\bar{X}/Q'$  Curves for an Elevated Release (100 meters)

this limit is around 1000 meters. When no limit is placed on vertical diffusion, estimates of  $\bar{x}/Q'$  are affected at distances beyond  $6 \times 10^3$  meters. These values will be underestimated less than an order of magnitude at  $10^5$  meters. Allowance may be made for the mixing depth in any particular region by use of maps published by Holzworth.

The above method and the CAMS curves are not designed to replace in any way the need for on-site meteorological data collection programs. These are only for initial estimates for an undefined site for use in plant design. It is anticipated that once a particular site is chosen, on-site data will be collected and used to make a more accurate assessment of the diffusion climatology both for the routine as well as accidental release calculations.

## 2. Accidents

The method for estimating atmospheric dispersion of an accidental release must entertain the possibility that the release may occur during any diffusion regime. This is because releases resulting from accidents are inherently of short duration (a few minutes, up to a day in length) and random with respect to timing. A method for estimating the atmospheric dispersion of accidental releases from nuclear power plants is presented in USAEC Regulatory Guide 1.3. The enveloped maximum curve method discussed in the Guides should apply equally well to plutonium fuel plants. The method is clearly explained in the safety guides, and is in wide enough usage that no further detail on the method will be presented here. It should be recognized, however, that the climatological model employed in the Regulatory Guide estimates less dilution and therefore is more conservative than the previously discussed CAMS model for routine releases.

## 3. Effects of Dry Deposition, Scavenging, and Resuspension on Accidental and Normal Operation Release Consequences

Dry deposition, scavenging, and resuspension are important considerations in the evaluation of the consequences from accidental and normal operating releases. First, these factors, with exception of resuspension, reduce the immediate atmospheric concentration of plutonium. Second, some

plutonium will deposit on the ground where recovery or containment may be possible. Third, an accurate prediction of atmospheric air concentrations following an accident will require consideration of these mechanisms. The atmospheric dispersion curves for normal operation (CAMS) shown in Figure 2 do not separately consider the effects of these three mechanisms; deposition, scavenging, and resuspension.

The deposition velocity of plutonium particles cannot be specified exactly because it will vary depending on the size distribution of the particles, the nature of the surface on which deposition occurs, the wind speed, and other meteorological variables. In addition, the size distribution of the particles with which the plutonium is associated can be expected to change downwind as a result of a number of natural processes. For a particular situation, once a deposition velocity is known, an estimate may be obtained by use of the methods recommended in Meteorology and Atomic Energy (1968).<sup>(21)</sup>

Inspection of this method reveals that an order of magnitude change either way from the example deposition velocity of 1.0 cm/sec results in a drastic change in deposition. The deposition velocity for plutonium particles with an average diameter of 0.3  $\mu\text{m}$  is 0.01 to 0.20 cm/sec for wind speeds of 2 m/sec to 13 m/sec.<sup>(26)</sup> This particle size has the greatest fractional penetration through a HEPA filter and hence has been assumed to be applicable to the normal operation release as well as the accident release, assuming the HEPA filter remains intact. The value of the deposition velocity will also depend on the nature of the release, in that different size distributions may be generated. For an uncontained fire, Mishima determined the size of airborne particles from the oxidation of metallic plutonium was about 4.2  $\mu\text{m}$  with no particles greater than 8  $\mu\text{m}$  detected.<sup>(27)</sup> If other nonradioactive particles are produced at the same time, such as during a fire, a combination of particles can be expected. Higher deposition velocities would be applicable for such a release. However, as long as the HEPA filters are intact, the size distribution will be mostly a function of the properties of the exhaust system. Laboratory measurements suggest the size will be around 0.3  $\mu\text{m}$  as assumed

earlier. However, this value is open to some question based on experimental evidence. Actual measurements at a plutonium fuel plant showed the radioactive particles downstream from HEPA filters were an order of magnitude greater in size.<sup>(28)</sup> This in turn would result in increased deposition velocities and increased importance of dry deposition.

The range of maximum deposition on ground level surfaces near the plant perimeter may be estimated using the ground level CAMS curves and a reasonable deposition velocity for the released particles of 0.05 cm/sec at 300 meters. An annual release of  $y$  grams produces maximum annual deposition rates of  $y \times 9 \times 10^{-8}$  g/m<sup>2</sup>.

Deposition rates for an accident will have to be calculated on a case by case basis, taking into account the actual situation. The possible cases include nearly complete deposition within a fairly short range up to almost no deposition at all.

More stable atmospheric conditions, although having a lower deposition velocity, generally have higher actual deposition as a result of the dominating effect of higher air concentrations under stable conditions. An onsite measurement of atmospheric stability will be required as one of the inputs if the deposition from an accidental release is to be calculated. The magnitude of the change in deposition between a very unstable case and a very stable case is about a factor of two, at 10<sup>3</sup> meters.

A similar estimate may be made for the accident case using the curves from Safety Guides 3 and 4. For an accidental release the maximum ground level deposition at 300 meters may be expected to be in the range of  $y' \times 3 \times 10^{-6}$  and  $y' \times 6 \times 10^{-5}$  g/m<sup>2</sup>/hour where  $y'$  is the accidental release rate in grams per hour.

At any prospective site, the potential for scavenging by precipitation should be considered. Assuming a uniform distribution of precipitation in the region, the maximum deposition from routine releases should occur at or in the immediate vicinity of the site. The washout coefficient is defined as the constant of the time-dependent exponential decay

$$x = x_0 \exp(-\lambda t)$$

where  $\chi$  is the air concentration at time  $t$ ,  $\chi_0$  is the initial air concentration, and  $\Lambda$  is the washout coefficient.<sup>(29)</sup>

Experimental and theoretical evidence has shown the washout coefficient is about  $10^{-4}$  to  $10^{-3} \text{ sec}^{-1}$  for uranium particles that have median mass diameter of between  $6 \mu\text{m}$  and  $15 \mu\text{m}$ , respectively.<sup>(30)</sup> This implies that for a five minute rainfall, between 26% and 3% of a plant release would be deposited within 1500 meters of the plant assuming a 5 m/sec wind speed. The variation is primarily the result of different rainfall rates; the washout coefficient is approximately proportional to the rainfall rate. The main effect of the wind speed is in the dispersion of the release. The washout rate is also a function of the particle size. Theoretical models based on inertial effects alone predict smaller washout coefficients for smaller particles. Slinn has provided a theory that predicts higher washout coefficients for smaller particle sizes.<sup>(30,31)</sup> Further research is needed in this area to develop the theory and data for prediction of the potential effects of scavenging at a site based on precipitation climatology.

Any material that is deposited on a surface can be resuspended to the atmosphere by natural processes. Most experiments have been primarily concerned with resuspended particles over a contaminated area. The problem of downwind concentrations with nonradioactive materials have been considered by Sehmel.<sup>(32)</sup>

Resuspension rates for material deposited on the ground are time dependent and tend to decrease with increasing time after initial deposition. Local conditions can be expected to strongly affect the rate; rainfall, winds, and surface characteristics being predominant. The exact relationships are not well enough understood at this time to account for these effects. The nature of the process, as presently understood, suggests that initially the resuspension may be relatively high, but then becomes smaller as the particles weather into the soil. This suggests that a long-term buildup can be reasonably expected, but at some rate less than the deposit rate.

It has been assumed that the amount of material available for resuspension decreases with a half-life of 30 to 45 days. Therefore the level of "surface" contamination  $Q(\text{g}/\text{m}^2)$  will eventually come to equilibrium if the contamination is deposited on the surface over an extended period of time at a specified Rate  $P$  ( $\text{g}/\text{m}^2$  per year) in accordance with the following relationship.

$$Q(\text{g}/\text{m}^2) = \frac{P(\text{g}/\text{m}^2 \text{ year})}{\frac{0.693}{t_{1/2} \text{ (years)}}$$

The amount of material that is resuspended is given in terms of a resuspension factor,  $K$ , with units of  $\text{m}^{-1}$ .

$$K = \frac{\text{airborne concentration (units}/\text{m}^3)}{\text{surface concentration (units}/\text{m}^2)}$$

In a review of suspension factors obtained by a literature search, Mishima<sup>(34)</sup> tabulated measurements ranging from  $10^{-3} \text{ m}^{-1}$  to about  $10^{-6} \text{ m}^{-1}$  for several materials under different conditions with various degrees of physical activity in the area. Stewart<sup>(35)</sup> concludes that under quiescent outdoor conditions a factor of  $10^{-6} \text{ m}^{-1}$  is reasonable, and under conditions of moderate physical activity in the area a factor of about  $10^{-5} \text{ m}^{-1}$  would be appropriate, a further discussion of resuspension may be found in Appendix A.

The importance of resuspension in the exposure of the public may be estimated for the reference plant. Using the  $\bar{x}/Q'$  from Figure 2 of  $1.8 \times 10^{-4} \text{ sec}/\text{m}^3$  at 300 meters from the point of release a deposition velocity of 0.05 cm/sec ( $5 \times 10^{-4} \text{ m}/\text{sec}$ ) appropriate to a windspeed of about 3 m/sec, and an annual release rate of 5 mg/year, a depletion rate may be obtained:

$$1.8 \times 10^{-4} \text{ sec}/\text{m}^3 \times 5 \times 10^{-4} \text{ m}/\text{sec} \times 5 \times 10^{-6} \text{ g}/\text{yr} = 4.5 \times 10^{-13} \frac{\text{g}}{\text{m}^2} \text{ per year.}$$

over a period of years, assuming a fixation half-life of 30 days (0.082 years) the surface contamination level will come to equilibrium at



$$\frac{4.5 \times 10^{-13} \frac{\text{g}}{\text{m}^2 \text{ year}}}{\frac{0.693}{0.082} \text{ year}^{-1}} = 5.3 \times 10^{-14} \text{ g/m}^2.$$

If the half-life is taken to be 45 days, the equilibrium surface contamination level would be  $8 \times 10^{-14} \text{ g/m}^2$ .

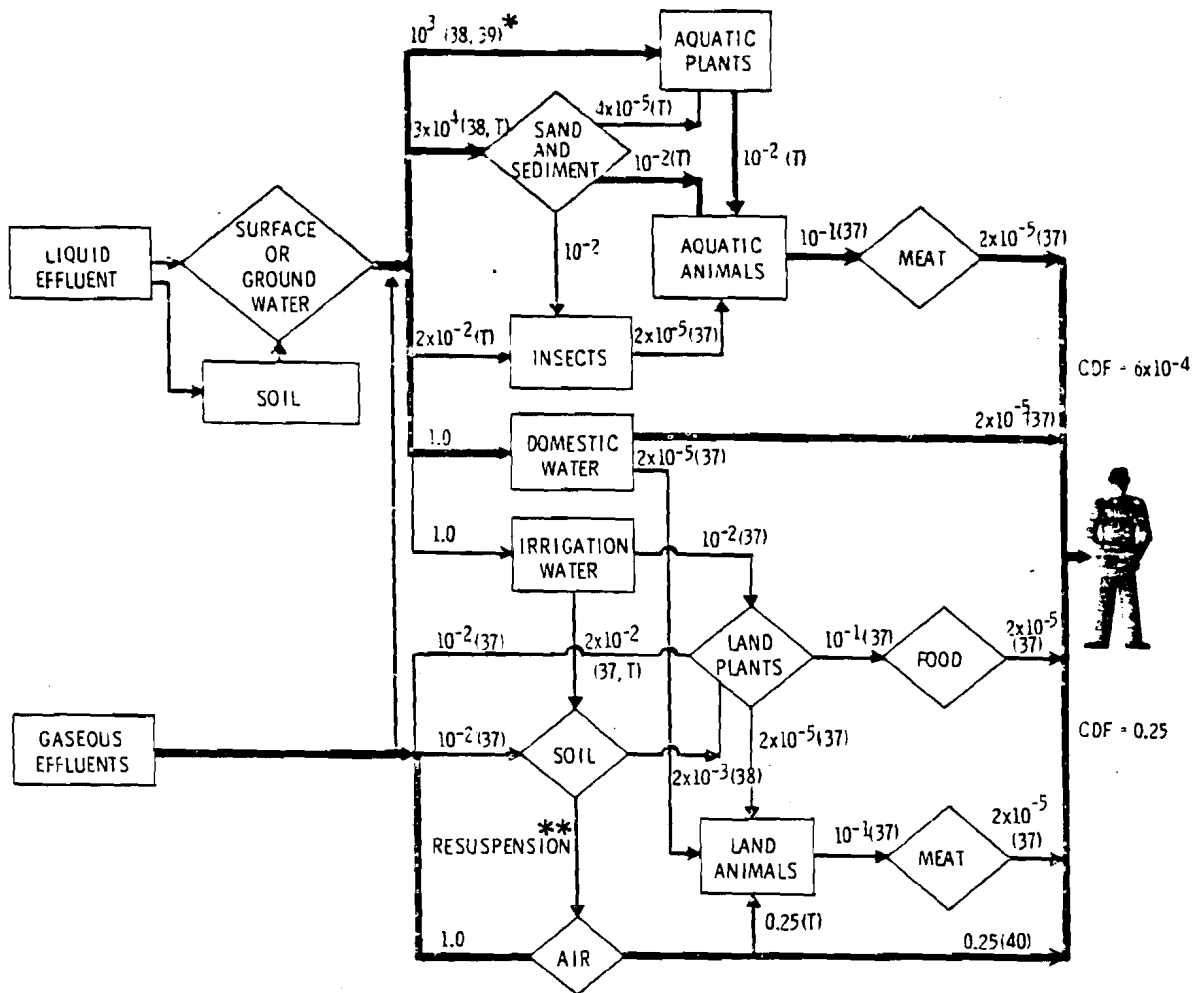
For mixture number III described in Table 23 Section X, with an alpha specific activity of  $4 \times 10^{-1} \alpha \text{ Ci/g}$ , this is 20 fCi $\alpha$  per square meter, or a factor of  $2 \times 10^{-6}$  below the "contaminated area" decision level proposed in Figure 33.

Using resuspension factors of  $10^{-6} \text{ m}^{-1}$  for quiescent conditions, and  $10^{-5} \text{ m}^{-1}$  for moderate physical activity conditions, airborne concentrations from resuspension would be  $5.3 \times 10^{-20} \text{ g/m}^3$  and  $5.3 \times 10^{-19} \text{ g/m}^3$  respectively. For mixture III this would be equivalent to  $7.5 \times 10^{-19} \mu\text{Ci/cm}^3$  under quiescent conditions and  $7.5 \times 10^{-18} \mu\text{Ci/cm}^3$  under moderate activity conditions.

#### B. PLUTONIUM PATHWAYS

Exposure transport routes to man were developed for plutonium as a means of locating points of expected long-term contaminant accumulations and points of population - contaminant interfaces.<sup>(36)</sup> Since the plutonium content is clearly limiting based on relative hazard of the two materials, (Pu and U), only plutonium exposure routes were considered. The more important plutonium pathways are shown in Figure 4.

Media appearing inside the diamond shapes on the figure are those commonly sampled in environmental surveillance programs. Individual discrimination factors (IDF) defined as the recipient to donor concentration ratio or as the fraction of incident contaminant retained by recipients are provided for each medium-to-medium transfer link. Identification of the predominant exposure pathways was made by propagating the IDFs into combined discrimination factors (CDF) for each recognized potential transport route from effluent release to human exposure. A CDF is defined as the product of all of the IDFs in an exposure pathway.



\* ( ) DENOTES THE SOURCE OF IDF'S. THE NUMERICAL VALUES ARE REFERENCE NUMBERS AND THE "T" MEANS THAT THE BASIS FOR THE VALUE IS DEVELOPED IN TABLE 5.

\*\* A UNITLESS VALUE IS NOT CURRENTLY AVAILABLE

FIGURE 4. Exposure Pathways and Discrimination Factors for Plutonium in the Biosphere

Discrimination factors which were not available from the literature were inferred by comparison as shown in Table 5. Two assumptions were made based on similar transport routes (noted in Table 5). The first assumption was that bottom-feeding aquatic animals exhibit the same IDF toward their sources of nourishment as do plant-feeding aquatic animals. Additionally, the irrigation water-soil-land plant pathway and the sediment-aquatic plant pathway were assumed to have the same IDF. Based on work by Langham,<sup>(37)</sup> an IDF of  $2 \times 10^{-5}$  was assigned to most animal ingestion processes,  $10^{-2}$  for surface contamination mechanisms, and  $10^{-1}$  for food processing. Other inferences indicated in Table 5 are based on work by Soldat<sup>(41)</sup> and Noshkin.<sup>(39)</sup> Exposure routes with CDF's less than  $10^{-5}$  were considered insignificant and are not shown on the diagram. Four transport routes have CDF's greater than  $10^{-5}$ ; these are indicated by heavy lines on the figure. The greatest CDF's for both liquid and gaseous effluent exposure pathways are indicated at the right of the figure.

TABLE 5. Discrimination Factors for Plutonium Pathways

Water	→	Aquatic Plants	→	Aquatic Animals	= 10	(38)
Water	→	Aquatic Plant			= $10^3$	(39)
		Aquatic Plants	→	Aquatic Animals	= $10^{-2}$	
Sand and Sediment	→			Aquatic Animals	= $10^{-2}$	(Assumption #1)
Water	→	Sand and Sediment	→	Aquatic Animals	= 300	(38)
Water	→	Sand and Sediment			= $3 \times 10^4$	
Irrigation Water	→	Soil	→	Land Plants	= $4 \times 10^{-5}$	(37)
Sand and Sediment	→	Aquatic Plants			= $4 \times 10^{-5}$	(Assumption #2)
Soil	→			Land Plants	= $2 \times 10^{-3}$	(38)
Irrigation Water	→	Soil			= $2 \times 10^{-2}$	
Air	→	Man			= 0.25	(40)
Soil	→	Air	→	Man	= $0.25 \times 10^{-5}$	
Soil	→	Air	→	Land Animals	= $0.25 \times 10^{-5}$	
Air	→	Land Animals			= 0.25	

The relative importance of each critical pathway was deduced by weighing the CDF for each route with quantities related to (a) the relative amounts of credible contaminants available to that pathway via environmental release of process materials and (b) the relative extent to which members of each route are used by human populations. By this process, the most important exposure pathway identified is inhalation of airborne plutonium. The domestic water and aquatic animal pathways are at least four orders of magnitude less significant.

Tables 6, 7 and 8 list the radiation dose rates (mrad/sec) received by certain aquatic organisms and their predators from exposure to a unit water concentration ( $\text{g}/\text{m}^3$ ) of the mixtures of transuranic nuclides specified by L. G. Faust as available for fuel manufacturing in the years 1975, 1980 and 1985 (Appendix C). These dose rates can be interpreted in terms of integrated dose per unit of "exposure" (mrad per  $\text{g}\cdot\text{sec}/\text{m}^3$ ) from a short-term accident situation, provided one is willing to make the highly conservative assumption that there is an instantaneous equilibrium between the water concentration, the organisms' diet and the organism itself. Because of the time lag in transfer through the trophic levels the body burdens of the organisms and the resulting doses will be less for short term exposure than estimated via the above method.

The formula and computer code CRITR used for these calculations has been described in an AEC report BNWL-1754.<sup>(41)</sup> Basically, the program first converts aqueous effluent release rates to water concentration (by applying mixing ratios to flow data) and then to concentration in the organism (by applying bioaccumulation factors) and finally to dose rate (utilizing effective energies per disintegration of the nuclide in the size of organism of interest) through Equation 1 below.

$$\begin{aligned}
 (\text{D.R.})_i &= 18.7 C_i E_i \\
 &= 18.7 W_i (B_A)_i \bar{E}_i
 \end{aligned}
 \tag{1}$$

TABLE 6. Estimated Dose Rates to "Aquatic" Organisms from Transuranium Nuclides in Water  
[mrad/sec per (g of mixture) / (m<sup>3</sup> of water)]\*

<u>Organism</u>	<u>Summary</u>					
	<u>Fresh Water</u>			<u>Salt Water</u>		
	<u>1975</u>	<u>1980</u>	<u>1985</u>	<u>1975</u>	<u>1980</u>	<u>1985</u>
Fish	5.2	6.9	7.4	4.8	6.4	6.8
Invertebrates	180	230	250	260	350	370
Algae	770	990	1100	1300	1700	1800
Muskrat	2.2	2.7	2.9	2.9	3.7	4.0
Heron	0.017	0.021	0.023	0.016	0.020	0.022

\* or mrad per (g sec/m<sup>3</sup>) assuming instantaneous equilibrium between organisms and water.

TABLE 7. Percent of Doses to Aquatic Organisms Contributed by Pu Isotopes\*

<u>Organism</u>	<u>Fresh Water</u>			<u>Salt Water</u>		
	<u>1975</u>	<u>1980</u>	<u>1985</u>	<u>1975</u>	<u>1980</u>	<u>1985</u>
Fish	54	57	57	50	54	53
Invertebrates	45	49	48	62	66	65
Algae	37	40	40	62	66	65
Muskrat	18	20	20	39	42	42
Heron	31	34	33	28	31	30

\* The remainder of the dose is due to <sup>241</sup>Am.

TABLE 8. Percent of Pu Dose Contributed by Individual Isotopes (All Organisms, Fresh or Salt Water)

<u>Nuclide</u>	<u>1975</u>	<u>1980</u>	<u>1985</u>
$^{238}\text{Pu}$	62*	72	74
$^{239}\text{Pu}$	15	9.2	8.4
$^{240}\text{Pu}$	19	15	14
$^{241}\text{Pu}$	4.0	3.5	3.6
$^{242}\text{Pu}$	0.0035	0.050	0.072

\* For example the dose to fresh water fish from  $^{238}\text{Pu}$  in the 1975 mixture is  $(5.2 \text{ mrad}) \times (0.62) \times (0.54) = 1.7 \text{ mrad}$

where  $(D.R.)_i$  = the dose rate to an organism from a deposition of nuclide i (mrad/yr)

$C_i$  = the concentration of nuclide i in the organism (pCi/kg)

$\overline{E}_i$  = the effective energy per disintegration of nuclide i in the particular size of organism involved (MeV/dis)

$W_i$  = the concentration of nuclide i in the water (pCi/liter)

$(B_A)_i$  = the bioaccumulation factor for nuclide i from water to the organism (pCi/kg per pCi/liter)

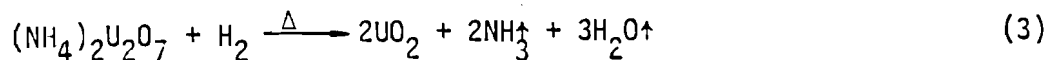
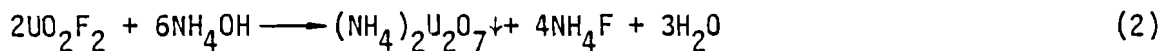
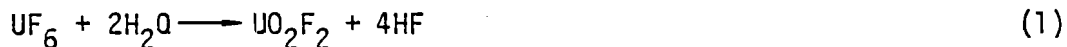
For the calculation of dose to predators, the concentration in diet is calculated from  $W$  and  $B_A$  and then the radionuclide intake rate is estimated. The accumulated body burden and dose rate is then calculated as ICRP calculates dose rate to man, except the appropriate values for mass and effective decay energy are substituted. Since little or no data on fractional uptake and effective half-life in these organisms are available, standard man values are assumed for these two parameters. In all instances the dose is calculated in rads because the factors of DF and QF are not applicable to organisms other than man.

## C. CHEMICAL EMISSIONS

A review of the processes which will be used in a mixed oxide facility indicates that the main chemical contaminants will be compounds of fluorine and nitrogen. Although chemicals used in the process will appear in both liquid and gaseous streams, attention is given exclusively to the airborne particles and gases. Liquid streams must meet stringent Federal and State release limits and license applicants will be required to obtain discharge permits if discharges are to natural water bodies. The principle of "zero" release will be followed to such a degree that evaporation and recycling will virtually eliminate liquid waste streams beyond the confines of the plant perimeter. Some gases and airborne particulate material will be available for inadvertent and routine releases. Sources, characteristics and standards for anticipated chemical contaminants are described below.

### 1. Fluorine Compounds

A potential exists for emissions of gaseous and particulate fluorine compounds from nuclear fuel fabrication plants. The primary source of fluorine is the starting compound  $UF_6$  which is converted to uranium oxide. One process used is to hydrolyze the  $UF_6$  in water and precipitate ammonium diuranate, which is then separated, dried and calcined to the oxide. This conversion is shown in the following equations:



A large fraction of the fluoride ion will remain in the aqueous waste streams. During the calcining process, gaseous compounds of fluorine and inorganic fluoride particles will be evolved in relatively small amounts.

All processes giving rise to gaseous fluorine compounds will be required to have efficient caustic scrubbers or other highly efficient methods for removing HF.

Particles generated from sprays, sintering, grinding, etc., will be filtered through two high efficiency filters. If caustic scrubbers are not efficient and high fluorine concentrations reach HEPA filters, severe degradation of the filters may result. However, there appears to be a limit to the concentrations of fluorine that can exist in the exhaust system due to the chemical reactivity of fluorine with the structural material. Perspective can be gained for the significance of fluorides that could be released by accounting in a realistic manner for the attenuation of fluoride through the process. Exact partitioning between solid-liquid and solid-gas is not known for the processes, but good approximations are possible. If a plant of 1 MT (2206 lb) of  $UO_2$  per day is assumed, the starting  $UF_6$  will be 2875 lb of which 929 lb is fluorine.

It is reasonable that no more than 10% will remain with the solid product, and the remainder will appear in the waste water. The calcining can be counted on to remove all except traces of the fluoride. The fluorides passing into a scrubber will thus be about 90 lb/day. Assuming that all the fluoride is a gas and the scrubber efficiency is  $\bar{99\%}$ , about  $(0.01) (90) = 0.9$  lb of fluoride (HF) per day will penetrate the scrubber.

A uniform release rate of 0.9 lb per day (410 g per day) from a 30 m stack would produce a ground concentration of about  $1.5 \times 10^{-7}$  g/m<sup>3</sup> if a 5 m/sec wind and slightly unstable air stability are assumed. In reference to fluoride standards to be discussed, it is not likely that fluoride levels released from the reference plant would be of great concern. There may be occasional episodes during off-standard operations which may momentarily increase levels.

Fluoride is an accumulative toxicant, and vegetation injury can be associated with accumulation over a long period. During chronic exposures, fluoride is gradually absorbed, transported, and concentrated in the tips and margins of the leaves. The amount of fluoride accumulated, and the extent of injury to the plant, is determined not only by fluoride concentration and exposure duration, but also species sensitivity, absorption capacity and plant maturity, as well as climatic and environmental factors



such as temperature, humidity, light intensity, precipitation, and soil type.<sup>(42)</sup> Injury to plants apparently depends also on the rate of fluoride absorption by the tissues. Severe defoliation and other damage occurs in plants when tissue fluoride concentrations rapidly accrue. If the same concentrations were accumulated over an extended period, little or no effect would be induced.

Flora are frequently categorized into three classes as to sensitivity to atmospheric fluoride or tissue concentrations: as susceptible, intermediate, and tolerant species; although it is recognized that differences in tolerance by individual species within each grouping is great and dependent on the variables previously described. In a review<sup>(43)</sup> of the current information on the concentrations in air of gaseous fluoride causing foliar markings, threshold levels were described as: (1) 3-4  $\mu\text{g}/\text{m}^3$  for the most susceptible species and 10  $\mu\text{g}/\text{m}^3$  or higher for species of intermediate susceptibility, for exposures of one day; and (2) for exposure periods longer than a month, about 0.5  $\mu\text{g}/\text{m}^3$  for the most susceptible and between 1-3  $\mu\text{g}/\text{m}^3$  for some intermediate species. It was also noted that although the relationship of accumulated foliar fluoride to the occurrence of lesions is complex, susceptible plant species show foliar lesions when tissue fluoride concentrations range from 20-150 ppm. The Washington State Airborne Fluoride Standards shown in Table 9 appear to be consistent with the experimental data and the concentrations cited as protective for most plant species,<sup>(44,45)</sup> although it has been noted that unique circumstances may exist in which the values may be either over- or under-protective.<sup>(46)</sup>

In summary, the considerations that need to be given in terms of important pathways are the effects of fluorides on animals via feed crops such as alfalfa, for which limits are listed in Table 10, and the growing of certain ornamental plants such as evergreens and gladioli for commercial use where fluorides have a detrimental effect. It appears certain that controls can be imposed to meet a restrictive limit on fluoride releases.

TABLE 9. State of Washington Ambient Air Fluoride Standards

Gaseous fluorides in the ambient air calculated as HF at the standard conditions shall not exceed:

- 3.7  $\mu\text{g}/\text{m}^3$  average for any 12 consecutive hours;
- 2.9  $\mu\text{g}/\text{m}^3$  average for any 24 consecutive hours;
- 1.7  $\mu\text{g}/\text{m}^3$  average for any 7 consecutive days;
- 0.84  $\mu\text{g}/\text{m}^3$  average for any 30 consecutive days;
- 0.5  $\mu\text{g}/\text{m}^3$  average for the period March 1 through October 31 of any year

TABLE 10. Washington State Fluoride Standards<sup>(44)</sup>

Fluoride content of dry forage shall not exceed:

- 40 ppm fluoride ions (averaged over a month), each month for a period of 12 consecutive months.
- 60 ppm fluoride ions for more than two consecutive months.
- 80 ppm once in any two consecutive months.

Dry forage to be sold shall not exceed 40 ppm fluoride ions.

If dry forage is used only part of the year, the fluoride content of the dry forage may be average over a 12 month period so that the rate is no more than 40 ppm per month.

## 2. Nitrogen Compounds

Two constituents evolved in the process are ammonia,  $\text{NH}_3$ , and nitrogen dioxide,  $\text{NO}_2$ .

### a. Ammonia

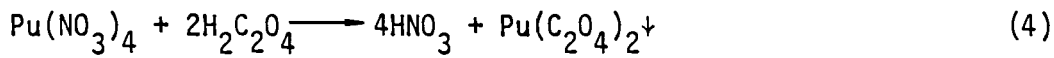
Ammonium hydroxide is used to convert the hydrolyzed  $\text{UF}_6$  to ammonium diuranate according to Equation (2). Reaction vessel vents will carry away ammonia from this system. Larger quantities of ammonia will be generated from the subsequent thermal reduction of the ammonium salt as shown by equation (3). Ammonia is a very soluble gas and should be readily removed by a scrubber to innocuous levels. Estimates of ammonia

evolution can be made to support this conclusion. The ammonia to be disposed for the 1 MT of fuel per day reference plant will be about 270 lb per day. If 99% is removed by a scrubber, about 2.7 lb/day would appear in the airborne effluents. The resulting maximum downwind concentration near the ground would be about  $0.4 \mu\text{g}/\text{m}^3$ , if uniform release is assumed. The Threshold Limit Value is  $35 \text{ mg}/\text{m}^3$ , five orders of magnitude greater than that estimated to be present with the assumptions used.

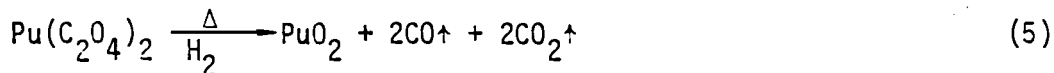
Ammonia is in the ambient air primarily due to decaying organic material, and the use of fertilizers. Junge cites research to show that ambient air contains from a fraction of a  $\mu\text{g}$  per  $\text{m}^3$  to  $14 \mu\text{g}/\text{m}^3$ .<sup>(47)</sup> Other work has shown that ammonia in rain represents about 60% of the total nitrogen present. Ammonia and nitrates were collected in rain at the monthly rate of 5.8 lb per acre.<sup>(48)</sup> Ammonia is used widely as a fertilizer and it seems highly unlikely that the anticipated concentrations would have any environmental effects above those from ammonia already present.

b. Nitrogen Oxides

In the conversion of plutonium nitrate to plutonium oxalate, nitric acid is formed per equation (4):



The precipitated plutonium oxalate is removed, carrying a small quantity of nitric acid. The oxalate is calcined in a stream of hydrogen, yielding  $\text{PuO}_2$ , CO,  $\text{CO}_2$  and  $\text{NO}_x$  from the carry-over of acid.



The evolution of nitrogen oxides from calcining  $\text{Pu}(\text{NO}_3)_4$  to the oxide can be shown to be an almost inconsequential release. For the reference plant producing a  $\text{UO}_2 - 4\% \text{ PuO}_2$  fuel, about 44 lb of plutonium or

about 108 lb of  $\text{Pu}(\text{NO}_3)_4$  will be calcined daily. The  $\text{NO}_2$  evolved would be about 7 lb/day. If this were released uniformly in a 5 m/sec wind, under slightly unstable meteorological conditions, the downwind concentration would be about  $1.1 \times 10^{-3} \text{ mg/m}^3$ . The national ambient air standard for  $\text{NO}_2$  is  $0.1 \text{ mg/m}^3$ , or two orders of magnitude greater than that estimated. (49) The Threshold Limit Value is  $9 \text{ mg/m}^3$ . (50) Conversion to oxide will likely be a batch operation, rather than a continuous one; hence, higher concentrations could be experienced over a shorter time period, but it is extremely unlikely that nitrogen dioxide concentrations will be of any concern. No credit was taken for reduction of the source by scrubbing. The scale of the operation is such that a compact water scrubber could achieve a factor of 10 lower release.

### 3. Accident Considerations - Toxic Chemicals

A few toxic chemicals are used in the fuel fabrication process. These are the usual industrial chemicals for which standard safe handling procedures should greatly limit the potential for accidental release. An applicant for a license to operate a fuel fabrication plant would be required to address the impact of accidents involving these materials on the plant and the environment. The chemicals identified which would be of concern are the following:

#### a. Uranium Hexafluoride, $\text{UF}_6$

Although a radioactive material,  $\text{UF}_6$  should also be regarded as a volatile, toxic chemical which could be released in a serious accident such as a gasoline fire following a break in the container. Such release might occur in an involvement of a  $\text{UF}_6$  carrier with a gasoline tanker truck.  $\text{UF}_6$  is a white solid at temperatures below  $133.5^\circ\text{F}$  and at atmospheric pressure sublimates at temperatures above this. Under pressure and at temperatures above  $147^\circ\text{F}$ ,  $\text{UF}_6$  is a clear liquid. When released to the air,  $\text{UF}_6$  is converted quickly to uranyl fluoride,  $\text{UO}_2\text{F}_2$ , and HF gas. A fine smoke is produced on release of  $\text{UF}_6$  to the atmosphere. Uranium compounds such as  $\text{UO}_2\text{F}_2$  are toxic to the kidneys when inhaled or ingested, and the chemical toxicity is more important than the toxicity due to radioactive

constituents, up to an enrichment of about 10%.<sup>(50)</sup> The Threshold Limit Value for uranium is  $0.2 \text{ mg/m}^3$  as uranium, and for fluorides is  $2.5 \text{ mg/m}^3$  as fluorine. These values represent the 40 hour time weighted average concentrations to which personnel may be repeatedly exposed day after day without adverse effect. The values appropriate to the accident case are those obtained by multiplying the TLV by the appropriate excursion factors, 3 for uranium and 2 for fluorides. The excursion TLV values are therefore  $0.6 \text{ mg/m}^3$  for uranium and  $5 \text{ mg/m}^3$  for fluorides.

Accidents which could result in significant releases of  $\text{UF}_6$  will involve rapid container failure while the  $\text{UF}_6$  is in liquid form. Because the handling systems are pressurized and the  $\text{UF}_6$  is in liquid form, a cylinder failure during transfer operations would result in a liquid spill or forceful injection to the air and surroundings. A significant external force would be required to bring about breaching of a container, and must be regarded as a very low probability event since no events of this kind have occurred in over 20 years of experience with shipping and processing  $\text{UF}_6$  in thousands of cylinders.<sup>(51)</sup>

The rate of release from a serious accident involving  $\text{UF}_6$  cannot be determined with any confidence because of lack of examples. The downwind airborne concentration at ground level can be estimated as shown in Table 11 for a release of 1 g/sec, which probably would be within a factor of 10 of a serious accident release rate.

TABLE 11. Dispersion Characteristics of a Ground Release Plume Centerline

Distance Downwind Meters	$xu/Q$ <sup>(51)</sup>					
	A Stability	B Stability	C Stability	D Stability	E Stability	F Stability
100	$8.5 \times 10^{-4}$	$6 \times 10^{-3}$	$4 \times 10^{-3}$	$8.2 \times 10^{-3}$	$5 \times 10^{-2}$	$3.3 \times 10^{-2}$
1000	$2.5 \times 10^{-6}$	$1.9 \times 10^{-4}$	$5 \times 10^{-4}$	$1.5 \times 10^{-4}$	$3 \times 10^{-4}$	$7 \times 10^{-4}$

If a 5 m/sec wind and the stability class with the lowest dilution factors (Class F) are assumed, a 1 g/sec release will result in a center-line ground concentration of about  $7 \text{ mg/m}^3$  at 100 meters, and  $0.15 \text{ mg/m}^3$  at 1000 meters. The excursion TLVs of  $0.6 \text{ mg/m}^3$  of uranium and  $5 \text{ mg/m}^3$  of fluoride would be exceeded at 100 meters but not at 1000 meters, at a release rate of 1 g/sec.

These estimates are made to suggest the levels of release which may be of concern in an accident involving the  $\text{UF}_6$ . Further definition of the hypothetical accident and the meteorological situation prevailing would be necessary in a detailed site selection study.

b. Ammonia,  $\text{NH}_3$

Ammonia can be used in large amounts as a source of hydrogen for the conversion step to prepare uranium dioxide (see equation 3). Many large cylinders of liquid ammonia would be available and subject to the same potential accidents as for the  $\text{UF}_6$  containers. Ammonia is a gas at ordinary temperatures and would disperse rather quickly. Applying the same release assumption of 1 g/sec released, results in a downwind ground level concentration of  $7 \text{ mg/m}^3$  at 100 meters. The excursion TLV factor for ammonia is  $27 \text{ mg/m}^3$  or about four times the indicated concentrations. Good industrial safety practices should virtually eliminate the possibility of a serious ammonia release.

c. Other Acids, Chemicals

Small amounts of nitric acid, hydrochloric acid, and ordinary chemicals are used routinely. These should not pose a problem from accidental release into the environment due to their low volumes.

## VI. DOSE CALCULATIONS

The behavior of material released from a plutonium fuels fabrication facility has been discussed in the previous section. Environmental consequences of atmospheric emissions from normal plant operation were examined using dispersion characteristics for a calculated annual maximum sector (CAMS).<sup>(52)</sup> Atmospheric dispersion following accidental releases was assumed to be the same as that postulated in USAEC Regulatory Guide 1.3. Doses were estimated from the 8 hour release curves for the accidents postulated in this report.

As concluded in a previous section on pathways, inhalation of the airborne plutonium is the most important exposure pathway for plutonium releases from a fuels fabrication facility. The organs of interest for this pathway are the bone for soluble plutonium and both lung and bone for insoluble plutonium.

### A. DOSE MODELS

Two lung models have been recommended by the ICRP. The initial lung model, recommended in ICRP Publication 2 and hereinafter referred to as the ILM, treats the inhaled material as either soluble or insoluble. When the material inhaled is soluble, then the uptake by other organs is assumed to be essentially instantaneous. The more sophisticated lung model, recently recommended in ICRP Publication 19 and hereinafter referred to as the TGLM, treats the inhaled material in a more complex way. The derived equations for estimating the dose to organs other than the lung are considerably more complex than those for the ILM. A computer program has been developed for calculating the dose to lung and other organs using the TGLM<sup>(53)</sup> and the results are compared with those obtained using the ILM. The reader is referred to the reference document for a detailed discussion of the TGLM computer program.

Two equations for calculating the dose to organs of interest via inhalation are presented for the emissions postulated from the reference facility; a continuous intake equation for normal operation and an equation for intake

over a short time interval for accidents. They are generally formulated to permit dose estimates for various model parameters and at different distances from the point of release. Relevant parameters in the dose models are particle size, inhalation time, human ventilation rate, chemical form, fractional uptake by the organ of interest, and the effective half-life of the deposited material. Additionally, the dose to the organ of interest from plutonium deposited in the organ is highly dependent on the isotopic composition. The isotopic composition of the plutonium assumed for the reference facility and the dose parameters used in this analysis are shown in Table 12.

The dose to the lungs and bones were calculated using two models which have been recommended by ICRP.<sup>(54,55)</sup> In lieu of detailed information, the following release parameters have been assumed:

- The particle size for all the released material was assumed to be in the respirable range.
- The inhaled material was assumed to be soluble when estimating the dose to the bone and insoluble when estimating the dose to the lung.
- The time of inhalation was assumed to be the same as the duration of release.

#### 1. Normal Operation Model

The 50 year dose to organs of interest via inhalation using the ILM from a continuous atmospheric release of a radionuclide is given by:

$$D = f_a k (P/\lambda_e) \left\{ \lambda_e t - [1 - \exp(-\lambda_e t)] \right\}$$

$$P = B Q' (\bar{X}/Q') 10^6$$

$$D = B (f_a k) (Q'/\lambda_e) (\bar{X}/Q') \left\{ \lambda_e t - [1 - \exp(-\lambda_e t)] \right\} 10^6$$

where:

- D - dose to organ of interest delivered over time t, rem
- $f_a$  - fractional uptake, via inhalation, by organ of interest
- k - dose conversion factor in rem per  $\mu\text{Ci}$  in organ of interest



- $P$  - inhalation rate,  $\mu\text{Ci}/\text{d}$   
 $\lambda_e$  - effective elimination rate constant,  $\text{d}^{-1}$   
 $\lambda_e = \lambda + \lambda_b$   
 $\lambda$  - radioactive decay constant,  $\text{d}^{-1}$   
 $\lambda_b$  - biological elimination rate constant,  $\text{d}^{-1}$   
 $t$  - duration of release/exposure,  $\text{d}$   
 $B$  - ventilation rate for standard man,  $\text{m}^3/\text{d}$  [ $20\text{m}^3/\text{d}$  (24 hour average rate)]  
 $Q'$  - atmospheric release rate,  $\text{Ci}/\text{sec}$   
 $\bar{X}/Q'$  - sector average dilution rate at ground level;  $\text{sec}/\text{m}^3$  as defined in Section V-A.

TABLE 12. Dose Parameters Used in Calculation Models

Nuclide	Half Life, Yr.	Activity Ci/g of Nuclide	Reference <sup>(a)</sup> Mixture, wt%	Activity Ci/g of Mix	Activity in Mix ( $\mu\text{Ci}$ ), %	Dose Conversion Factor <sup>(b)</sup>		
						$\left(\frac{\text{rem}}{\mu\text{Ci}\cdot\text{y}}\right)$ BONE <sup>(c)</sup>	$\left(\frac{\text{rem}}{\mu\text{Ci}}\right)$ LUNG ILM <sup>(c)</sup>	$\left(\frac{\text{rem}}{\mu\text{Ci}}\right)$ LUNG IGLM <sup>(d)</sup>
Pu-236	2.85	550	$7.3 \times 10^{-8}$	$4.0 \times 10^{-5}$	$2.9 \times 10^{-5}$			
Pu-238	87.8	17.5	1.9	0.33	2.4	750	500	2100
Pu-239	$2.44 \times 10^4$	0.0616	63.	0.039	0.28	720	470	2000
Pu-240	$6.54 \times 10^3$	0.227	19.	0.043	0.31	720	470	2000
Pu-241	15.	113	12.	13.6	97.	37	0.47	1.8
Pu-242	$3.87 \times 10^5$	0.00391	3.8	0.00015	0.0011	670	450	1900
Am-241	433.	3.25	0.60	0.020	0.14	750	500	2100
Reference Mixture				14. ( $\mu, \delta$ ) 0.43 ( $\mu$ only)		59	16	67

- a. Does not add to 100% because isotopic data are accurate to only two significant figures.  
 b. For material deposited in organ of interest.  
 c. ICRP Pub 2.  
 d. ICRP Pub 19.

## 2. Accident Model

The dose to the organ of interest via inhalation again using the ILM, from a radionuclide accidentally released to the atmosphere is given by:

$$D = f_a k P_s \left\{ [1 - \exp(-\lambda_e t)] \right\}$$

$$P_s = b Q^{-1} T (E/Q) = b Q (E/Q)$$

$$D = b (f_a k) Q (E/Q) \left\{ [1 - \exp(-\lambda_e t)] \right\}$$

where:

- D - dose to organ of interest delivered over time t, rem
- $f_a$  - fractional uptake, via inhalation by organ of interest
- k - dose conversion factor for organ of interest, rem per  $\mu\text{Ci}$  in organ
- $P_s$  - quantity inhaled,  $\mu\text{Ci}$
- $\lambda_e$  - effective elimination rate constant,  $\text{d}^{-1}$
- t - dose time, d
- b - ventilation rate for standard man,  $\text{cm}^3/\text{sec}$   
b =  $350 \text{ cm}^3/\text{sec}$  (8 hour working rates)  
=  $230 \text{ cm}^3/\text{sec}$  (24 hour daily rate)
- $Q^{-1}$  - atmospheric release rate,  $\text{Ci}/\text{sec}$
- T - duration of release exposure, sec
- Q - quantity released, Ci
- E/Q - time integrated air concentration normalized to quantity released,  $\text{Ci}\text{-sec}/\text{m}^3$  per Ci

$$\frac{E}{Q} = \frac{1}{\pi \sigma_y \sigma_z \bar{u}_h} \exp[-(y^2/2\sigma_y^2 + h^2/2\sigma_z^2)]$$

### B. NORMALIZED DOSE CURVES

The dose to the organ of interest per unit mass of radionuclide or reference mixture released was estimated by combining dose and atmospheric dispersion models for both continuous and short duration releases. Dose

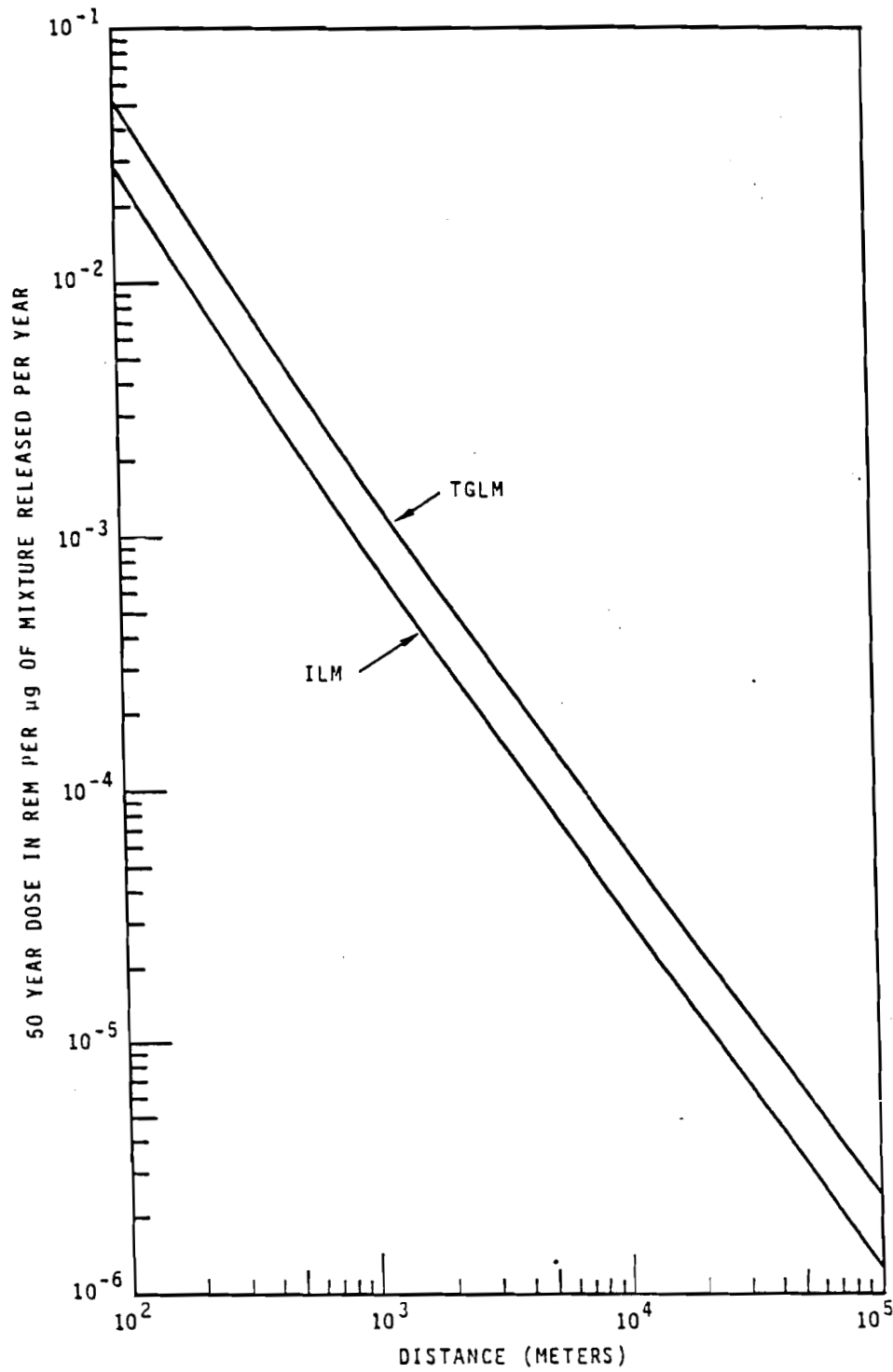
calculations for the lung and bone were made using both ICRP recommended lung models: the ILM and TGLM. The CAMS curves were used for an estimate of the atmospheric dispersion for the normal operation releases and Regulatory Guide 1.3 was used for the accident releases. When dealing with a specific site, meteorological data for that site should be used.

#### 1. Normal Operation Releases Curve

The 50-year doses to the bone and lung from continuous emission from the reference facility using the ILM and the TGLM are shown in Figures 5 through 8. Release heights corresponding to ground level and 100 meter release are illustrated. The curves are normalized to a constant release rate of 1  $\mu$  gram of reference mixture per year.

#### 2. Accident Release Curves

A comparison is made between the dose estimates for the bone and the lung using the ICRP recommended lung models for short duration releases at ground level. Only the 8 hour release curves are shown but the relationship remains about the same for the other release times. Figures 9 and 10 show the difference between the dose estimate using the two lung models for the bone and the lung, respectively. The treatment of soluble and insoluble material by the two lung models can be seen by examining the two figures. No curve is shown in Figure 10 for the dose commitment to the lung from inhalation of the reference mixture in soluble form. The ILM has no provisions for estimating the lung dose from soluble material. The remainder of the accident release curves, both for the bone and the lung, were developed using the ILM.



**FIGURE 5.** Comparison of the Dose to Bone from Inhalation of a Continuous Release of the Reference Mixture (Soluble) Using the Two ICRP Recommended Lung Models, Ground Level Release, Maximum Sector Curves

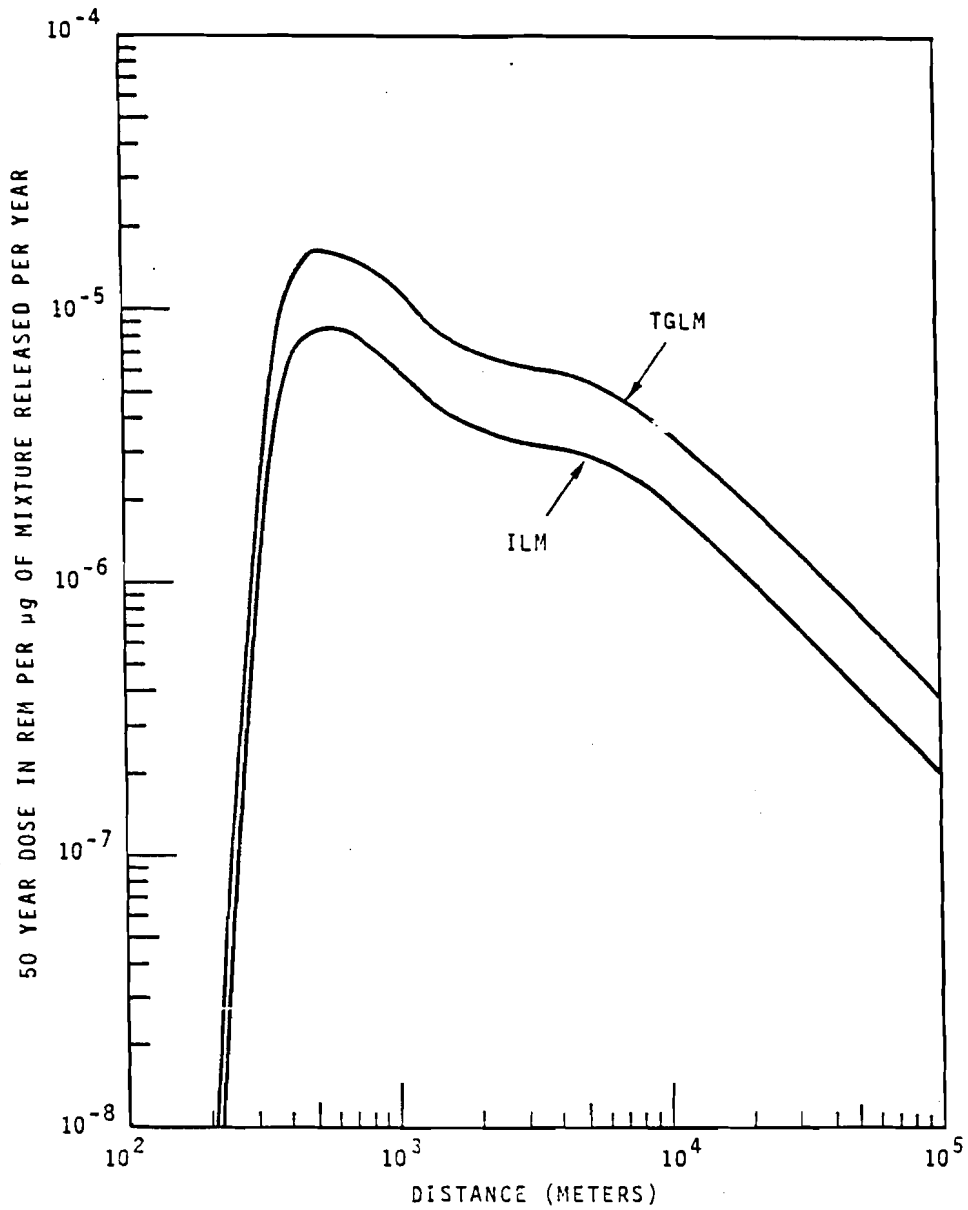


FIGURE 6. Comparison of the Dose to Bone from Inhalation of a Continuous Release of the Reference Mixture (Soluble) Using the Two ICRP Recommended Lung Models, Release Height 100 m, Maximum Sector Curves

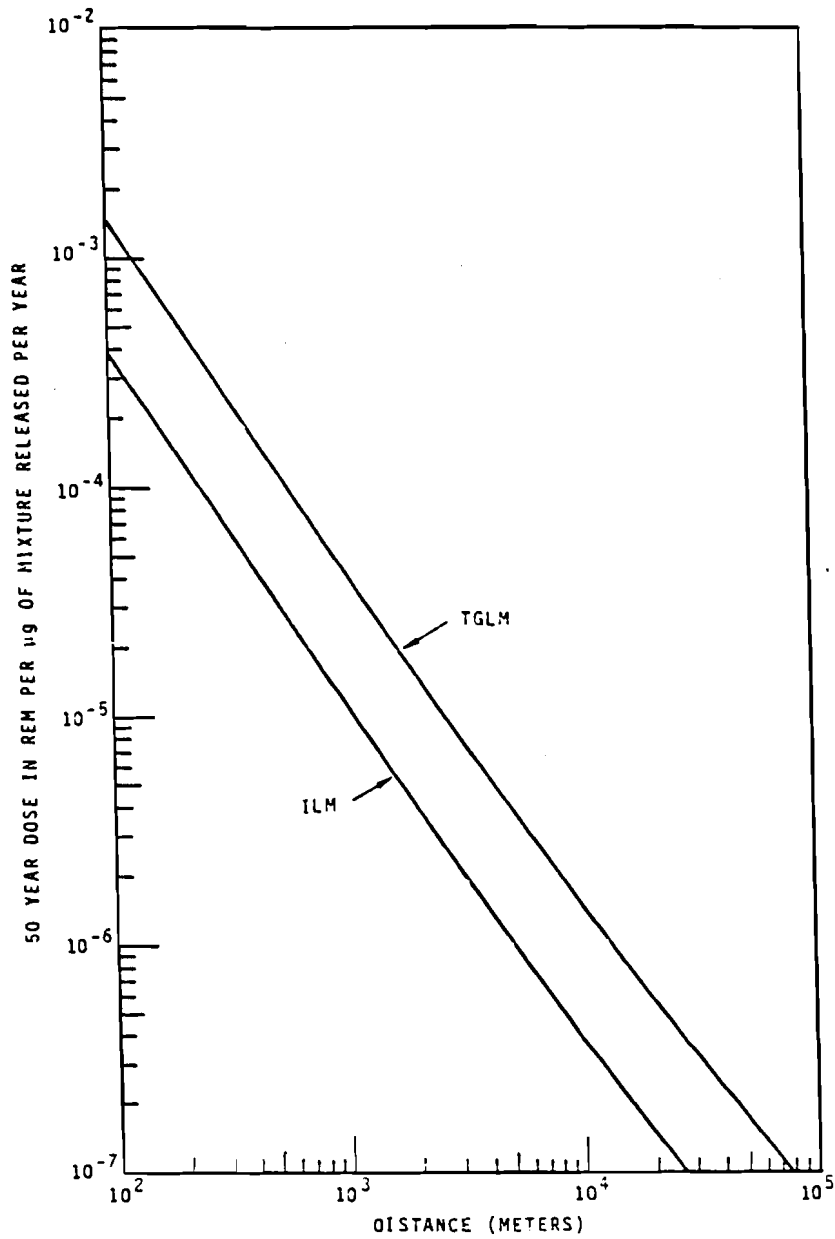
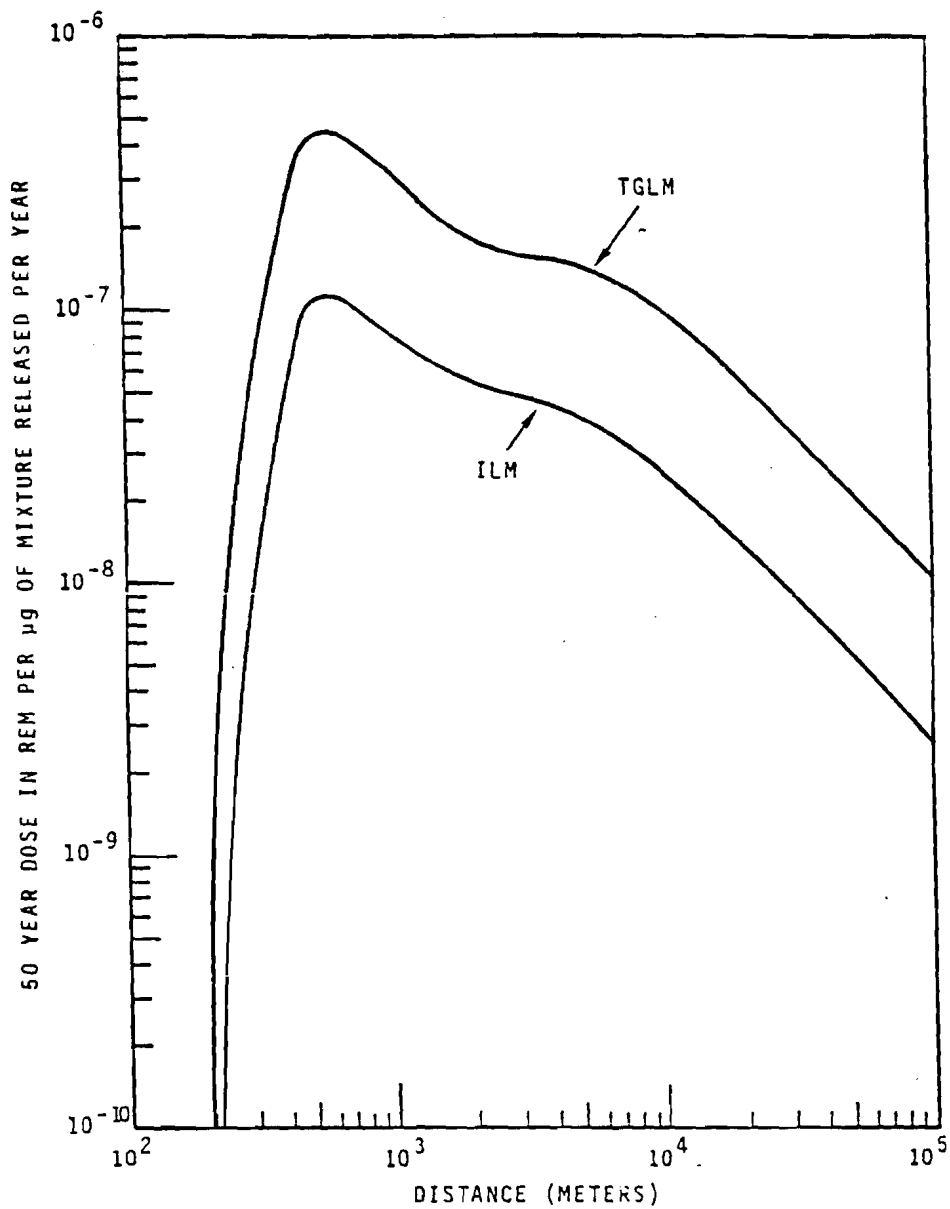


FIGURE 7. Comparison of the Dose to Lung from Inhalation of a Continuous Release of the Reference Mixture (Insoluble) Using the Two ICRP Recommended Lung Models, Ground Level Release, Maximum Sector Curves



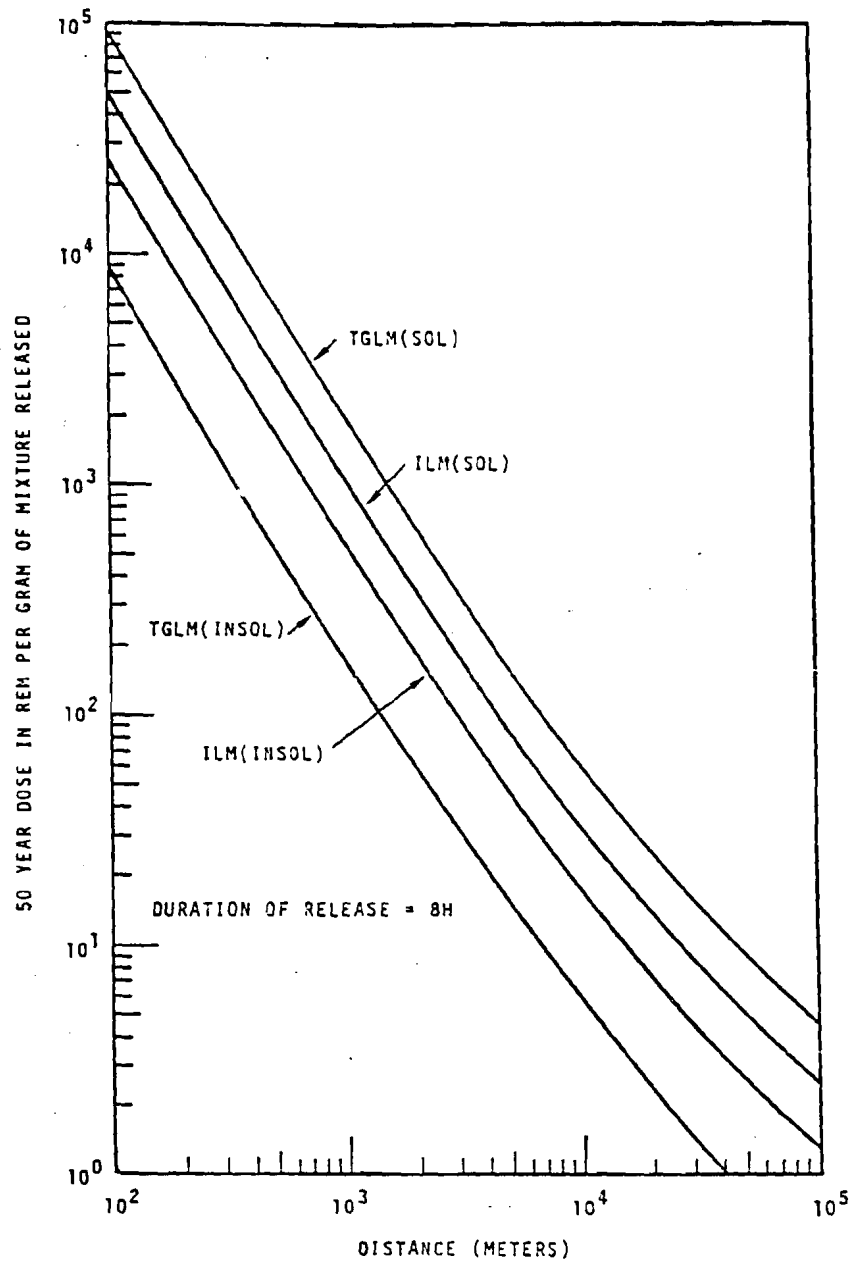
**FIGURE 8.** Comparison of the Dose to Lung from Inhalation of a Continuous Release of the Reference Mixture (Insoluble) Using the Two ICRP Recommended Lung Models, Release Height 100 m, Maximum Sector Curves

The 50-year dose to bone following an accidental atmospheric release was calculated as a function of distance for a one gram release of each radionuclide in the reference mixture. The calculation assumes inhalation exposure at ground level for release heights of ground level and 100 meters. The calculations were made for four release times. The 50 year bone dose commitment for acute inhalation following ground level releases are shown in Figures B-1 through B-5 (Appendix B) for each radionuclide. A similar curve is shown for the reference mixture in Figure 11. The 50 year bone dose commitments for the 100 meter releases of the individual radionuclides are shown in Figures B-6 through B-10 (Appendix B). Figure 12 shows the commitment for the reference mixture when released at a height of 100 meters.

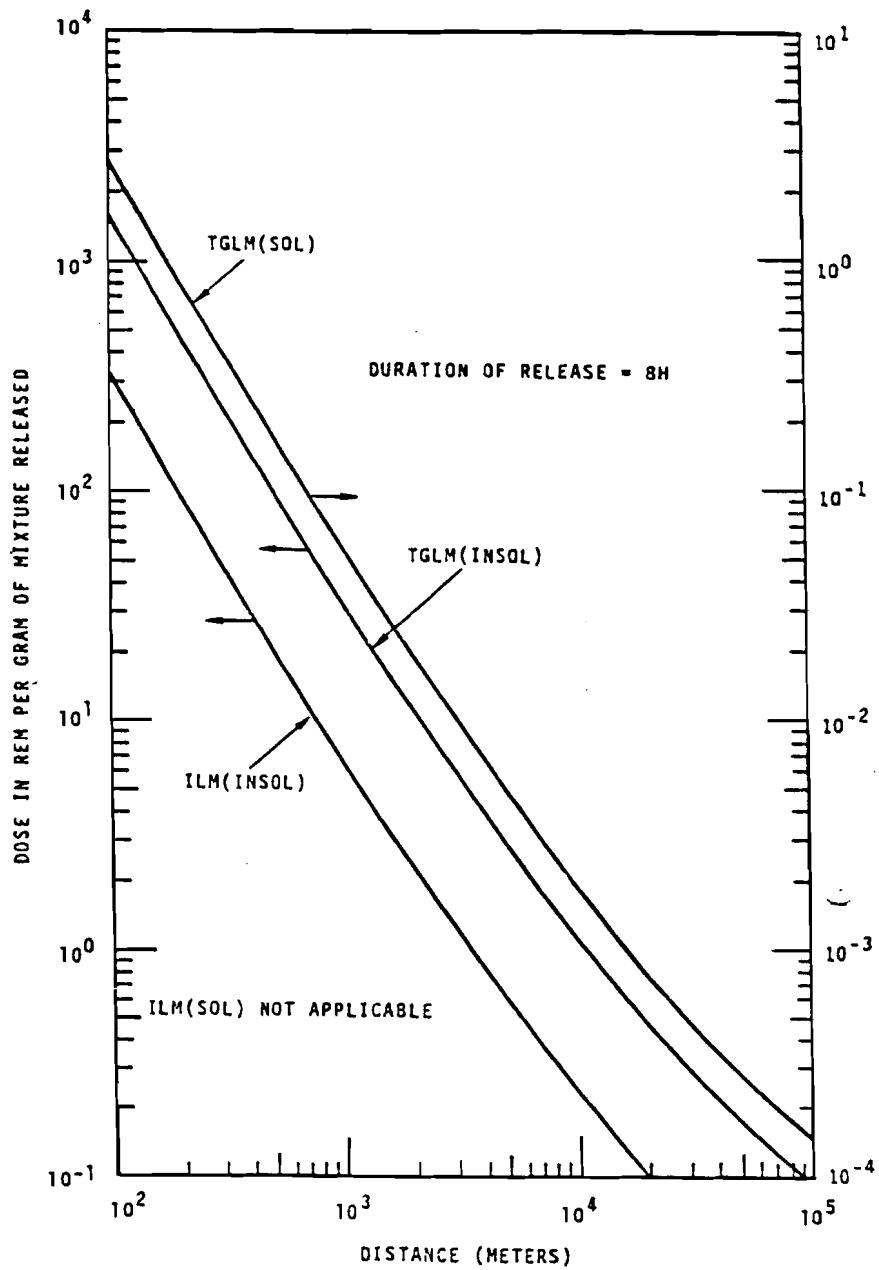
The accumulation of dose with time for each of the isotopes of the reference mixture following acute inhalation and subsequent deposition in the bone is illustrated on Figure 13. The figure demonstrates that the bulk of the dose from the mixture used in this study both initially and after 50 years, is due to  $^{238}\text{Pu}$  and  $^{241}\text{Pu}$ . The dose contribution from  $^{242}\text{Pu}$  is negligible.

Similar calculations were made for the dose to the lung. Based on the lung model recommended by ICRP in its Publication 2, the dose to the lung from insoluble plutonium is delivered in about a two year period following acute inhalation. Figures B-11 through B-15 (Appendix B) display the dose commitment per gram of each of the radionuclides released at ground level. The dose commitment for a ground level release of one gram of the reference mixture is shown in Figure 14. Similar dose curves for the lung are shown in Figures B-16 through B-20 (Appendix B) for 100 meter releases of each radionuclide. The dose commitment to the lung following a 100 meter release of one gram of the reference mixture is shown in Figure 15.





**FIGURE 9.** Comparison of the Dose to Bone from Acute Inhalations of the Reference Mixture Using the Two ICRP Recommended Lung Models, Ground Level Release



**FIGURE 10.** Comparison of the Dose to Lung from Acute Inhalation of the Reference Mixture Using the Two ICRP Recommended Lung Models, Ground Level Release

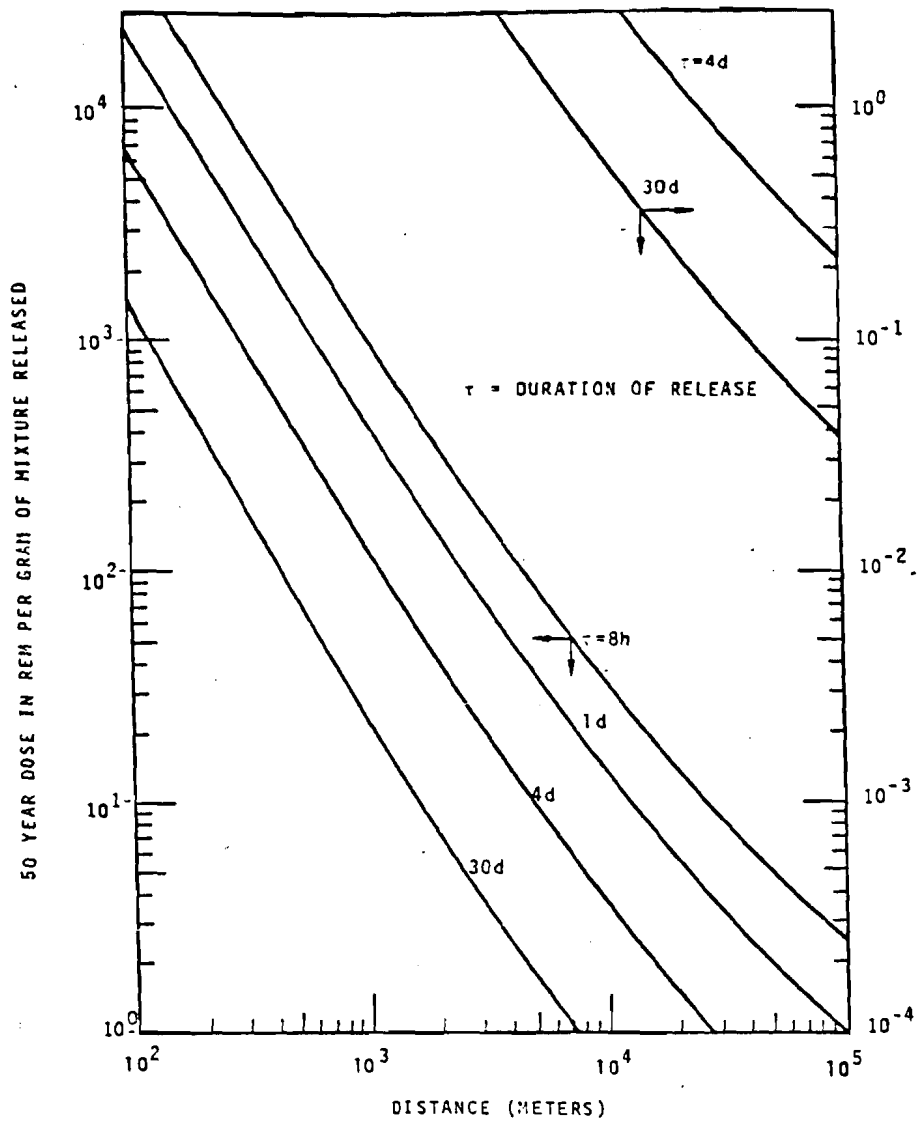
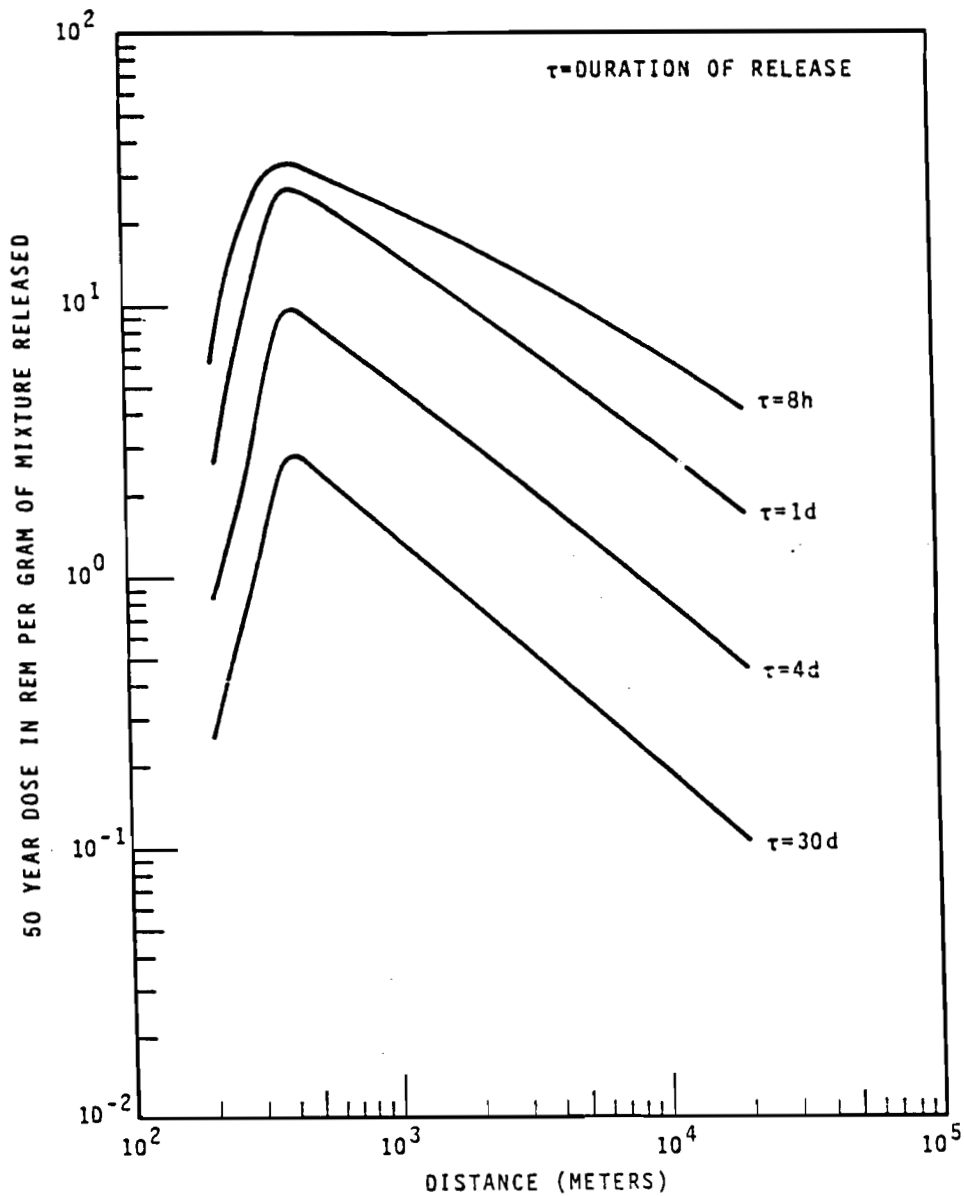


FIGURE 11. Dose to Bone from Acute Inhalation of the Reference Mixture of Plutonium and Americium, Ground Level Release



**FIGURE 12.** Dose to Bone from Acute Inhalation of the Reference Mixture of Plutonium and Americium, Release Height 100 m

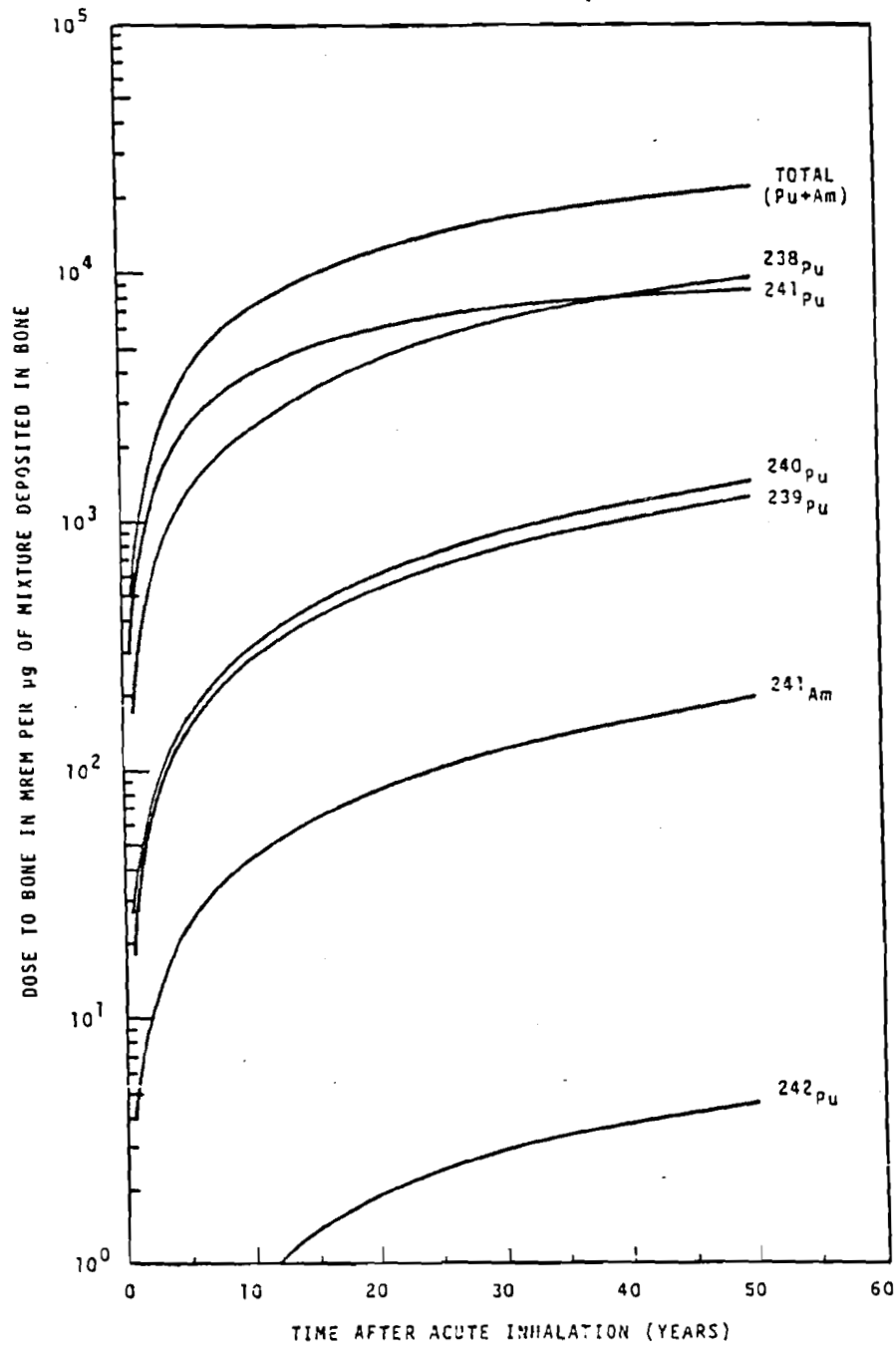


FIGURE 13. Dose to Bone as a Function of Time Following Acute Inhalation of the Reference Mixture of Plutonium and Americium (Soluble)

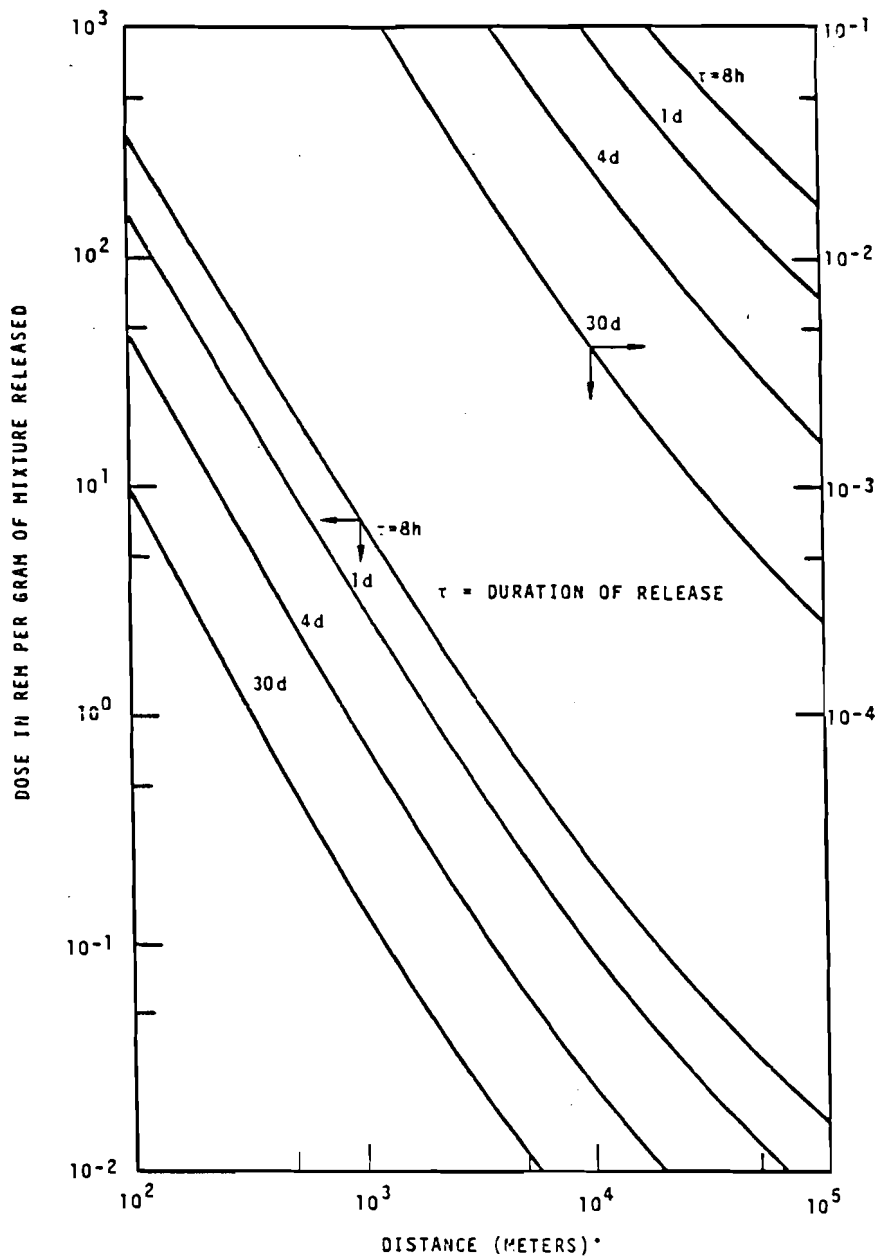
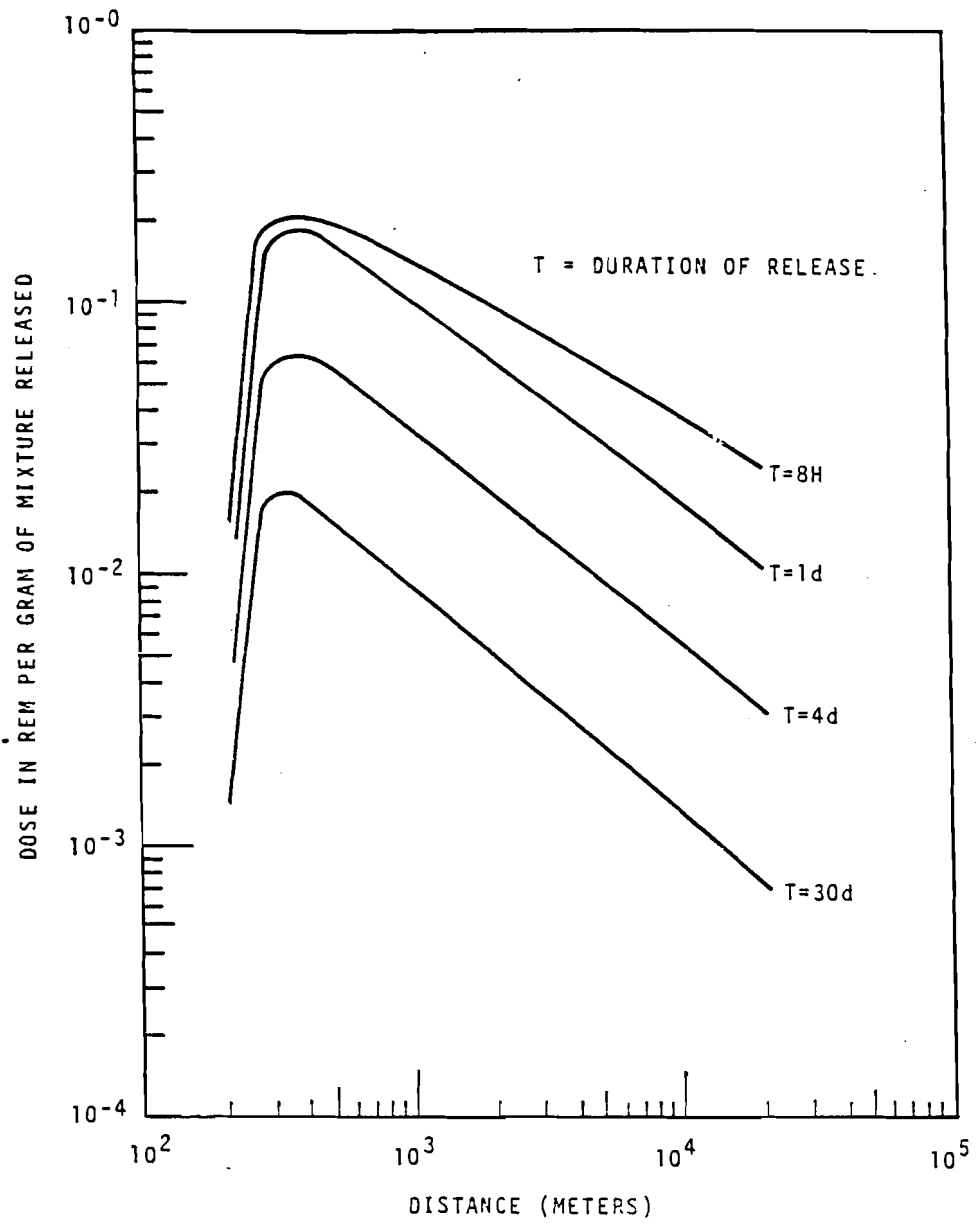


FIGURE 14. Dose to Lung from Acute Inhalation of the Reference Mixture of Plutonium and Americium, Ground Level Release



**FIGURE 15.** Dose to Lung from Acute Inhalation of the Reference Mixture of Plutonium and Americium, Release Height 100 m

## VII. SOURCE TERMS

In order to evaluate the risk to individuals in the environs of a fabrication plant, releases from normal operation and for various accidents have been estimated. The source term from normal operation results from having material in process. Theoretically small quantities of plutonium can, without equipment or operator error, find its way out of the confined systems into the plant environs. This type of release would be nearly continuous. Normally, the release would be marginally detectable in the environs. However, due to its high probability of occurrence, its consequences and subsequent environmental risk must be evaluated relative to less frequent source terms resulting from accidents.

Postulated accidents have been characterized according to consequences and probability or estimated frequency of occurrence. The accidents range from those with trivial off-site consequences to those which could cause significant off-site consequences. The assignment of occurrence rates for accidents resulting from equipment malfunction or operator error in fuel fabrication facilities is difficult due to lack of statistics. The limited statistics are a result of the relatively small number of these types of plants and their excellent safety records. However, accident data from non-nuclear facilities have been assembled in an effort to extrapolate to a plutonium plant. Due to the special emphasis on safety at fuel facilities, it is expected that these data will be conservative by at least an order of magnitude. In addition, general statements can be made about the occurrence frequency of accidents.

- Accidents resulting in trivial offsite consequences will occur more frequently than those resulting in significant offsite consequences.
- Accidents will occur more frequently in non-routine operation than routine operations.
- Accidents will occur less frequently where safety is achieved by engineered features rather than administrative controls.



## A. NORMAL OPERATION

The general process for plant operation along with the expected quantities and the physical and chemical forms of the fuel material in each step have been defined for the reference plant. In order to estimate the source term for normal plant operation, parameters related to the mobility, dispersion and deposition of plutonium compounds have been reviewed. Primary variables include particle size, physical and chemical form of uncontained material, air flow within the enclosure or glove box, and temperature of the environment. Other considerations relating primarily to operational practices include batch size, the form of containment within the enclosure, the uncontained time within the enclosure, and the degree of physical activity during the process step.

In estimating the source term from normal operation, a summation of the individual source terms from the various process steps may grossly overstate the real case. Due to the inherently conservative approach taken in characterizing the process parameters, the source term for each process step would be overestimated. Additional conservatism would be interjected since a simple summation of individual source terms would assume that all of the process steps occur simultaneously. Because of these considerations, the calculated annual source term for normal operation would have to be viewed as a maximum value rather than an expected average.

In order to interject realism into determining the source term for normal operation, measured releases from existing plutonium fuel fabrication plants have been reviewed. Assuming further development, standardization, and implementation of sound design criteria and improved operating practices, it is reasonable to assume that the releases from future fabrication will be less than from the plants of the past. This being the case, the source term used in the analysis for the normal operation case should be the one projected from industry experience rather than an individual process analysis.

Plutonium release rates from various fuel fabrication facilities making oxide fuels were examined. No consistency or trend was observed

between total plutonium released and fuel throughput. One of the major limitations of this approach was that the majority of the release results were "less than" the detection sensitivity of the methods employed at the facilities. It was considered that the release rate may not be highly sensitive to the plutonium throughput, but may only be dependent on the quantity of material maintained in process and the degree of activity in each step. This being the case, it was concluded that the emission rates from a 1 MT of fuel per day facility may be similar to the emission rates of much smaller facilities. In this light, a consistency in the release from present plutonium oxide and mixed oxide facilities was noted. This consistency may be attributed to the filter loading phenomenon previously discussed and/or to the inability of the present monitoring methods to discern a difference. The release rate for normal operation from the reference facility has been inferred to be less than 5  $\mu\text{g}$  of plutonium/year. This is consistent with previous work. (56)

*The source term is therefore: Quantity - 5  $\mu\text{g}/\text{yr.}$ , Particle Size - all in respirable range, Chemical Form - insoluble in body fluids,\* Height of Release - ground level, Duration of Release - continuous.*

## B. ACCIDENTS

The accidents have been characterized in general terms in order to develop an envelope of conditions that could occur in real plant situations. Little emphasis has been placed on the actual sequence of the events during the accident, since these are characteristics of specific plant and process design features. The consequence or resultant exposures are theoretical and should not necessarily be expected as a result of plant operation. This is true because of the statistical nature of these types of events and because of the conservatism applied to the calculations. In order to place the

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\* Particle size, however may be small enough that particles behave as though the material is soluble. Hence, in the bone dose calculation the material is assumed to be soluble and in the lung dose calculation the material is assumed to be insoluble.

risks of releases from these postulated accidents into perspective with the effects of normal operation, the calculated effects of the accidents should be scaled by their estimated frequency of occurrence. The product of consequence and frequency of occurrence provides an "environmental risk factor" for each operating function, whether it be normal operation or an accident situation.

In examining the various accidents deemed credible in the reference facility, it became obvious that the environmental effects would be inconsequential unless the final containment barrier was compromised. With the amount of fuel material limited in each process area, the typical operationally induced accidents resulted only in inconvenient in-plant conditions with negligible environmental effects. For this reason, the consequences of accidents coincident with the postulated partial failure of the final filtration system have been examined. Probabilities for these filter failures have been assigned to allow comparative analysis.

Accidents are unique occurrences. Their consequences depend for the most part, upon the sequence of events leading to and following the initial malfunction and to the amount and character of fuel material initially present. For this reason, not every conceivable accident can be discussed. Efforts were concentrated on analyzing the accidents and their parameters in the process areas having the potential for the greatest environmental consequences. Criteria for selection of these accidents were: amount of plutonium present, fraction of plutonium particles in the respirable range, difficulty generating plutonium aerosols, probability of occurrence and exposure by other means (e.g., criticality). Based on this set of criteria, it was obvious that the attention should be focused on four process areas or steps in the fabrication process. These areas are fuel storage, conversion, powder treatment, and scrap recovery. In the other areas, the fuel material is not easily dispersible, it is diluted by  $UO_2$  and/or contained, was present only in small quantities or the majority of the particles are not in the respirable range. A large quantity of dispersible plutonium is in the homogenization tank during blending of the nitrate solution. This solution, which is mixed in criticality safe, interconnect-

ing tanks, will typically be 1000 liters or greater. For the reference facility, this blending operation occurs in an area with a high degree of protection or a hardened facility like a vault.

Source terms were developed for the following types of accidents: criticality, explosion, localized fire, glovebox damage and major facility fire.

### 1. Criticality Accident

One of the major safety considerations in a fuel fabrication plant is criticality. The occurrence of a criticality event can have severe in-plant consequences if process shielding and emergency procedures are not adequate. To date four nuclear excursions have occurred in United States fuel fabrication and scrap recovery facilities.<sup>(57)</sup> None have occurred since 1964. All of these involved fissile solutions entering "unsafe" containers. Although there has never been any significant environmental consequences resulting from this type of accident, the considerations in evaluating the environmental effect of nuclear excursions are still examined.

Since an accident is a unique event, it is impossible to conceive *a priori* the exact mechanism(s) which could cause it or to relate the exact steps which it will follow. Knowing this in approaching a safety analysis, it is more important to examine the parameters in criticality events which have bearing on the final analysis. These parameters include:

- The total number of fissions - determines the amount of fission products generated.
- The "steady state" power level - defines the radiation exposure rate and determines the fission product and energy release rate. Probably a second order effect.
- The "peak power" pulse - defines the initial radiation exposure and the initial energy release which is primary to terminating the event and dispersing the radioactive material.

In examining these parameters, one must also consider the potential exposure pathways in the environment from a criticality event; prompt gamma

and neutron radiation exposure, external and internal exposure from released fission products, and internal exposure from plutonium released as a result of the criticality. As an example of a criticality event in the reference facility, we have assumed a criticality event in an "unsafe" tank of plutonium nitrate. Simplifying assumptions have been made in a conservative manner for the lack of a specific facility design or process procedure. In all cases, the assumptions maximize the environmental consequences and are important only in the mechanism(s) and the rate of terminating the criticality. For this study, it has been assumed that the event was caused by exceeding the critical volume by 10 liters during the filling of the tank and it resulted in  $10^{18}$  total fissions. Termination of the event was assumed to be by evaporation and expulsion of the 10 liter excess volume from the tank. An 8 inch wall of ordinary concrete was assumed to shield the plant environs from the direct radiation from the event. An empirical method for estimating credible number of fissions in a plutonium solution system is reported in Appendix E.

The dose to an individual from the prompt gamma and neutron radiation resulting from the criticality accident was examined as a function of distance from the facility. Table 13 summarizes these calculations. Even though experimental evidence<sup>(58)</sup> does exist which indicates that the neutron flux decrease can be approximated by the inverse distance to the fourth power for distances greater than 100 meters, inverse squared formulation was used. A quality factor of 10 was used for correcting the absorbed neutron dose to rem.<sup>(59, 60, 61)</sup> The doses in the table have been corrected for the assumed shielding. Values for fractional transmission through the shielding wall of 0.17 and 0.22 were used for the neutron and gamma radiation, respectively.<sup>(61)</sup>

As a result of the excursion, fission products are formed and a large amount of energy released. About 85% of the approximate 200 MeV released per fission is available for heating the solution. The energy input to the solution provides the power limiting and ultimate terminating mechanism for the accident. For this accident greater than  $6 \times 10^6$  calories of heat energy is initially available to the system. The solution would, more than likely, critically pulse at an "equilibrium" fission rate, until the solution is below the critical volume.

TABLE 13. Prompt Gamma and Neutron Dose to an Individual as a Function of Distance from a Criticality ( $10^{20}$  Fissions)

<u>Distance, m</u>	<u>Gamma Dose<sup>(a)</sup> Rem</u>	<u>Neutron Dose Rem</u>	<u>Total Dose Rem</u>
$10^2$	$4.6 \times 10^{-1}$	5.5	6
$10^3$	$4.6 \times 10^{-3}$	$5.5 \times 10^{-2}$	$6 \times 10^{-2}$
$10^4$	$4.6 \times 10^{-5}$	$5.5 \times 10^{-4}$	$6 \times 10^{-4}$

a. A total fission gamma energy of 5 MeV per fission.

Vaporization of some of the solution during the initial power pulse could cause a rapid volume expansion or pressurization of sufficient magnitude to rupture the glovebox. This is assumed to occur.

The excursion is terminated following the evaporation of the 10 liters of excess solution. Airborne release fractions for plutonium in nitrate solution vary widely depending upon temperature and activity of the solution, the plutonium concentration and air flow over the solution. The heat energy from the excursion would raise a 1000 liter solution about 15 °F if the heat were uniformly distributed. However, it was conservatively assumed that all of the heat was used to create localized boiling, vaporization of the excess volume, and surface film breakup. A release fraction of 0.2% was conservatively used for this accident. Work by Mishima et al.<sup>(62)</sup> indicates that as much as 0.18% of the plutonium in a dilute solution was made airborne during evaporation of approximately 90% of the solution in a deep form beaker of the solution at a rolling boil. The release fractions would be in the range of  $10^{-2}$  to  $10^{-4}$ % for less extreme conditions.

Assuming a solution containing 150 g of Pu per liter, a maximum of 3 g of plutonium could become airborne. The hood filter was assumed to have plugged releasing everything into the room and out the final filter system.

*The source term for this accident would be: Quantity -  $3 \times 10^{-5}$  g for two HEPA filters and  $3 \times 10^{-3}$  g for one HEPA filter, Particle Size -*

*all in respirable range, Chemical Form - soluble, Height of Release - ground level, Duration of Release - less than two hours.*

The fission products generated as a result of the criticality event also offer a dose potential and thus merit a consideration. For this analysis, simplifying assumptions are made to permit a view of the maximum condition possible. These assumptions are:

- All of the noble gases generated are released from the building. One fourth of the iodine is released. The material is released over a 10 minute interval.
- The radioactive material was released at ground level and dispersion was during a Pasquill type F meteorological inversion.

The inventory of fission products generated during the criticality were calculated using the computer code RIBD<sup>(63)</sup> and the activity at selected times after termination of the event is tabulated in Table 14. Cumulative yields for thermal neutrons were used in determining the activity of the fission products. The volatile solids and remaining fission products are not released from the building to any significant extent and therefore were not considered in the dose calculations.

The total body dose from the passage of the cloud following the criticality accident was calculated as a function of distance from the facility using the computer code RACER.<sup>(64)</sup> Radioactive decay is considered during the dispersion of the radioactive material. The dose to a 20 g thyroid from radioactive iodine inhaled during the passing of the cloud was calculated using a code INDOSE.<sup>(65)</sup> The results of these calculations are summarized in Table 15.

## 2. Explosions

Several types of explosions are conceivable in this type of facility. The most commonly examined are explosions in sintering furnaces, autoclaves, and plutonium solution tanks. These types of events are highly localized and will result in damage only to small areas. No explosion can be conceived which would affect the entire facility and result in major damage to the building.

TABLE 14. Fission Product Inventory From Criticality Accident

$10^{18}$  Total Fissions (32 Mw-sec) Quantity in Curies

Nuclide Classification	Duration of Event	Decay Time						
		Shutdown	10 min	60 min	2 hr	5 hr	10 hr	1 day
Noble Gases	0.5 sec	2.5E6 <sup>(b)</sup>	2.4E3	2.1E2	1.1E2	5.9E1	3.2E1	1.4E1
	5 min	3.8E4	1.8E3	2.0E2	1.1E2	5.9E1	3.2E1	1.4E1
	1 hour	4.0E3	5.3E2	1.5E2	9.9E1	5.4E1	3.1E1	1.4E1
Halogens	0.5 sec	6.0E5	4.7E2	2.7E2	2.1E2	7.4E1	3.4E1	1.5E1
	5 min	2.1E4	3.7E2	2.7E2	2.1E2	7.4E1	3.4E1	1.5E1
	1 hour	2.1E3	2.8E2	2.5E2	1.8E2	6.5E1	3.2E1	1.4E1
Volatile Solids	0.5 sec	9.0E5	2.3E3	6.7E2	2.4E2	1.8E1	5.8E0	3.3E0
	5 min	3.4E4	2.0E3	6.4E2	2.3E2	1.8E1	5.8E0	3.3E0
	1 hour	4.1E3	1.1E3	4.2E2	1.5E2	1.4E1	5.6E0	3.3E0
ARFP <sup>(a)</sup>	0.5 sec	1.5E6	1.5E4	2.0E3	7.5E2	3.3E2	1.8E2	6.4E1
	5 min	1.1E5	1.2E4	1.9E3	7.3E2	3.3E2	1.8E2	6.4E1
	1 hour	1.6E4	4.7E3	1.2E3	6.0E2	3.1E2	1.7E2	6.2E1

a. ARFP = All remaining fission products

b. 2.5E6 means  $2.5 \times 10^6$



TABLE 15. Total Body and Thyroid Doses to an Individual Located Downwind from a Criticality ( $10^{18}$  Fissions)

<u>Downwind Distance, m</u>	<u>Dose, rem</u>	
	<u>Total Body</u>	<u>Thyroid</u>
$10^2$	1.4	$1.1 \times 10^1$
$10^3$	$3.1 \times 10^{-2}$	$2.0 \times 10^{-1}$
$10^4$	$2.2 \times 10^{-4}$	$4.3 \times 10^{-3}$

Hydrogen explosions in pellet sintering furnaces have a limited amount of energy. The damage that could result from this type of event would likewise result in limited consequences. The explosion would probably be directed out the ends of the furnace. The glove box could be breached and pellets and possibly a small amount of mixed oxide fines could be spread around the room. It would be virtually impossible to produce significant quantities of plutonium particles in the respirable range from damage to the pellets or dispersion of the fines. This type of accident would result in an in-plant contamination spread and is a negligible source term to the environs.

The rupture of an autoclave wall during operation, could result in the high speed projection of fuel pins at the building walls or ceiling. Building design against missile precludes penetration of the structure from this type of accident. Significant source terms of respirable aerosols of plutonium are not credible either within or outside the building from this type of event.

A chemical explosion involving a plutonium solution could result in the production of a substantial quantity of airborne plutonium particles. If the vessel is open, some liquid would be pushed out of the vessel and some plutonium particles in the respirable range would be generated. If sufficient force is not available to cause extensive film break-up, it seems probable that most of the liquid would impact on and adhere to adjacent surfaces. If an explosion is of sufficient magnitude to rupture a heavy walled, closed vessel, a considerable number of fine particles could be generated by the liquid passing through the jagged opening.

In any case, the number of particles that persist in the air is limited. Swain and Haberman reviewed data from non-nuclear sources and calculated that 33 mg plutonium per cubic meter was the maximum airborne concentration a few minutes after an accident.<sup>(65)</sup> Castleman, Horn and Lindauer using an exploding wire technique to generate very fine particles found concentrations as high as 71 mg plutonium per cubic meter a few minutes after generation.<sup>(67)</sup> By calculation, a total of  $10^6$  plutonium particles with an aerodynamic equivalent diameter (AED) of 10  $\mu\text{m}$  per cubic centimeter would result in an airborne concentration of 2 g of plutonium per cubic meter. Considerable care and effort are required to generate monodispersed particles. Generally particles generated by a single mechanism are log-normally distributed. Thus, the anticipated weight concentration of 10  $\mu\text{m}$  AED particles would be much lower than that calculated for monodispersed particles. An upper value of 100 mg Pu per cubic meter for particles in the respirable range appears reasonable. Because of this upper mass limit for airborne plutonium particles, the consequences for explosion accidents appear to be limited by the material that can be maintained in the air and not by the total volume or mass initially involved. An explosion will, of course, splatter the solution on the wall and floor which may later become airborne as it dries. This is expected to be a small contribution to the overall source term because of the small release fraction from this condition without considering that corrective action will be implemented to contain the contamination spread.

The room in which the explosion occurs is assumed to have a volume of  $10^4$  cubic meters. At a maximum airborne concentration of 100 mg plutonium per cubic meter of air, the total amount of airborne plutonium which could be sustained in the room following the explosion would be about 1000 g.

*The source term from this type of accident is: Quantity -  $10^{-2}$  g for two HEPA filters and 1 g for one HEPA filter, Particle Size - all in respirable range, Chemical Form - soluble, Height of Release - ground level, Duration of Release - less than 2 hours (an exponential purge rate).*

### 3. Local Fire

The occurrence of a fire near the nitrate blending tanks represents one of the greatest potentials for release of plutonium. Airborne releases from fire around a blending tank can be envisioned in several ways depending on whether the tanks are vented or closed. If the blending vessel is a vented container, the fire could heat the liquid and cause it to boil. Boiling a dilute plutonium nitrate solution until a 90% volume reduction is attained could result in an airborne release no greater than 0.2%.<sup>(62)</sup> Fractional releases from concentrated solutions could be higher. After a substantial quantity of liquid is lost, the liquid thickens and vapor trapped under the surface tends to throw solids into the air. The salt content would be high and the tendency would be for the majority of material to form a coarse cake which is sintered to the surface of the vessel. Less than 0.5% of a uranium nitrate solution (applicable to plutonium) as a thin film on a stainless steel surface involved in a gasoline fire was made airborne by air at a nominal velocity of 2.5 mph passing over the surface.<sup>(68)</sup> Approximately 80% of the uranium aerosol was in the respirable size range. A smaller fraction is anticipated under the accident conditions due to the lack of airflow and depth of the vessel.

If the blending tank is a closed vessel, fire in the area around the vessel would result in pressurization of the vessel. Pressurization of the vessel could result in an airborne release in several ways. The pressure within the vessel could cause the nitrate solution to be jetted from various openings. If the opening is large, some small particles could be formed by film break-up along the edges of the stream with subsequent evaporation of the liquid to form particles. In this case, the fractional release is small. Small particles also could be formed by forcing the liquid through small openings, however, high pressures are required. A  $10 \mu\text{m}$  AED  $\text{PuO}_2$  particle is formed by a  $41 \mu\text{m}$  drop of nitrate solution of 150 g plutonium/liter concentration. Each particle contains approximately  $2 \times 10^{-10}$  g Pu. To generate an airborne concentration of  $20 \text{ mg/m}^3$ ,  $10^8$  particles/ $\text{m}^3$  of air of this size would be necessary. The number of holes required to produce this number of particles in a short period of time is large and it is not likely that such a condition could exist.

If the pressure is high enough, the vessel could rupture. If the rupture occurred above the liquid level some plutonium would become airborne but not a significant quantity. If the rupture occurs near the bottom of the tank and is large, a large stream of solution would pour onto the floor and extinguish part or all of the fire. If the fire is extinguished the fractional airborne release is low -- less than 0.003%.<sup>(62)</sup> If the fire continues and dries the nitrate solution, the airborne release could be as high as 0.5%.

For the source term calculation, it was assumed that there were 150 kg of plutonium as a nitrate in 1000 liters blending solution. If the blending tank is vented, a fire that boils the vessel dry could release as much as 0.7% of the plutonium present -  $1.5 \times 10^5 \text{ g} \times 0.007 = 1050 \text{ g}$  plutonium into the available air space. If the vessel is closed and is ruptured by the internal pressure, the nitrate would spill on the floor and be dried by the fire. Fractional airborne release could be as high as 0.5% -  $1.5 \times 10^5 \text{ g} \times 0.005 = 750 \text{ g}$  plutonium into the available air space. An internal detonation could generate a large quantity of fine droplets into the available air space if it were of sufficient magnitude to rupture the vessel. For a maximum release, a conservative estimate would assume an instantaneous uniform air concentration. The airborne concentration would be approximately  $100 \text{ mg plutonium/m}^3$  of air for a few minutes.

Using 1050 g of airborne plutonium as the release to the building, source terms were calculated for the fire accident.

*The source terms would be: Quantity -  $1.1 \times 10^{-2} \text{ g}$  for two HEPA filters and 1.1 g for one HEPA filter, Particle Size - 80% in respirable range, Chemical Form - insoluble, Height of Release - ground level, Duration of Release - less than two hours.*

#### 4. Glovebox Damage

An accident involving the loss of primary barrier has been examined for the powder treatment areas. The sized  $\text{PuO}_2$  powder prior to the  $\text{UO}_2$  blending represents one of the greatest sources of respirable plutonium

in a fabrication facility. For this reason, an accident has been explored to focus attention on some of the characteristics of this material.

It was assumed that mechanical damage occurs in the area of the PuO<sub>2</sub> powder line rupturing the glovebox and breaking an adjacent compressed air line. The line sags and directs a jet of air into an open container holding the PuO<sub>2</sub> powder. It is also assumed that the material is awaiting blending and involves the entire batch limit of 11.3 kg of Pu. Under such conditions, greater than 50% of the powder could be made airborne. Mishima, et al, found that 60 to 70% of relatively coarse oxalate powder (Median Mass Diameter of 50 m) could be made airborne by air-drawn tangentially across the powder.<sup>(69)</sup> Royster and Fish found approximately 80% of 5 μm particles will be made airborne from a stainless steel surface by air at a velocity of greater than 60 m per sec impinging on the surface at various angles.<sup>(70)</sup> Most of the plutonium powder for the operation is assumed to be in the respirable range (a PuO<sub>2</sub> sphere less than 3.3 μm). Deagglomerating a powder composed of such fine particles is not a trivial task. But making a "conservative" assumption that the material airborne is deagglomerated and uniformly distributed in a room of 10<sup>4</sup> cubic meters, the maximum amount of plutonium which could be made instantaneously airborne in the room would be 1000 g. Continuously applying the jet of air on the powder over an extended period would, of course, put more plutonium in the air but it is expected that the situation would be corrected immediately. The glovebox filter is assumed to plug causing all flow of material to be out of the glovebox, thereby bypassing the hood filter system.

*The source term for this accident situation would be: Quantity - 10<sup>-2</sup> g for two HEPA filters and 1 g for one HEPA filter, Particle Size - all in respirable range, Chemical Form - insoluble, Height of Release - ground level, Duration of Release - less than two hours (an exponential purge rate).*

##### 5. Major Facility Fire

It would require a major disaster to breach facility confinement and release unfiltered plutonium to the environs. There are only a few accidents

in this "plant disaster" class that can theoretically produce damage of sufficient magnitude to compromise the final confinement barriers. Specific plants will, as a minimum, be structurally designed and built to satisfy criteria relative to earthquakes and tornadoes. However, finite possibilities exist that the facility could be stressed by forces beyond those used for design. Major facility fires also seem incredible in buildings where combustibles are limited, but experience indicates they can occur. In summary, plant disaster accidents that can cause major facility damage are not "incredible" but highly improbable.

The bulk of plutonium in the facility will be in storage in a hardened area like a vault. For the reference facility, only a few hundred kg of plutonium will be in-process, with about one-half in dispersible form.

A plant fire that could cause catastrophic breaching of the final barrier could not be conceived. With the expected concrete construction of a facility of this type, the low fire loading, and the expected airtight nature, total burning is incredible. For analysis, the final barrier was assumed to remain intact after a facility fire involving the gloveboxes and other combustibles in process and the deluge system was assumed to remain operable. It was estimated that less than 1% of the in-process dispersible inventory could become airborne within the facility.

*Assuming two stages of filtration the source term would be: Quantity- $10^{-2}$  g, Particle Size - all in respirable range, Chemical Form - insoluble, Height of Release - elevated, Duration of Release - extended (greater than two hours and less than eight hours).*

#### 6. Other Accident Considerations

From the previous accident discussions, it should be obvious that there are many other accidents that can be hypothesized for a fuel fabrication facility. However, for lack of specific design details in the reference facility, the accidents focused on the process areas and operations offering the greatest potential environmental consequences. Less dramatic events such as small liquid or powder spills and ruptured drybox

gloves occur more frequently than the accident cases examined, but the environmental consequences would be insignificant and the considerations would be far less instructive. For this reason, additional information relating to the generation of plutonium aerosols for other accident conditions has been included in Table 16 and in Appendix F. The release values in the table were selected as maximum values from the data available, with rounding toward the highest values found. Judicious application of release fractions should include a careful review of the conditions under which the release values were obtained. Postulated accidents may not conform well to the experimental conditions; hence, the analysis must recognize discrepancies between postulated and experimental circumstances. Abstracts of most of the technical reports relating to plutonium releases are presented in Appendix F. The reader charged with the responsibility of assessing accident consequences should be thoroughly familiar with the details of the experimental measurements described in the original documents.

TABLE 16. Summary of Experimentally Determined and Estimated Airborne Release Fractions of Plutonium Under Various Thermal and Aerodynamic Stresses

	<u>Stress Imposed</u>	<u>% of Source Initially Airborne</u>
•	<u>Fire</u>	
	Metal - Ignited airflow less than 100 cm/sec (2.2 mph)	0.01 <sup>(a)</sup>
	Partial disruption of molten metal, airflow greater than 100 cm/sec	1.0 <sup>(a)</sup>
	Dioxide Powder - Airflow less than 100 cm/sec	0.5
	Airflow 100 cm/sec	1.5
	Airflow greater than 100 cm/sec	15
	Involved with flammable material natural convection	0.05
	Involved with flammable material forced draft	40
	Oxalate Powder - Airflow less than 100 cm/sec	1.0
	Fluoride Powder - Airflow less than 100 cm/sec	0.1
	Nitrate Solution - Airflow less than 100 cm/sec	0.5
	Airflow greater than 100 cm/sec	1.0
	Gasoline fire, stainless steel surface, 20 mph	10
	Involved with flammables, natural convection	0.2
	Involved with flammables, forced draft	10

• Explosion

Any explosive mechanism of sufficient magnitude to completely destroy the integrity of the containment is assumed to make airborne all the source material directly involved or fill the available air space with an aerosol which has a mass concentration of 100 mg Pu/cm<sup>3</sup> after 10 minutes.

• Other

If air at a velocity of greater than 10 mph is directed upon or through a finely divided powder (particles less than 50 microns AED), 100% of the powder is assumed airborne.

a. Percent of source airborne associated with particles less than 10 microns Aerodynamic Equivalent Diameter (AED).



## VIII. PROBABILITY OF RELEASES

There are two kinds of releases, chronic and accidental. The chronic releases occur as a result of normal operation. These releases are kept "as low as practicable" by using the highest design standards and then performing a periodic inspection and maintenance program. The accidental releases are normally produced by a sequence of events which ultimately result in a release. Delineation of events before, during and after an accidental release inevitably disclose that prior to the system breakdown, with its resultant release, there occurred a chain of events in which a series or combination of system component failures or deficiencies led to a release. These accidental releases are usually not the result of a single event. All too frequently, man-machine interactions are important contributors to the accident chain. The causal relationships can be obtained from operating data if sufficient facts about the accident situations are reassembled. A fault tree analysis technique is the best safety analysis tool for showing these causal relationships.

The following subsections describe the failure modes for both the containment building and the internal equipment. The failure probabilities for the containment structure will be based on the potential for major natural disasters such as tornadoes, earthquakes, fires and meteor strikes. The equipment failure data will be based on the personal experience of the authors of this report, failure data for similar equipment in related industries, and also from data tabulated by the Nuclear Safety Information Center.

### A. PROBABILITY OF MAJOR EVENTS WHICH COULD BREACH CONTAINMENT

The accidents which could result in a breach of the containment are shown in Table 17. The basis for each of the numbers will be given in separate paragraphs which follow.

#### 1. Tornado

The probability of a tornado striking a facility has been considered in nuclear facility safety analysis reports. Articles by Doan<sup>(71)</sup> and

TABLE 17. Frequencies of Major Accidents

<u>Event</u>	<u>Probable Frequency Of Occurrence Per Plant - Year</u>	<u>Range</u>
Tornado	$6 \times 10^{-4}$	$4 \times 10^{-3} - 6 \times 10^{-6}$
Earthquake		
Intensity IX	$2 \times 10^{-5}$	$10^{-2} - 10^{-8}$
Airplane Impact	$10^{-5}$	$10^{-4} - 10^{-6}$
Meteorites	$10^{-10}$	$10^{-9} - 10^{-11}$
Flood	$10^{-4}$	$10^{-2} - 10^{-6}$
Fire	$2 \times 10^{-4}$	$4 \times 10^{-4} - 4 \times 10^{-5}$

Dunlap and Wirdner<sup>(72)</sup> summarize the analyses carried out for these facilities. Doan states that the worst site, in Oklahoma, has a probability of a strike in any year of  $3.62 \times 10^{-3}$ . The probability of a tornado in the western United States, based on a study by Smith and Mirabella<sup>(73)</sup> is  $1.3 \times 10^{-6}$ /year. All locations in the U.S. fall within that range. The average for the contiguous 48 states is  $6.0 \times 10^{-4}$  strikes/year. Doan states that "severe tornadoes of design proportions have a probability of occurrence about two decades below the above figures."

Dunlap and Weidner discuss tornado driven missiles considered in nuclear power plant design for the midwest. They are:

- (1) A 4 in. x 12 in. x 12 ft long wood plank weighing 108 lb traveling on end at 300 mph and striking any place on the sides of the reactor building.
- (2) A 3 in. diameter schedule 40 pipe 10 ft long traveling on end at 100 mph striking any place over the full height of the structure.
- (3) A passenger car weighing 4000 lb traveling on end at 50 mph with a contact area of  $20 \text{ ft}^2$  and at a height not more than 25 ft above ground level.

In the western states, the missile velocities are ratioed down by the factor of 200/300 since the highest expected velocity is 200 mph for that region.

In addition to the missiles, the passing of a tornado gives rise to an atmospheric pressure fluctuation which must be considered in any design. The lower pressure is expected to last for 3 seconds and then recover to normal atmospheric pressure in the next 3 seconds. For the midwest a pressure transient of 3 psi in 3 seconds is consistent with the 300 mph design basis tornado. In the western U.S. the maximum pressure drop is expected to be 1.5 psi and the rate of fluctuation is expected to be below 0.4 psi/second. <sup>(73)</sup> Thus it would take about 4 seconds for the decrease of 1.5 to occur in the west.

In the analysis it is important to consider the cumulative effect of the pressure fluctuations, the missiles and the vortex on the plant structures. If the internal building pressure adjusts to the pressure fluctuation, then the effect of the pressure fluctuation on equipment such as gloveboxes should be considered.

## 2. Aircraft Impact

Another missile source is an aircraft impact into the side of the facility. Chelapati, Kennedy and Wall <sup>(74)</sup> analyzed the aircraft hazard for nuclear power plants. They determined that the probability of a strike per year varied from  $1.4 \times 10^{-5}$  for distances greater than 5 miles from an airport to  $3.4 \times 10^{-5}$  for distances less than 5 miles from the airport. No variation in the number of flights over a given region were considered in their analyses. Thus the hazard could vary by at least a factor of 100 about this mean.

Chelapati et al., looked at the thickness of concrete required to prevent penetration of the barrier. If the barrier was reinforced concrete 1 ft thick, approximately 1/3 of all crashes will produce missiles which can penetrate the barrier. Most airplane crashes result in fire of varying duration. The addition of fire can add to the severity of the event.

### 3. Fire

Fire data were derived from statistics of the chemical industry. Although the plutonium fuel fabrication industry has many operations which are more nearly light manufacturing in nature, the scrap recovery, co-precipitation, and powder blending operations have a great deal of similarity to common chemical processes.

Hence, the use of fire data from the chemical industry can be justified even though it may be conservative. Major fires in the chemical industry for the years 1966-70 were 25.<sup>(75)</sup> It was assumed that the population from which this statistic was derived was the entire number of plants listed under Chemical Industry (SIC #28) which is given in the 1967 industrial census as 11,799.<sup>(76)</sup> This results in a probability of  $4 \times 10^{-4}$  major fires per plant per year. Recognizing the difference in industry characteristics, a lower range value of  $2 \times 10^{-4}$  was arbitrarily assigned.

### 4. Earthquake

The risk from seismic events is very dependent on the geographic location of a particular facility. The region west of the Rockies is considered to have much greater seismicity than the rest of the country. California in particular has very high seismicity because of the numerous active faults in the state. Algermissen<sup>(77)</sup> has estimated the seismic risk in the various regions of the country. Gutenberg and Richter<sup>(78)</sup> have summarized the seismicity of the entire earth for the 1904-1952 time period. Of particular interest to this study are reports by G. A. Bollinger<sup>(79)</sup> and a book by Wiegel.<sup>(80)</sup> The Bollinger article discusses the historical and recent seismic activity in South Carolina. In this article Bollinger states "the great Charleston, South Carolina earthquake of August 31, 1886 provides an example of a major United State earthquake outside the Pacific coast region. No prior severe shocks were known to have occurred in this region since its settlement by the English in 1670. Because of this long aseismic history, the 1886 event is often cited as an example that no region is completely safe from earthquake hazard." The estimated magnitude of this earthquake was 7.7 on the Richter scale.

On the Modified Mercalli scale this earthquake had a maximum intensity of X and a region of about 3,000 square miles experienced damage in excess of IX on the Modified Mercalli scale. In this 3,000 square miles, buildings were shifted off foundations, cracked, thrown out of plumb, ground was cracked and underground pipes broken. For purposes of this report the risk calculation will use an intensity of IX to calculate the probability of an earthquake striking a fabrication plant. The use of this intensity is not meant to indicate that the earthquake will result in a release, it was used as a point of reference. More severe earthquakes will have a lower probability of occurrence and thus less seismic risk.

Bollinger shows that the probability of an earthquake striking a region of 3,000 square miles along the east coast is about 0.005/year. Assuming the east coast consists of  $10^6$  square miles, then the risk of an earthquake at any point on the east coast is approximately  $\frac{0.005 \times 3000}{10^6}$ . Thus the probability of an earthquake at some generic eastern site would be approximately  $2 \times 10^{-5}$ /year.

In California, the probability of a Richter magnitude 7.5 earthquake is 0.076/year.<sup>(80)</sup> This would result in an intensity of greater than IX on the Modified Mercalli scale over about 2,000 square miles. The probability of an earthquake with a magnitude greater than 8.0 is 0.021 and 6,000 square miles can be expected to experience heavy damage. The probability of a magnitude 8.5 earthquake is estimated to be 0.006/year. Approximately 14,000 square miles can be expected to experience heavy damage from this earthquake. Thus the probability of an earthquake striking a random point in California (150,000 square miles) is:

$$p = \frac{0.006 \times 14,000 + (0.015)(6,000) + 0.055(2,000)}{150,000}$$

$$= 0.002/\text{year}.$$

It should be recognized that some areas, particularly along faults, experience a higher probability than being involved in a severe earthquake once every 500 years.

## 5. Meteorites

As part of the space effort, significant research has been done on the probability of meteorites impacting the earth. Gera and Jacobs<sup>(81)</sup> summarized current knowledge on the subject and concluded that the probability of a  $2 \times 10^7$  kg meteorite impacting an area is  $10^{-2}/\text{km}^2\text{-yr}$ . The frequency of falls is inversely proportional to the weight of meteors, and for a 2,000 kg meteor, the probability is  $10^{-8}/\text{km}^2\text{-yr}$ . This meteorite with an impact velocity of approximately 4,500 ft/sec with a resulting crater of over  $3 \text{ ft}^2$  area, would certainly be capable of penetrating a containment structure. However, the fabrication plant does not occupy a square km but occupies about 1 acre of land. Thus the probability of a meteor striking the fabrication plant is about  $4 \times 10^{-10}/\text{yr}$ .

## 6. Flood

Most fabrication plants are designed to be above the Probable Maximum Flood (PMF). If the facility were inundated, the greatest danger would be from water seepage. The consequence of such seepage was not investigated in this report.

## B. RELEASE PROBABILITY FROM IN-PLANT ACCIDENTS

### 1. Data Sources

Although plutonium fabrication experience is limited, certain amounts of operating data are available. Experimental plutonium bearing fuel elements have been manufactured at Pacific Northwest Laboratories and Argonne National Laboratory for the past 15 years. The handling of plutonium for weapons work at Pacific Northwest Laboratories, Rocky Flats, Mound Laboratory and Los Alamos also has yielded invaluable experience on the handling and control of plutonium. Similarly, there has been a significant amount of experimental chemistry conducted in gloveboxes at all of the laboratories. While this work involves equipment somewhat different from that used in fuel fabrication, the handling of hazardous radioisotopes in various chemical regimes contributes valuable data for safety analyses. Although statistics from these operations are probably directly applicable, it should be noted that they were compiled in many cases in experimental

rather than industrial facilities. The very nature of this experimental work leads to a comparatively high accident rate.

Other sources of information have been the Safety Related Occurrences in Nuclear Facilities.<sup>(82)</sup> The data in this publication are most applicable to the problem at hand. Unfortunately, this is of limited use in its present form. The classification of the accidents reported is not directly applicable and little or no work has been done on reduction of the data to accident frequency. In 1967, a Reliability and Maintainability Data-Source Guide was prepared for the U. S. Naval Applied Science Laboratory, Brooklyn, New York.<sup>(83)</sup> The guide lists twenty-three Government reliability and maintainability data sources and over ninety-five sources of technical and scientific information for related engineering data. A preliminary review of one of the more promising reliability and maintainability data sources (FARADA PROGRAM) indicated that the data had been reduced to directly usable form, namely, failure rates per kinds of equipment based on operating experience.<sup>(84)</sup> The Failure Rate Data (FARADA) information is compiled into loose-leaf handbooks (SP-63-470) of five volumes, which are now updated quarterly. These data have some value in the study even though it is oriented entirely to the Army, Navy, Air Force, and NASA.

In summary, it is better to have some data, used judiciously, than none. It should be emphasized that there is no substitute for operating statistics. If preliminary analysis based on limited or inferred data are reexamined with time, they have positive benefits. If they are "cast in concrete" and the limited nature of their origins forgotten, they have dubious value.

## 2. Equipment Failure Rates and Accident Frequencies

In most cases, the failure of a single piece of equipment will not result in an uncontrolled release of plutonium to the environs. However, equipment failures or accidents involving resin columns, gloveboxes, criticality, and sintering furnaces, could, by themselves, result in small release through the normal building exhaust system. In all cases, the magnitude of accidental releases as well as release from normal

operation are highly dependent on the efficiency of the building final filter system. Analysis of the previously discussed failure rate data source has lead to estimates of failure rates for selected pieces of equipment. Additionally, the reliability of the HEPA filters for operating at quoted or design efficiencies has been developed. This information is summarized in Table 18 and Table 19. A discussion of the development of

TABLE 18. Estimates of the Occurrence Frequencies and Failure Rates of Major Equipment

	<u>Failures or Events Per Year</u>
Resin Columns <sup>(82)</sup>	<0.1
Dry Glovebox Operations <sup>(82)</sup>	<0.1
Autoclave <sup>(82)</sup>	<0.01
Sintering Furnaces <sup>(85)</sup>	<0.05
Ventilation Equipment <sup>(82)</sup>	
Direct Driven Fan	~0.01
Belt Driven Fan	0.4
Controls	0.08
Criticality	$8.6 \times 10^{-3}$

TABLE 19. Efficiency of a Two Stage Building Exhaust Filter System Under Various Conditions

<u>Condition</u>	<u>Most Probable Filter Transmission</u>
Filters not tested prior to installation or in place, 8% of filter operating less than design efficiency	$7 \times 10^{-5(a)}$
Filters tested prior to installation and in place, routine inspection and replacement program	$1 \times 10^{-5}$

a. Assumes a filter efficiency of 0.95 for the defective filters.



some of these values follows. The estimated occurrence rate of a criticality in a fuel plant is also included in Table 18. The basis for this value is summarized in the text.

a. Resin Columns

Four incidents have been reported resulting in a release of radioactivity into the facility as a result of thermochemical instabilities in an ion-exchange processing. Plutonium solutions were involved in three and a uranium solution in the other.<sup>(86)</sup>

Thermal transients, if not properly controlled, arise from radiolytic heating, excessive applied heat, oxidation due to nitration under uncontrolled conditions, or heating by strong oxidants. During the excursion, the column heats, becomes pressurized, and could rupture to discharge resin and solution. The accident has been successfully modeled and the operating conditions required to avoid the incident are well known.<sup>(87)</sup> Conscientiously applied administrative controls, reliable monitoring equipment and adequate safety features would presumably successfully control the process.

There would still exist some finite probability of a simultaneous breakdown of the administrative controls (operator inattention, etc.), monitoring equipment failure (thermocouples and amplifiers) and inoperative safety devices (rupture disks or check valves). If failure rate data can be obtained or projected from operating history on equipment in related operations, and the contribution from operator error adequately evaluated,<sup>(88)</sup> the frequency of a resin column fire and explosion can be developed.

Prior work,<sup>(89)</sup> assuming that the column inventory was about 1400 grams and that the glovebox ruptured postulated that the resulting plutonium release through the filters to the stack is about 0.00003 g (30  $\mu$ g) for this incident.

b. Glovebox Explosions

Since 1967, four glovebox explosions or fires have been reported in which significant amounts of materials were released. One involved a hood, another an explosion in a vacuum dry box, a third was a glovebox explosion and fire involving  $^{238}\text{Pu}$  and the fourth was the Rocky Flats

plutonium facility fire. The last involved plutonium metal and is not representative of a process in a fuel fabrication plant.

The safety problem from a glovebox explosion and fire again would be expulsion of plutonium compounds into the work area and possible carrying out of plutonium through the exhaust system to the environs.

There are several passive and active safety actions used in the plant to prevent glovebox fires and contain them. Normal operating procedures would specify that:

- Flammable solvents with flash points below 140 °F be not permitted inside the boxes.
- Inert atmospheres be used in boxes where heat is present.
- Trash (such as wipes) be kept to a minimum inside boxes and stored in covered metal containers.
- PuO<sub>2</sub> and UO<sub>2</sub> not actually in process inside the gloveboxes be kept inside metal containers.
- The dry operation involves PuO<sub>2</sub> and UO<sub>2</sub> which are not flammable materials.

Well-designed gloveboxes contain heat detectors and fire extinguishers mounted inside the boxes with devices for automatically initiated fire suppression. Filters are preceded by fiberglass prefilters, and the final filter bank is protected from hot or burning debris automatically by a temperature activated deluge system.

The probability of a glovebox explosion and fire is low, but again a finite and a tentative number has been assigned based on available information. The accidents analyzed in this study involving a fire and an explosion in a glovebox are extreme examples of this type of accident. It is expected that these types of accidents would have a probability of occurrence at least a factor of 100 less than the listed value in Table 15.

#### c. Sintering Furnace

Published operating failure rate data (explosions) were not available for sintering furnaces although it appears that the numbers of sintering

furnaces in fuels fabrication facilities are available. A major vendor for example, has 30 units in service at uranium plants and one at a plutonium facility and four vendors furnish over 95% of the large units commercially in use.

Accidents have occurred at sintering furnaces resulting from detonations of explosive mixtures of oxygen from air and hydrogen, the reducing gas. The explosive mixtures develop usually from:

- Incomplete removal of air (oxygen) from a cold furnace at startup before introducing the forming gas containing hydrogen.
- In-leakage of air (oxygen into the furnace during operation with the furnace at temperature).
- In-leakage of air (oxygen) in the line exhausting the forming gas mixture ( $H_2 > 12\%$ ) without first diluting the  $H_2$  content to less than a non-explosive content (about 5%  $H_2$ )

Basic safety features on all sintering furnaces include an automatic nitrogen flush with hydrogen isolation until the flush is complete (timed out) for furnace startup, and, for protection when the furnace is at temperature, and automatic transfer from  $H_2$  to  $N_2$  make-up if the hydrogen pressure fails or a power failure develops.

Forming gas mixtures are established in some sintering furnaces entirely by valving and monitoring individual gas flow streams. The probability for operator error that would result in the furnace filling with  $H_2$  is finite. When mixing gases automatically at the furnace with a metering device, orifices have become plugged and the gas mixture can suddenly revert to 100%  $H_2$ . Flow alarms are recommended but apparently seldom used. Their reduction of the risk is dependent upon their own reliability.

A reasonable assessment of the potential for an accident in a sintering furnace would be estimated from compiling operating experience, if possible; by applying failure rates to the safety equipment and to controllers; and by assessing the contribution of operating errors.

d. Criticality

The objective of this section is to estimate the probability of a criticality accident in fuel fabrication facilities. This probability,  $P_{ca}$ , will be estimated as:

$$P_{ca} = \frac{A_f}{Q_f}$$

where  $A_f$  = Number of criticality accidents in Fuel Fabrication related plants to date

$Q_f$  = Quantity of fuel processed through Fuel Fabrication related plants to date

The number of criticality accidents to date is four.<sup>(90)</sup> These accidents have all occurred in operating facilities in the United States from activities that could be related to fuel fabrication if the scrap recovery operation is included, as all four occurred during scrap recovery. Two involved plutonium, two highly enriched uranium, and all four occurred during wet chemistry operations. No criticality accident has occurred in the United States during the processing of dry material and no criticality incident has occurred in the processing of wet or dry material of low enrichment. The locations, dates, and personnel exposures resulting from these accidents are summarized in Table 20.

Four accidents in 25 years is obviously very limited experience and does not give a comprehensive picture of the ways criticality accidents can occur. A more proper appraisal perhaps would be to include "near-misses"; but quantitative information of this type is not available. Nonetheless, if all facilities similar to those in which the above accidents have occurred are included in the assessment of  $Q$ , the probability estimate should be valid for the intent of this study.

The scope of the probability estimate thus includes facilities that process, fabricate, recover, or otherwise handle non-irradiated plutonium, slightly enriched uranium, and fully enriched uranium. Fuel reprocessing facilities and facilities for processing U-233 and Pu-238 are not included at this time.

TABLE 20. Summary of Criticality Accidents Included in This Study

<u>Location</u>	<u>Date</u>	<u>Fissile Material Involved</u>	<u>Form of Fissile Material</u>	<u>Deaths</u>	<u>Personnel Exposures, Rem</u>	
Y-12 Plant	6/58	U(93) <sup>(a)</sup>	Solution	0	461	298
					428	86
					413	86
					341	29
LASL	12/58	Plutonium	Solution	1	12,000	
					134	
					53	
Hanford	4/62	Plutonium	Solution	0	110	
					43	
					19	
UNC	7/64	U (93)	Solution	1	10,000	
					80	
					80	

a. U(93) means uranium whose 235-U enrichment is 93 wt%.

The choice of "plant years" for Q is recognized as not the best basis, but it is the only basis that could be estimated quantitatively at this time. A better basis would be total fuel throughput, with allowance for differences in the fuel forms and fuel reactivities processed. Such figures of fuel processed through USAEC plants, of course, are not available. So a different basis must be used. Consideration was given to plant floor area, number of employees, plant design capacity and simply years of operation, for the basis. These figures, too, are difficult to obtain and lead to various degrees of inaccuracy. Consequently, for the purposes of the present study, it was decided to use "plant years" for the probability estimate.

The "number of plant years" of operation to the present is estimated to be 432. This estimate was obtained by summing plants' years of operations since 1942, as shown on Table 21.<sup>(91 - 94)</sup>

Imbalances in production rates should be partially compensated by the large number of small scale plants that are included in the tally. It is

recognized that some of the dates may be off by a few years; but overall, the estimate is considerably better than a guess. Improvement of the accuracy of this information will be the subject of a later study.

TABLE 21. Estimate of Plant-Years of Production Since 1942 Involving Uranium and Plutonium Fuel Fabrication

<u>Plant</u>	<u>Estimated Dates</u>	<u>Estimated Plant Years</u>
Hanford	1944-1973	29
Savannah River Laboratory	1954-1973	19
Los Alamos Scientific Laboratory	1943-1973	30
National Lead Company of Ohio	1944-1973	29
Oak Ridge National Laboratory	1943-1973	30
Lawrence Radiation Laboratory	1949-1973	24
Argonne National Laboratory	1949-1973	24
Aerojet General Nuclear	1955-1970	15
Atomics International	1955-1973	18
Babcock & Wilcox	1957-1973	16
Clevite Research Corporation	1957-1969	12
Combustion Engineering	1955-1973	18
Curtiss-Wright Davison	1955-1973	10
Gulf General Atomics	1958-1973	15
General Electric	1955-1973	18
Gulf United Nuclear	1971-1973	2
M & C Nuclear, Incorporated	1961-1965	4
Exxon	1971-1973	2
Mallinckrodt Nuclear Corporation	1960-1970	10
Martin Company	1960-1970	10
Kerr-McGee	1969-1973	4
National Carbon Company	1960-1965	5
National Lead Company	1962-1973	11
Engelhard Industries, Incorporated	1957-1970	13
Nuclear Development Corporation of America	1957-1968	11
Nuclear Materials and Equipment Corporation	1960-1971	11
Sylvania-Corning Nuclear Corporation	1960-1968	8
Westinghouse Electric Corporation	1955-1973	18
United Nuclear	1957-1973	<u>16</u>
	Total	432

From the foregoing, the probability of a criticality accident in uranium and plutonium fuel fabrication facilities is estimated to be as follows:

$$P_f = \frac{4 \text{ criticality accidents}}{432 \text{ plant years}}$$

$$P_f = 9 \times 10^{-3} \frac{\text{Criticality accidents}}{\text{plant year}}$$

This probability estimate can undoubtedly be further improved by considering greater production detail. Improvements that could be considered at a future date are as follows:

- Obtain more accurate dates of plant startups
- Make allowance for reactivity of fuels processed.
- Obtain a better basis for quantities of fuel produced to date.
- Consider types of fuel operations - wet versus dry.

e. High Efficiency Filter (HEPA)

In fuel fabrication facilities, the filter in the glovebox and building exhaust systems provide the final protection between the building process air and the environment. For this reason it is very important that the filters be within the design specifications. On the average, 3% to 6% of the HEPA are defective upon receipt from the vendor with the percentage of rejects on some shipments being significantly higher.<sup>(95, 96)</sup> Filters are judged defective for excessive penetration, filter medium failure, frame failure, gasket failure, and damage during shipment. Recent tests on 13,000 filters showed that rejections due to these defects occurred in the ratio 3.2:4.0:2.3:1.0:1.3. About 35% to 45% of the defective filters have filter efficiencies less than design specifications.<sup>(96)</sup> The efficiency for the filters judged to be defective due to excessive penetration is in the 95% to 99% range.<sup>(95)</sup>

Failures during installation due to improper seating or careless handling seem to occur at somewhat lower percentages, namely about 1 to 2%.<sup>(97)</sup> Hence, if testing isn't done prior to or following installation, 4 to 8% of all HEPA filters installed will leak and be significantly less efficient than the design value.

Failures of the filters while they are in service are a function of their environment. Continuous monitoring of the pressure drop across the filters should be coupled with a routine filter replacement program to insure a reliable filter system. The practice of such quality assurance procedures by AEC contractors over the past 25 years has made in-service HEPA failure an almost unheard of phenomenon.

Because of these statistical defects, a fuel fabrication facility should have a pre-installation program for examining the HEPA filters to assure they meet design specifications.

### 3. Operating Data Obtained from National Safety Information Center

The National Safety Information Center (NSIC) organizes in several ways the incidents that are reported to the AEC by facility licensees. The reporting requirements for facility licensees are described in Parts 20, 40, 50, 70 and 73 of Title 10 of the Code of Federal Regulations. Information from reports of abnormal occurrences or unusual events provided most of the source material in the compilations developed by NSIC. Despite the extensive cataloging and organization of the information, more information about each occurrence seems necessary to contribute to a study on risk using frequencies of accidents, failure rates of equipment, contributions by design inadequacies and the like. However, several informative comparisons are possible using the data as indicated in the following tabulations. A summary of the reported occurrences is shown in Table 22.

To be properly applicable in accident reviews, each of the personnel exposure incidents, for example, should be reviewed to determine the actual operational step of the fabrication process in which it occurred. This should be repeated for each of the other three categories.



TABLE 22. Occurrence Frequencies

	<u>No. for 1970-71</u>	<u>Avg/yr</u>	<u>Est. Frequency,<sup>(a)</sup> No./plant/yr</u>
• Personnel Exposure <sup>(b)</sup>	52	26	3.2
• Contamination in the Work Areas	10	5	0.6
• Local Fire	3	1.5	0.2
• Contamination Outside Work Area	2	1	0.1

- a. In order to normalize to a one metric ton per day plant, existing facilities were weighted as shown: General Electric (3), Westinghouse (2), Babcock & Wilcox (1/2), Combustion Engineering (1/2), Numec (1 1/2), Kerr-McGee (1), Gulf (1), Nuclear Fuel Services (1 1/2).
- b. Personnel Exposure reports involved those occurrences required by 10 CFR 20 Sect. 405.

For purposes of demonstration, totals on incidents for each of the categories were averaged from these plants for the last two years that this information was published; namely, 1970 and 1971, and they are shown in column two. From strictly conjecture and only for the purposes of illustration, it was suggested that in total, the averages were approximately equal in output to eight fuel fabrication plants of one tonne per day capacity. This was used to develop the estimated frequency; number per plant per year.

The same data has been organized as to the cause of release in Table 23.

TABLE 23. Occurrence Causes

	<u>No. for 1970-71</u>	<u>Avg/yr</u>	<u>Est. Frequency, No./plant/yr</u>
• Equipment Failure	24	12	1.5
• Operating Error	17	8.5	1.1
• Administrative Control	12	6	0.75
• Design Error	10	5	0.62
• Maintenance Error	4	2	0.25
• Installation Error	0	0	--

Again, detailed information as to kinds of equipment that failed and at which work stations must be sought out for proper analysis. These data were extracted from the abstracts summarizing incidents only from plants involved in fuel fabrication.

The following table, using slightly different categories than above, was gathered from the same sources, and occurrences from related industries, primarily the preparation of radioactive pharmaceuticals, were included. Information was usually not available in the abstract to determine much about each occurrence. Therefore, the incidents at this time are categorized only in general terms in Table 24.

TABLE 24. Occurrence Frequencies for Fuel Fabrication Plants Plus Related Industry

<u>Event</u>	<u>70</u>	<u>71</u>	<u>Avg/yr</u>	<u>Est. Frequency No./Plant/Yr</u>
Spills	6	19	13	1.3
Failure of Glovebox or Hood	23	18	20	2.0
Exceeded Design Limitations	10	13	12	1.2

If one knew what the related industries, in total, contributed to the statistics, accident occurrence rates could be developed considering a wider base which includes the related industries. Arbitrarily assuming that the activities of the pharmaceutical houses and related industry included above were equivalent to 2 additional fuel fabrication plants, accident occurrence frequencies have been estimated. To repeat, to compile a meaningful study of accident frequencies, the records from which the data above were summarized must be reviewed in detail. From this, an accident frequency rate per powder handling glovebox per year or per scrap recovery glovebox per year, can be developed. This is the kind of compilation needed to develop useful concepts of risks for the generic one MT/day fuel fabrication plant. It appears that records may be available to obtain it if additional effort is expended.

## IX. ASSESSMENT OF ENVIRONMENTAL RISK

Individuals living in the environs of a plutonium fuels fabrication facility will be exposed to emissions from normal plant operation and those resulting from accidents. Since accidental releases are normally of greater magnitude and occur infrequently, they must be placed in perspective with releases from normal operation before their relative risk can be assessed. One method of doing this is to take the insurance man's approach by weighting the estimated consequence of a release by its frequency of occurrence. The output of this approach is commonly referred to as "risk". When this is done, all release modes can be directly compared and the total risk of the plant quantitatively assessed.

In making such a risk assessment, one must conduct a comprehensive safety analysis. This analysis must include estimates of the magnitude of credible release modes; both from accidents and normal operation. A typical logic diagram for an analysis of this type for a fuels fabrication facility is shown in Figure 16. Potential release modes are input into the Facility "Model" which is characterized by material and process parameters and building design criteria. The magnitude of the inplant release and the facility design characteristics defines the quantity of material released. The released material is dispersed and acted upon in the plant surroundings in a manner predicted by the Environmental "Model". Different atmospheric dispersion models should be used for continuous and short duration releases. Estimates of the dose to the organs of interest for an individual exposed to the dispersed material are made using the Dose "Model". The results of these dose calculations are referred to as the "consequences of a release".

Coupling this output with the probability of the release, one gets a prediction of the environmental risk of that particular release mode. A summation of the "risk" of all credible releases, both from normal operation and from accidents, provide a measure of the total imposed risk of the facility. An analysis such as this permits direct risk comparison of dissimilar facilities.

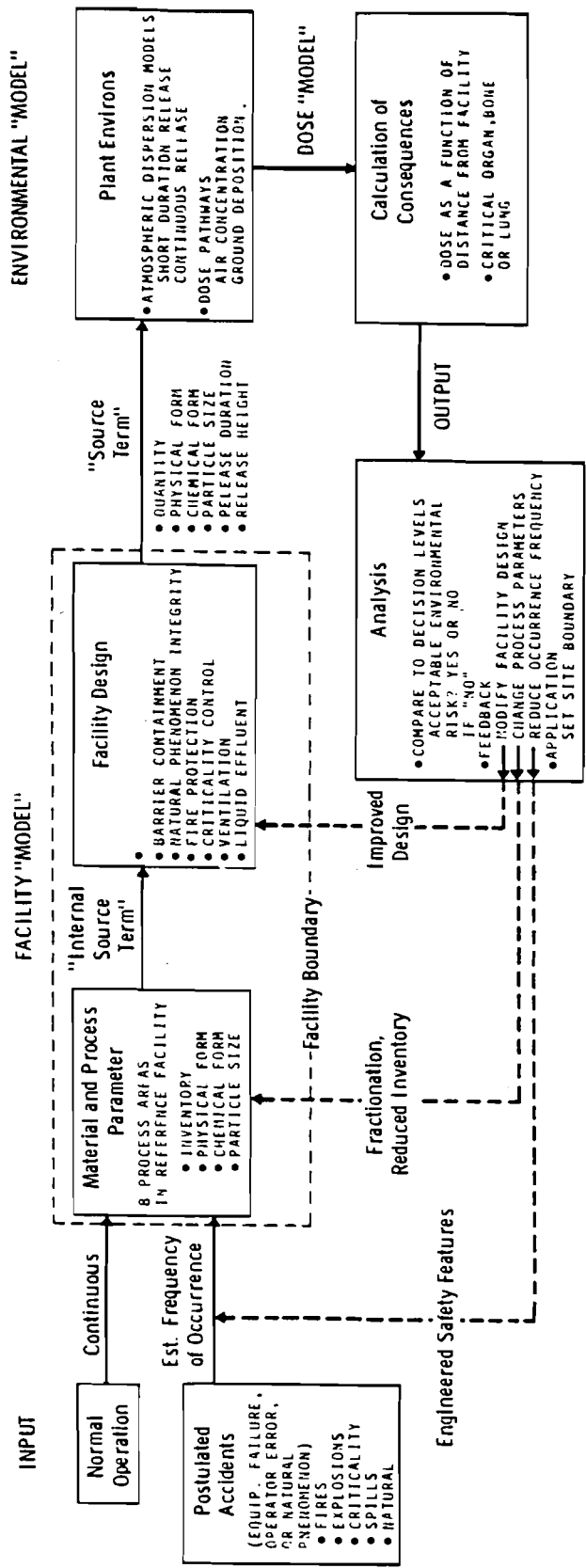


FIGURE 16. Safety Analysis Logic for a Plutonium Fuel Fabrication Facility

A summary of the risk analysis for the reference facility is presented in Table 25. The risk for all of the release modes discussed in this report have been quantified in the final column called, "Annual Dose Commitment Risk." This column shows the annual risk of incurring appropriate dose commitments to the noted critical organs. Unlike a reactor where "Annual Risk" is used as an index, risk for a plutonium facility must be assessed on a "Dose Commitment" basis. This is because the dose to the critical organs from deposited plutonium is delivered over a long period of time and therefore cannot be adequately expressed on an annual dose "received" basis as in the case of a reactor facility.

For releases for normal operation and accidents discussed in this report, the annual dose commitment risk to an individual 1,000 meters from the facility is estimated to be less than 16 mrem to the bone, 1.5 mrem to the lung, 1.7 mrem to the thyroid, and 0.4 mrem to the whole body. It is expected that the remaining spectrum of accidents will not significantly increase these values.

For this facility, the analysis clearly indicates that the dose to the bone is the major consideration in evaluating the environmental impact of a plutonium fuels facility. Based on the results shown in Table 25, the largest annual dose commitment to an individual in the environs results from an accident involving plutonium in a soluble form. Therefore, special emphasis should be directed toward refining the probability of such releases and, as necessary, provide additional engineered safety features and administrative control for operations involving plutonium in this chemical form. Additionally, if a criticality accident of greater magnitude can be deemed credible, the thyroid dose from radioactive iodine becomes a significant factor. The removal effectiveness of iodine for the facility filter system should be reviewed to determine if credit can be taken for iodine removal in the safety analysis. If this is found inadequate, consideration should be given to the addition of filters containing iodine removal media.

In summary, this report provides a comprehensive view of the technological considerations germane in analyzing the safety of a plutonium fuel

TABLE 25. Annual Risk of an Individual in the Environs of a Plutonium Fuels Fabrication Facility

Condition	Source Term, g	Radioactive Contaminant	Critical Organ	Dose at $10^3$ (c) meter, mrem	Probability of Release, yr <sup>-1</sup>	Annual "Dose Commitment" Risk, mrem(e)
Normal Operation	$5 \times 10^{-6}$ (S)(a)	Pu Mix(b)	bone	4/50 yr(d)	1	4.0
			lung	0.06/50 yr	1	0.06
Criticality	$3 \times 10^{-5}$ (S)	Pu Mix direct radiation Noble gases and Halogens iodine	bone	28/50 yr	$8.6 \times 10^{-3}$	0.24
			whole body	16	$8.6 \times 10^{-3}$	0.14
			whole body	31	$8.6 \times 10^{-3}$	0.27
Explosion	$10^{-2}$ (S)	Pu Mix	thyroid	200	$8.6 \times 10^{-3}$	1.7
Fire	$1.1 \times 10^{-2}$ (I)	Pu Mix	bone	9400/50 yr	$\approx 10^{-3}$	9.4
			lung	70/2 yr	$<10^{-2}$	$<0.70$
Glovebox Damage	$10^{-2}$ (I)	Pu Mix	bone	700/50 yr	$<10^{-2}$	$<7.0$
			lung	60/2 yr	$<10^{-2}$	$<0.60$
Facility Fire	$10^{-2}$ (I)	Pu Mix	bone	600/50 yr	$<10^{-2}$	$<6.0$
			lung	60/2 yr	$2 \times 10^{-4}$	0.01
Resin Column Fire	$3 \times 10^{-5}$ (S)	Pu Mix	bone	600/50 yr	$2 \times 10^{-4}$	0.12
			bone	28/50 yr	$<1 \times 10^{-1}$	$<2.8$
Normal Operation				TOTALS:	Bone	4.0 mrem
					Lung	0.6 mrem
Accident				TOTALS:	Bone	$<25$
					Lung	$<1.3$
					Thyroid	1.7 mrem
					Whole Body	0.41 mrem

a. I = insoluble, S = soluble.

b. Pu Mix = reference mixture of plutonium and americium.

c. Estimated using the ICRP Pub 2 lung model (ILM).

d. 4/50 yr means that the 4 mrem is delivered to the bone over a period of 50 yr.

e. The probable dose to which an individual is annually committed to receive. This could be instantaneously received or it could be delivered over an extended period as in the case of the bone and lung.

fabrication facility. Additionally, it quantifies the risk for the fuel fabrication operations and more importantly, identifies the areas where special safety emphasis should be placed. Effort should continue toward developing improved safety procedures and refining the release probabilities for operations involving the greatest risk.

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APPENDIX A  
PLUTONIUM FUEL PROCESSING AREAS IN REFERENCE PLANT

APPENDIX A

PLUTONIUM FUEL PROCESSING AREAS IN REFERENCE PLANT

PROCESS AREA I - FUEL STORAGE

Typical Operations: Plutonium and mixed plutonium-uranium in various forms is stored. Plutonium nitrate solutions are blended in large volume vessels to achieve isotopic uniformity.

Inventory: 1000-3000 kg Pu < 5000 kg U (mixed with 4 wt% Pu)

Chemical Forms: Pu (NO<sub>3</sub>)<sub>4</sub>, soluble  
PuO<sub>2</sub>, insoluble  
PuO<sub>2</sub>-UO<sub>2</sub>, insoluble  
Small quantities of miscellaneous soluble Pu compounds

Physical Forms: PuO<sub>2</sub> powders, dispersible, particle size 0.1-80 μm  
PuO<sub>2</sub>-UO<sub>2</sub> powders, dispersible, particle size 0.1-1400 μm  
PuO<sub>2</sub>-UO<sub>2</sub> pellets, essentially nondispersible  
Solutions (primarily nitrate) containing soluble Pu, dispersible  
Slurries containing insoluble Pu, dispersible  
PuO<sub>2</sub>-UO<sub>2</sub> fuel rods and elements, nondispersible

Comments: 1. The homogenization tank, or innerconnected tanks, for blending the plutonium nitrate solution will typically have a volume of 1000 liters or greater. This tank or tanks will be of heavy wall construction which will provide a high degree of protection for the contents.



2. The quantitative division of the plutonium between the various chemical and physical forms will vary in time.
3. All of the plutonium in the plant, except for the in-process inventory of the order of a few hundred kilograms of plutonium, will be in the fuel storage area. Typically, about half of the in-process inventory of plutonium will be in the dispersible forms of solutions, slurries and powders; the remainder will be in the form of pellets or encapsulated rods. The decontaminated rod and fuel element storage will be separated from other storage to avoid contamination.
4. The uranium storage area will be separate from the plutonium and mixed plutonium-uranium storage area. Typical uranium forms will be  $UF_6$ ,  $UO_2$ ,  $UO_3$ ,  $UO_2(NO_3)_2$  and  $U_3O_8$  (yellowcake). The typical maximum uranium inventory will be  $10^4$ -  $10^5$  kg.

#### PROCESS AREA II - CONVERSION

Typical Operations: Plutonium nitrate solution is precipitated as Pu (IV) oxalate by the addition of oxalic acid. Hydrogen peroxide is sometimes added in this step for valence adjustment of the plutonium. The Pu oxalate is filtered, dried, and calcined to  $PuO_2$  at a temperature of 650-850°C. The  $PuO_2$  powder from the calciner has particle sizes ranging from 0.3 to 80  $\mu m$ . The larger particles are loosely bound agglomerates. Approximately 95% of the particles are larger than 1  $\mu m$  and the average particle size is 6-10  $\mu m$ .

Inventory: 50 kg Pu

Chemical Forms: Pu oxalate, soluble  
 $PuO_2$ , insoluble

Physical Forms: Pu oxalate slurry and wet powder, dispersible  
dry PuO<sub>2</sub> powder, dispersible

Comments: All operations will be conducted in equipment that is critically safe by geometry. Therefore the inventory is limited by process throughput requirements rather than by criticality safety considerations.

### PROCESS AREA III - POWDER TREATMENT

Typical Operations: The calcined PuO<sub>2</sub> powder is crushed and screened to obtain particles of a few micrometers and then blended with UO<sub>2</sub> powder. The blended PuO<sub>2</sub>-UO<sub>2</sub> is milled, agglomerated (either by dry compaction or binder addition), and granulated to 14-180 mesh (85-1400 μm) granules to produce a free flowing feed powder for the pellet press. The powder is cold pressed to yield green pellets (0.3-0.5 inch diameter) of 40-55% of the theoretical density.

Inventory: 50 kg Pu (total) 11.3 kg Pu (per batch)  
After blending the Pu is mixed (4 wt% PuO<sub>2</sub>-96 wt% UO<sub>2</sub>) with UO<sub>2</sub>

Chemical Forms: PuO<sub>2</sub>, insoluble  
UO<sub>2</sub>, insoluble

Physical Forms: PuO<sub>2</sub> powder, dispersible  
UO<sub>2</sub> powder, dispersible  
PuO<sub>2</sub>-UO<sub>2</sub> powder, dispersible  
Green PuO<sub>2</sub>-UO<sub>2</sub> pellets, not easily dispersed

Comments: These batch-type operations will be conducted dry or with a controlled degree of moderation because optimally moderated criticality safety limits are too restrictive on batch size. A large batch size is especially desirable in blending operations to assure uniform powder characteristics.

#### PROCESS AREA IV - PELLET TREATMENT

Typical Operations: The green pellets are sintered at 1500-1700 °C in a reducing atmosphere (6-15 wt% hydrogen in nitrogen). If an organic binder was used, this operation may be preceded by presintering. The sintered pellets are centerless ground (up to 2-4% of the material is removed) to a specified diameter, washed, dried and outgassed (at ~600-800 °C in high vacuum).

Inventory: 1700 kg PuO<sub>2</sub>-UO<sub>2</sub> (60 kg Pu) in pellets  
1.6 kg Pu in solution or slurry

Chemical Forms: PuO<sub>2</sub>-UO<sub>2</sub>, insoluble

Physical Forms: PuO<sub>2</sub>-UO<sub>2</sub> green (not easily dispersed) and sintered (essentially nondispersible) pellets  
PuO<sub>2</sub>-UO<sub>2</sub> powder in solution or slurry, dispersible

Comments: The centerless grinding operation yields PuO<sub>2</sub>-UO<sub>2</sub> dust or slurry depending on whether the operation is performed wet (the usual case) or dry. The washing operation yields a PuO<sub>2</sub>-UO<sub>2</sub> solution or slurry.

#### PROCESS AREA V - PELLET LOADING AND FUEL ROD ENCAPSULATION

Typical Operations: The out-gassed PuO<sub>2</sub>-UO<sub>2</sub> pellets are loaded into fuel rods. The rods are then welded closed and decontaminated.

Inventory: 850 kg PuO<sub>2</sub>-UO<sub>2</sub> (30 kg Pu)

Chemical Forms: PuO<sub>2</sub>-UO<sub>2</sub>, insoluble

Physical Forms: Sintered PuO<sub>2</sub>-UO<sub>2</sub> pellets, essentially nondispersible  
Fuel rods containing sintered PuO<sub>2</sub>-UO<sub>2</sub> pellets, nondispersible

PROCESS AREA VI - TESTING AND TREATMENT OF FUEL RODS AND ELEMENTS

Typical Operations: The decontaminated rods are degreased, etched, leak tested, autoclaved (at pressures of ~1000 psi), assembled into fuel elements, put through final inspection and prepared for shipment.

Inventory: 1700 kg PuO<sub>2</sub>-UO<sub>2</sub> (60 kg Pu)

Chemical Forms: PuO<sub>2</sub>-UO<sub>2</sub>, insoluble

Physical Forms: Fuel Rods and elements, nondispersible

PROCESS AREA VII - LABORATORY

Typical Operations: Various analytical and quality control operations are conducted. These may include density measurements, moisture analysis, oxygen-to-metal ratio analysis, gas release analysis, various chemical composition analyses (e.g., emission spectroscopy), radio-chemical analysis, and autoradiography.

Inventory: 5 kg Pu (total) with the majority mixed with UO<sub>2</sub>  
≤230 g Pu (per batch)

Chemical Forms: Diverse, both soluble and insoluble

Physical Forms: Diverse, assume all are dispersible

Comments: 1. Normally much less than 230 g Pu will be handled in any one operation.  
2. A wide variety of chemical reagents are used in the operations.

PROCESS AREA VIII - PROCESSING AND RECOVERY OF SCRAP AND WASTE

Typical Operations: Scrap and waste that may be processed include:

1. Noncontaminated PuO<sub>2</sub>-UO<sub>2</sub> and PuO<sub>2</sub> powders

2. Noncontaminated  $\text{PuO}_2\text{-UO}_2$  pellets
3. Chemically contaminated  $\text{PuO}_2\text{-UO}_2$  pellets
4. Chemically contaminated  $\text{PuO}_2\text{-UO}_2$  and  $\text{PuO}_2$  powders
5. Combustible Pu-contaminated materials (e.g., paper, clothing, filters).
6. "Noncombustible" Pu-contaminated materials (e.g., glove box gloves, plastics, tools)
7. Pu-contaminated aqueous and organic solutions and slurries from process waste streams (e.g., grinder sludge), laboratory operations, clean-up operations, etc.

The scrap and waste is treated in various ways depending on its characteristics. The processing for item one could be merely recalcining prior to return to the powder treatment area. Other operations that may be required to process the waste and scrap include:

1. Calcining of the combustible materials in an oxidizing atmosphere to remove organics
2. Dissolution in  $\text{HNO}_3$
3. Leaching of "noncombustible" materials
4. Anion exchange or solvent extraction of chemically contaminated materials
5. Distillation to increase the fissionable material concentration in solutions
6. Packaging of unrecoverable waste for disposal

Inventory: 25 kg Pu, may or may not be combined with U

Chemical Forms: Diverse, both soluble and insoluble

Physical Forms: Diverse, assume all are dispersible

APPENDIX B

ISOTOPE DOSE CURVES

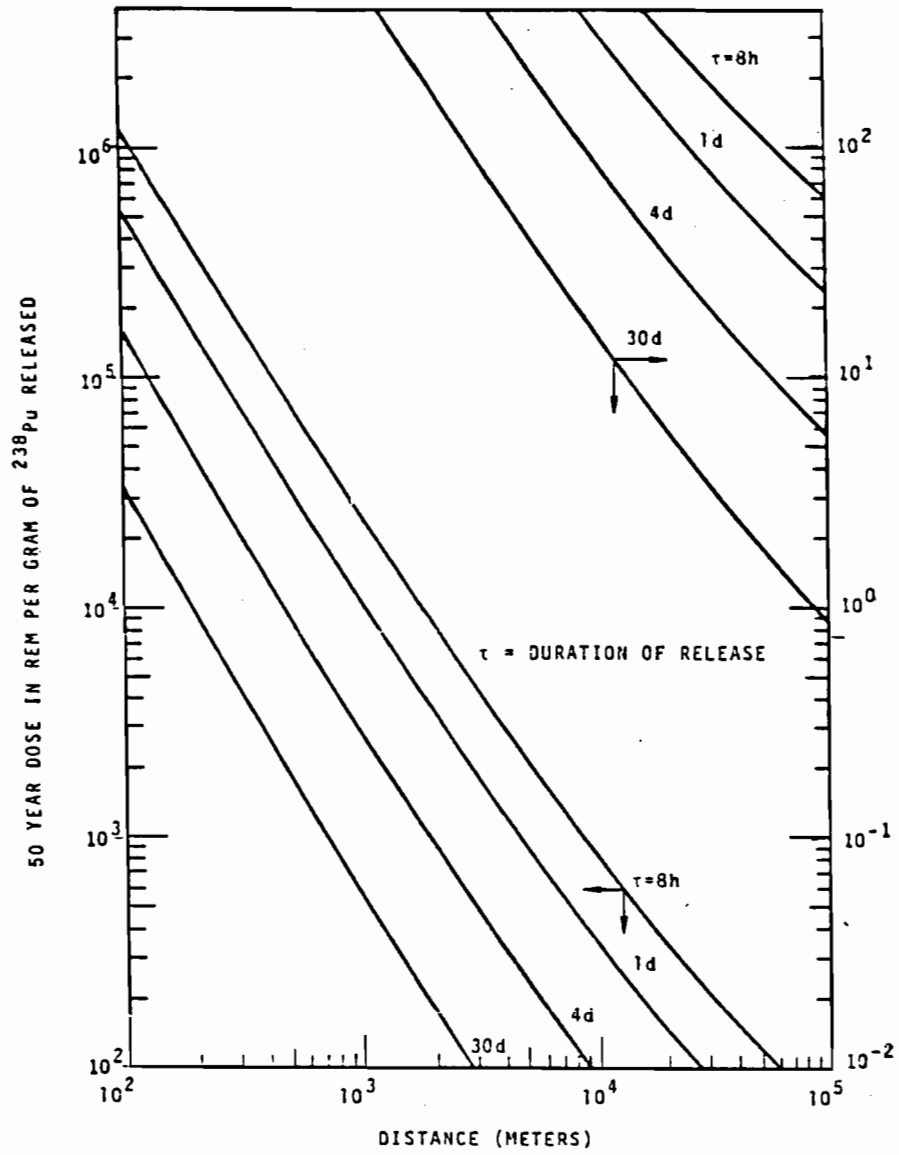


FIGURE B.1. Dose to Bone from Acute Inhalation of Plutonium-238, Ground Level Release

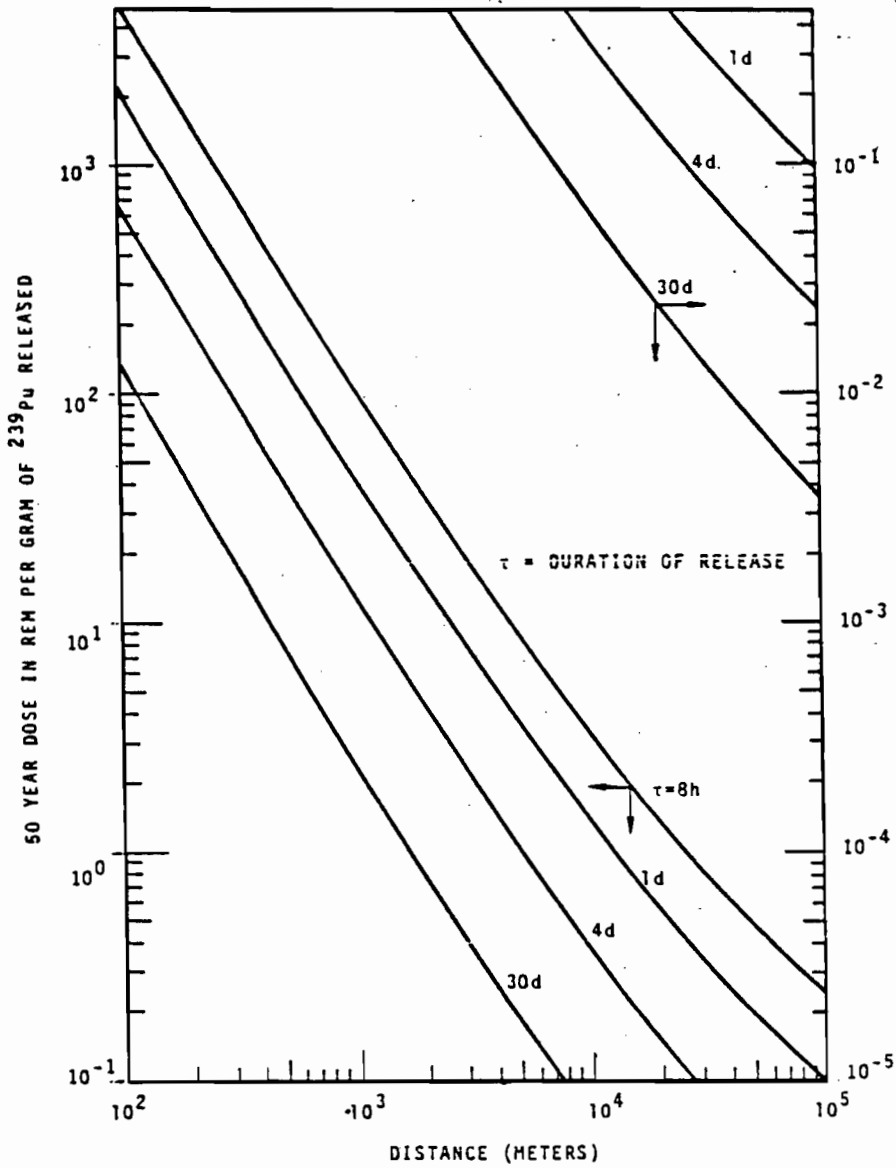


FIGURE B.2. Dose to Bone from Acute Inhalation of Plutonium-239, Ground Level Release



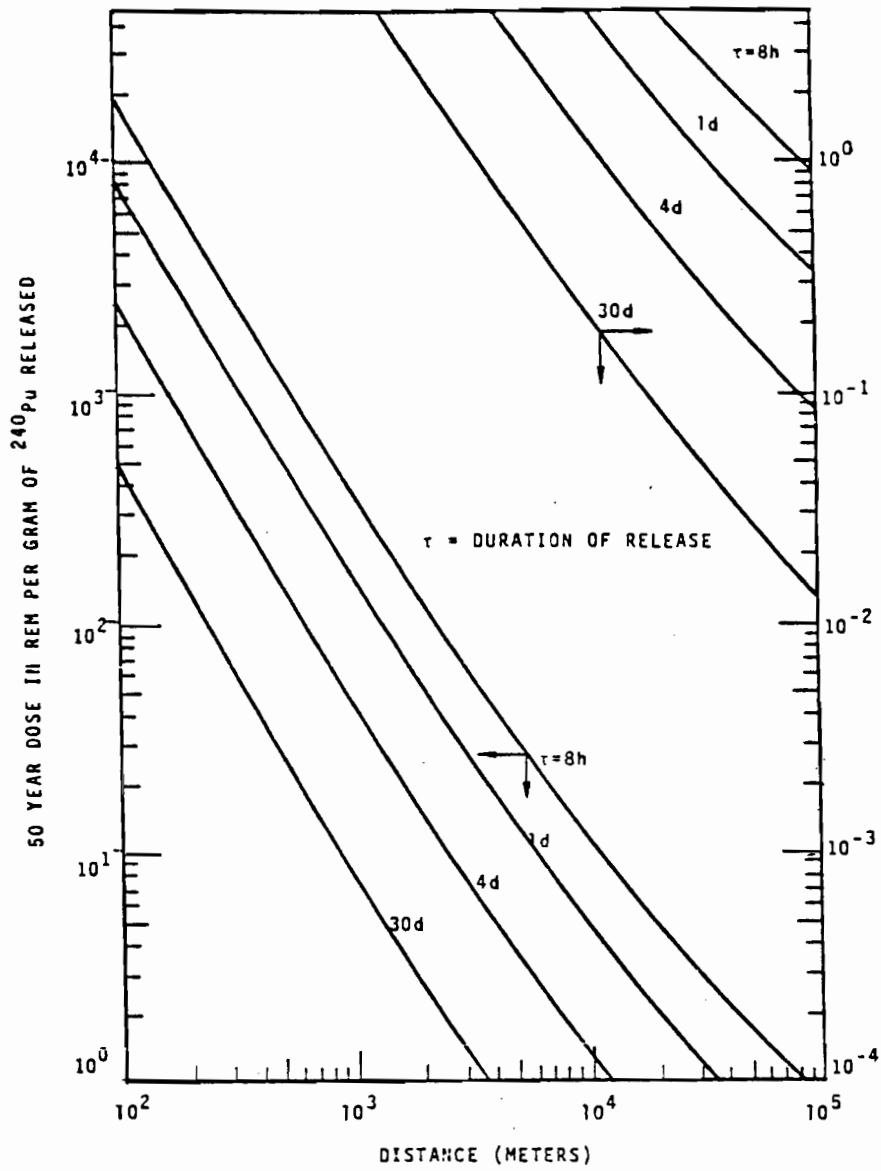


FIGURE B.3. Dose to Bone from Acute Inhalation of Plutonium-240, Ground Level Release

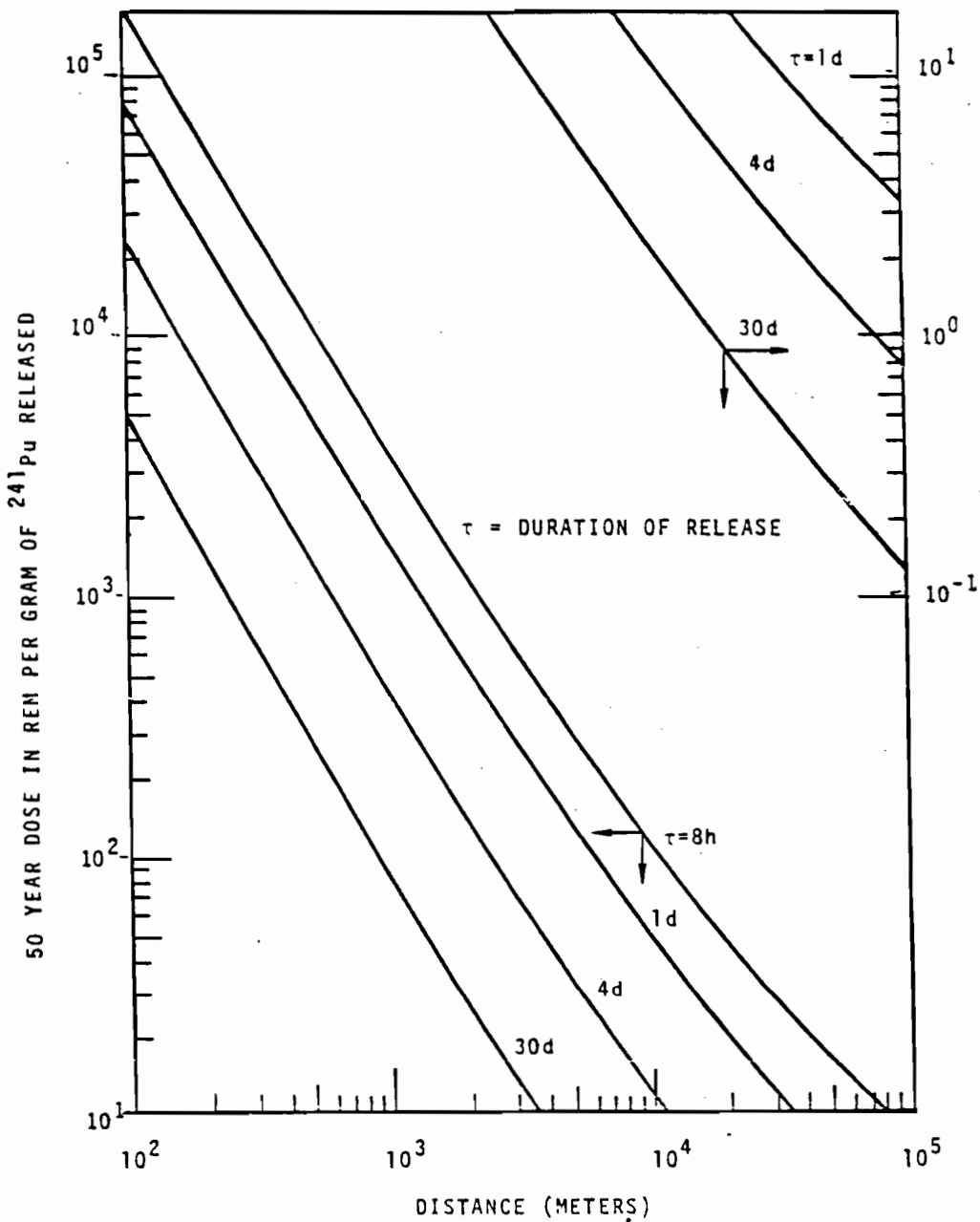
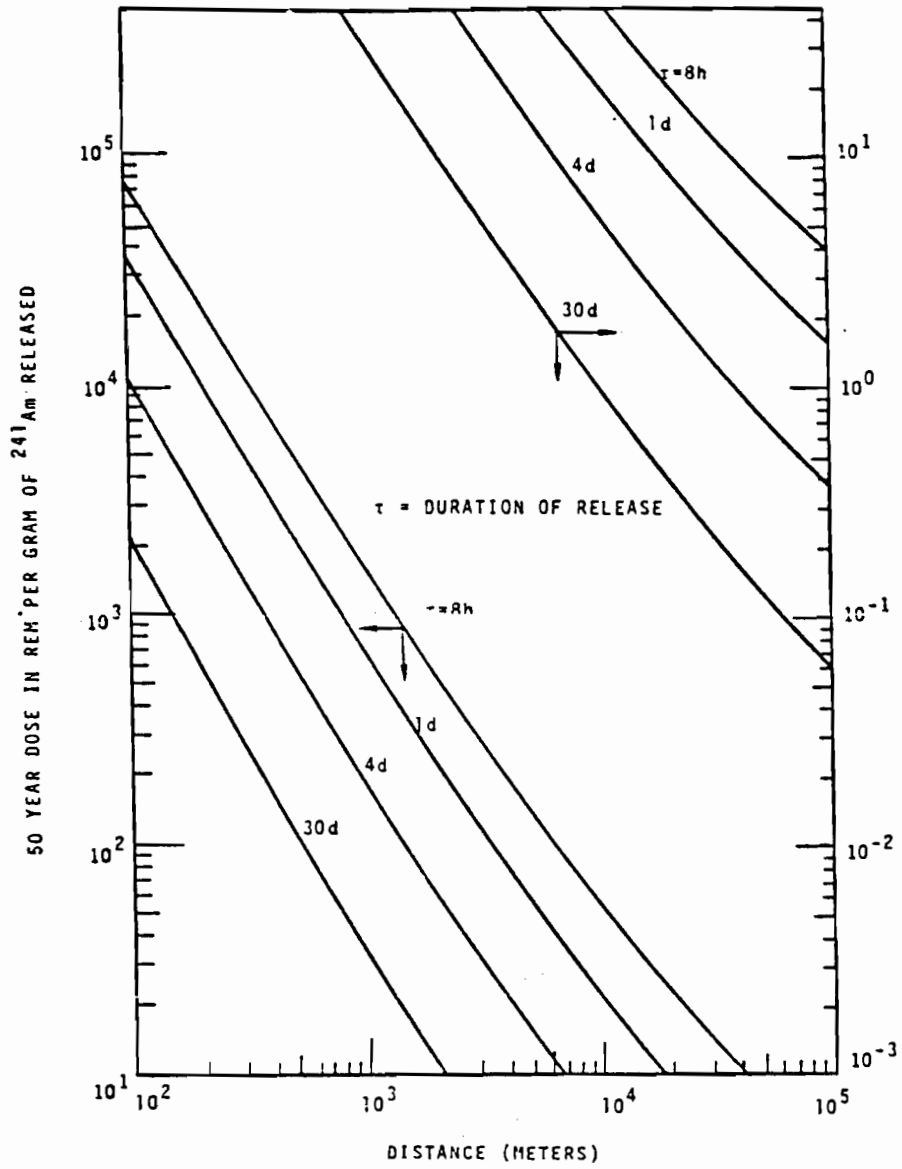


FIGURE B.4. Dose to Bone from Acute Inhalation of Plutonium-241, Ground Level Release



**FIGURE B.5.** Dose to Bone from Acute Inhalation of Americium-241, Ground Level Release

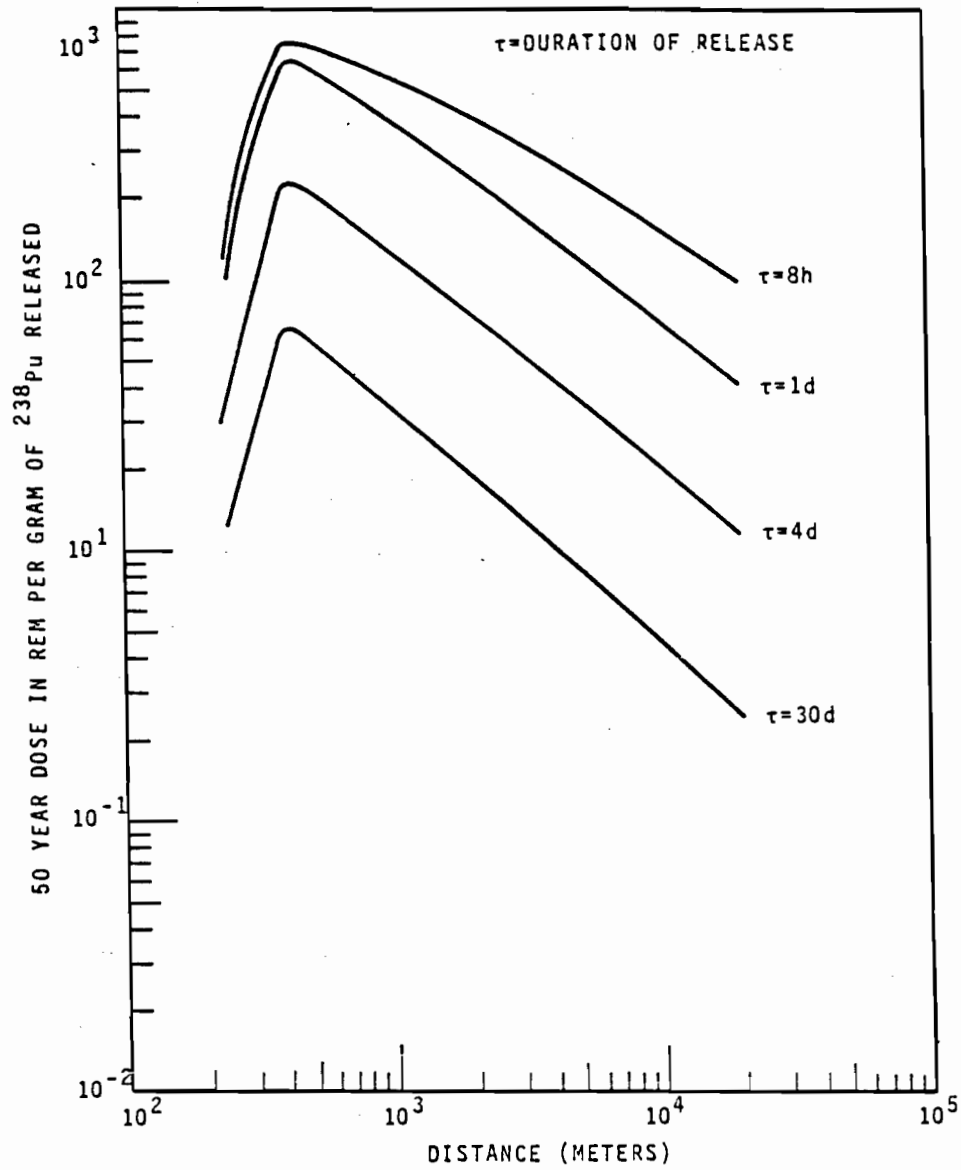
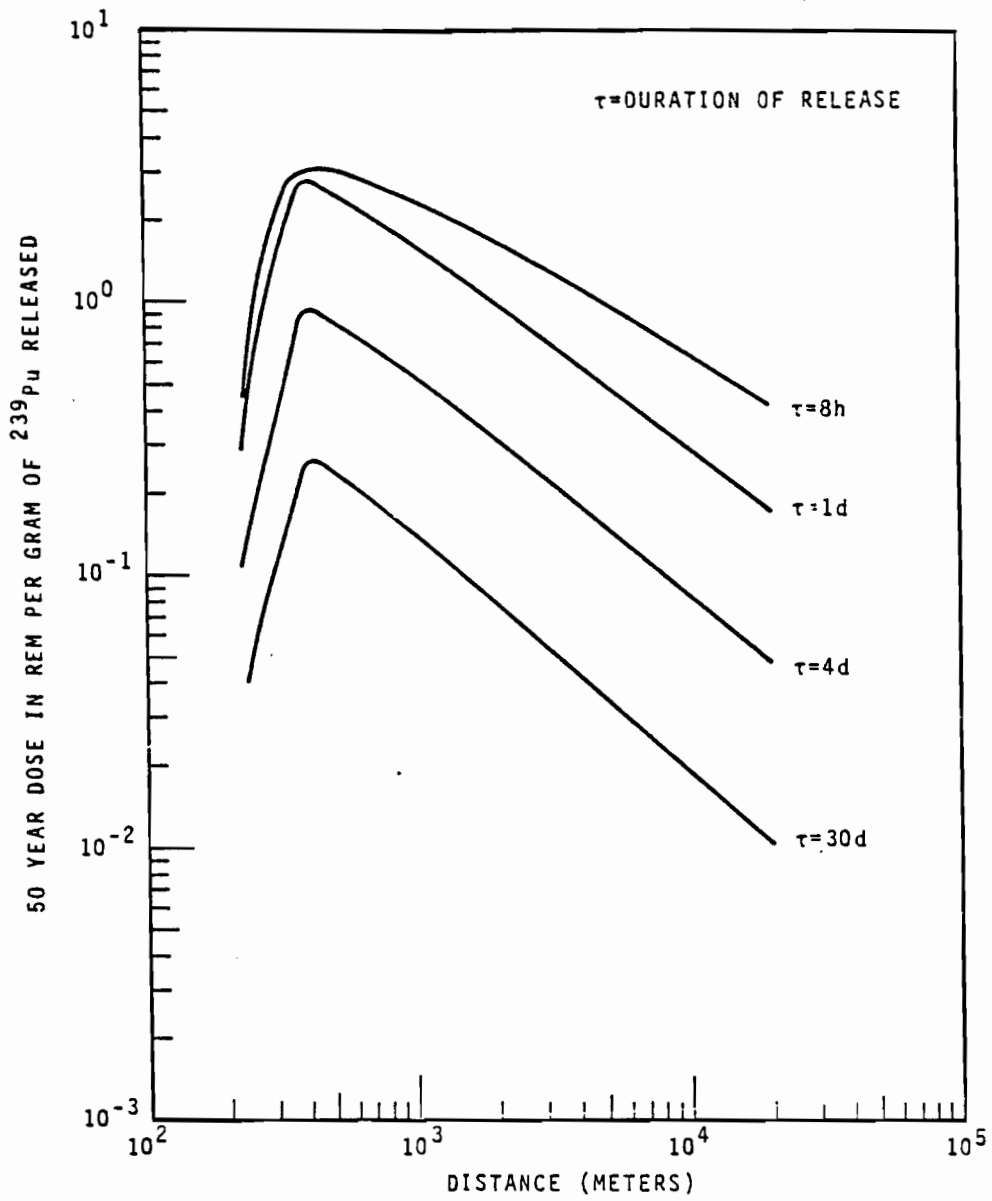
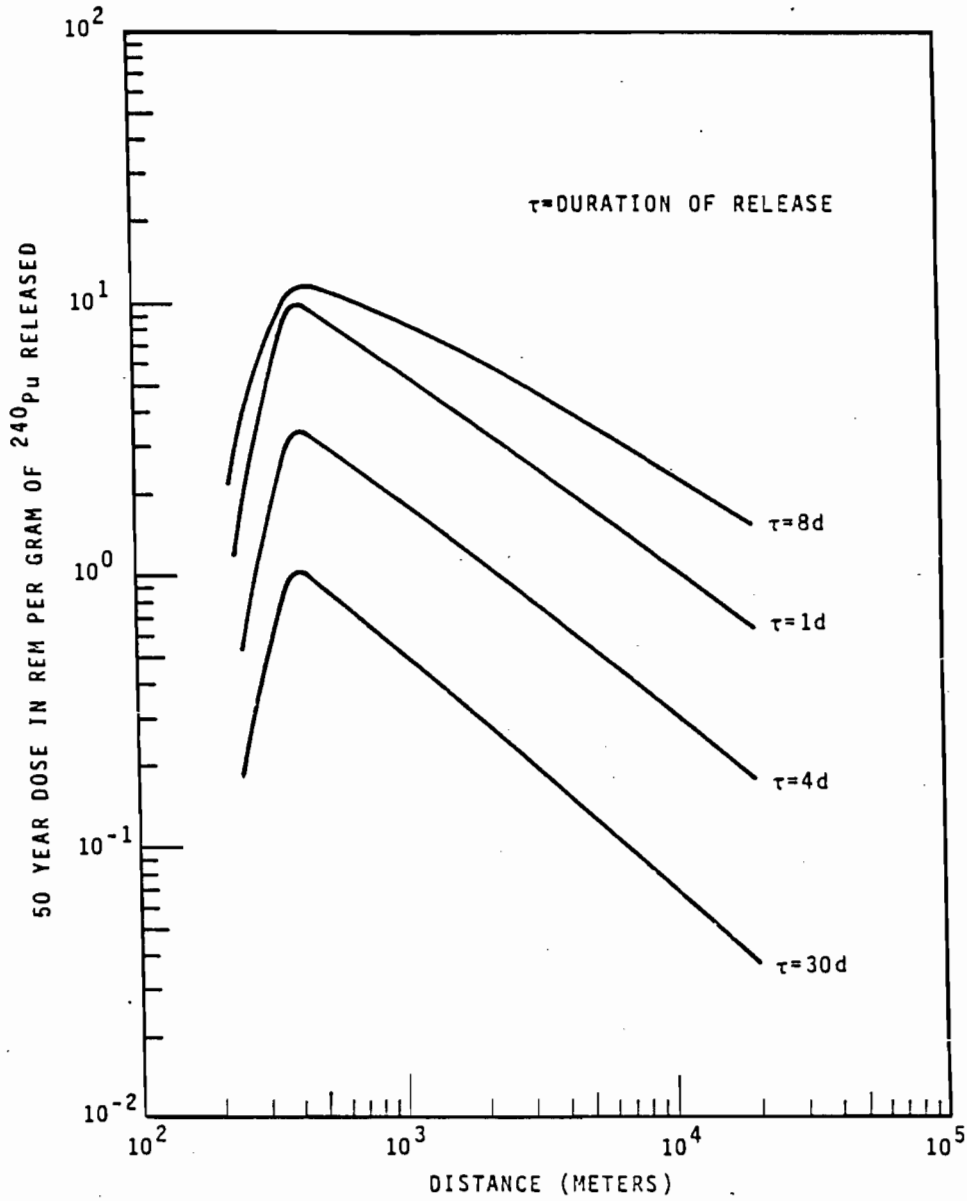


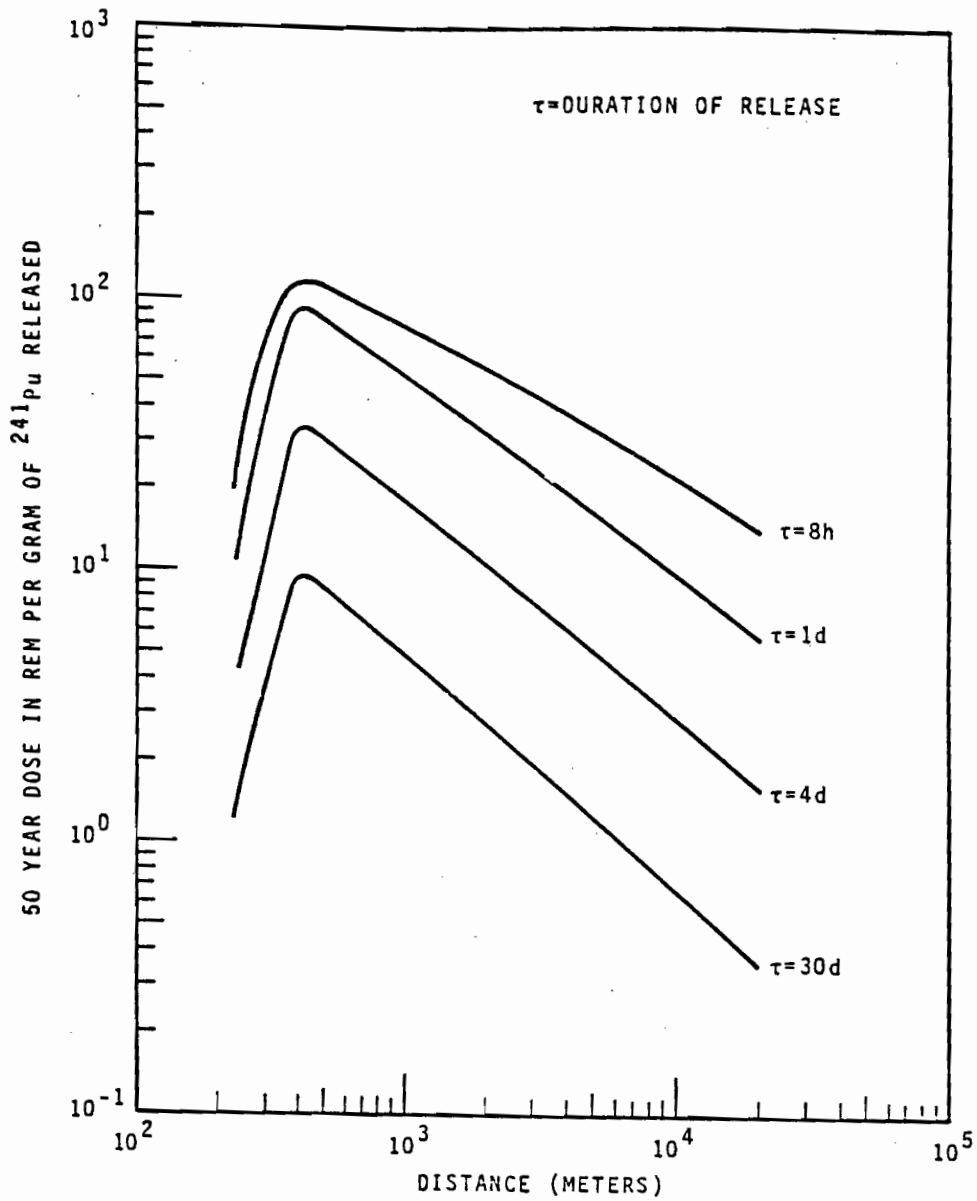
FIGURE B.6. Dose to Bone from Acute Inhalation of Plutonium-238, Release Height 100 Meters



**FIGURE B.7.** Dose to Bone from Acute Inhalation of Plutonium-239, Release Height 100 Meters



**FIGURE B.8.** Dose to Bone from Acute Inhalation of Plutonium-240, Release Height 100 Meters



**FIGURE B.9.** Dose to Bone from Acute Inhalation of Plutonium-241, Release Height 100 Meters

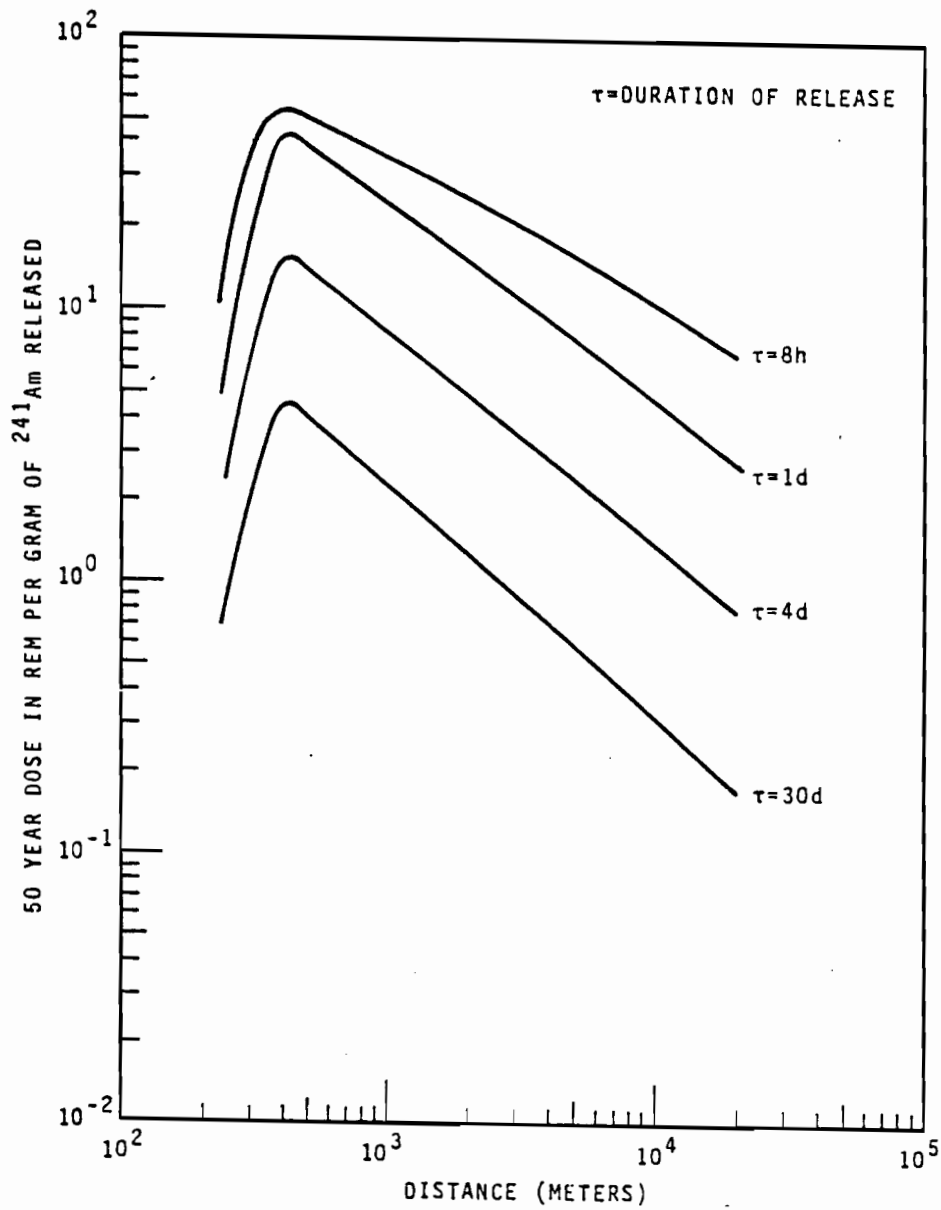


FIGURE B.10. Dose to Bone from Acute Inhalation of Americium-241, Release Height 100 Meters



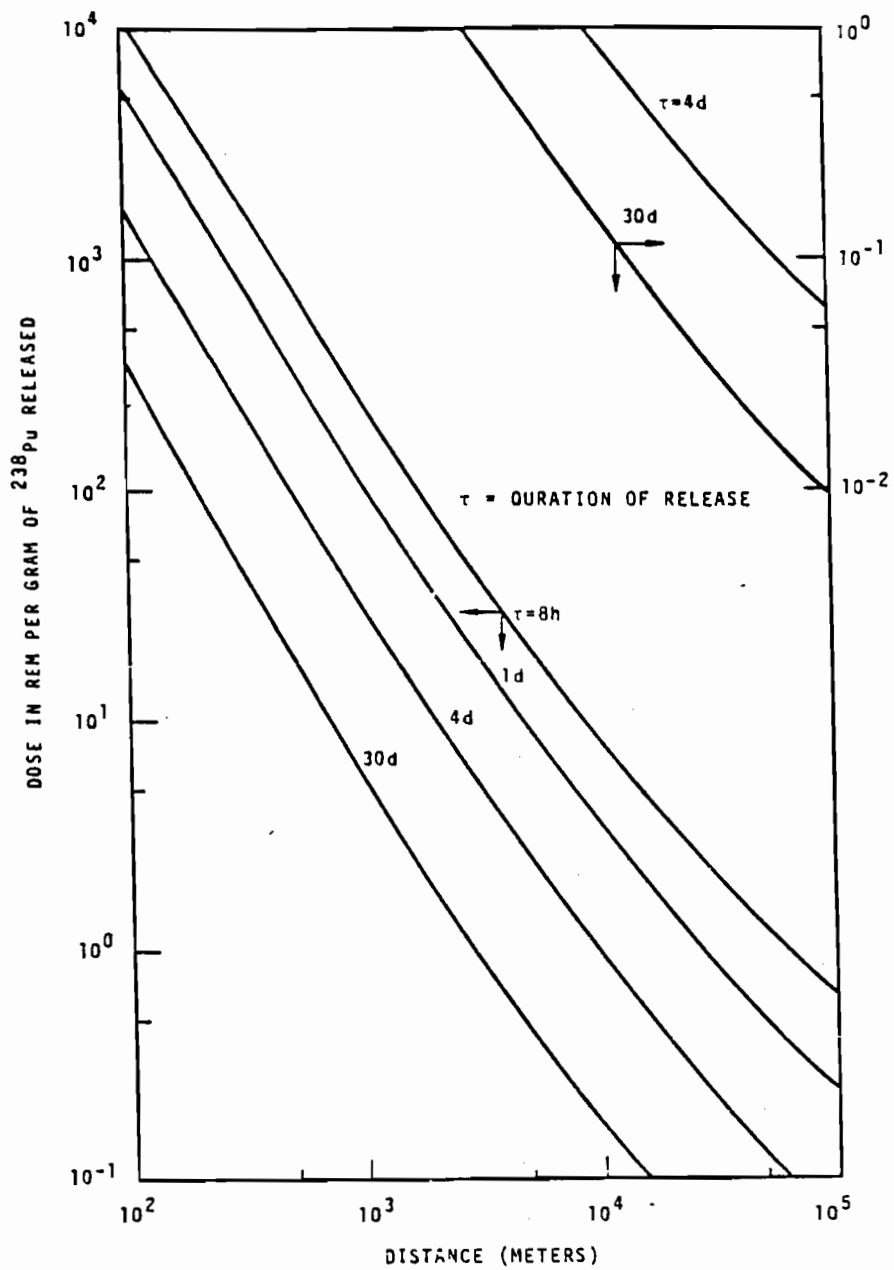


FIGURE B.11. Dose to Lung from Acute Inhalation of Plutonium-238, Ground Level Release

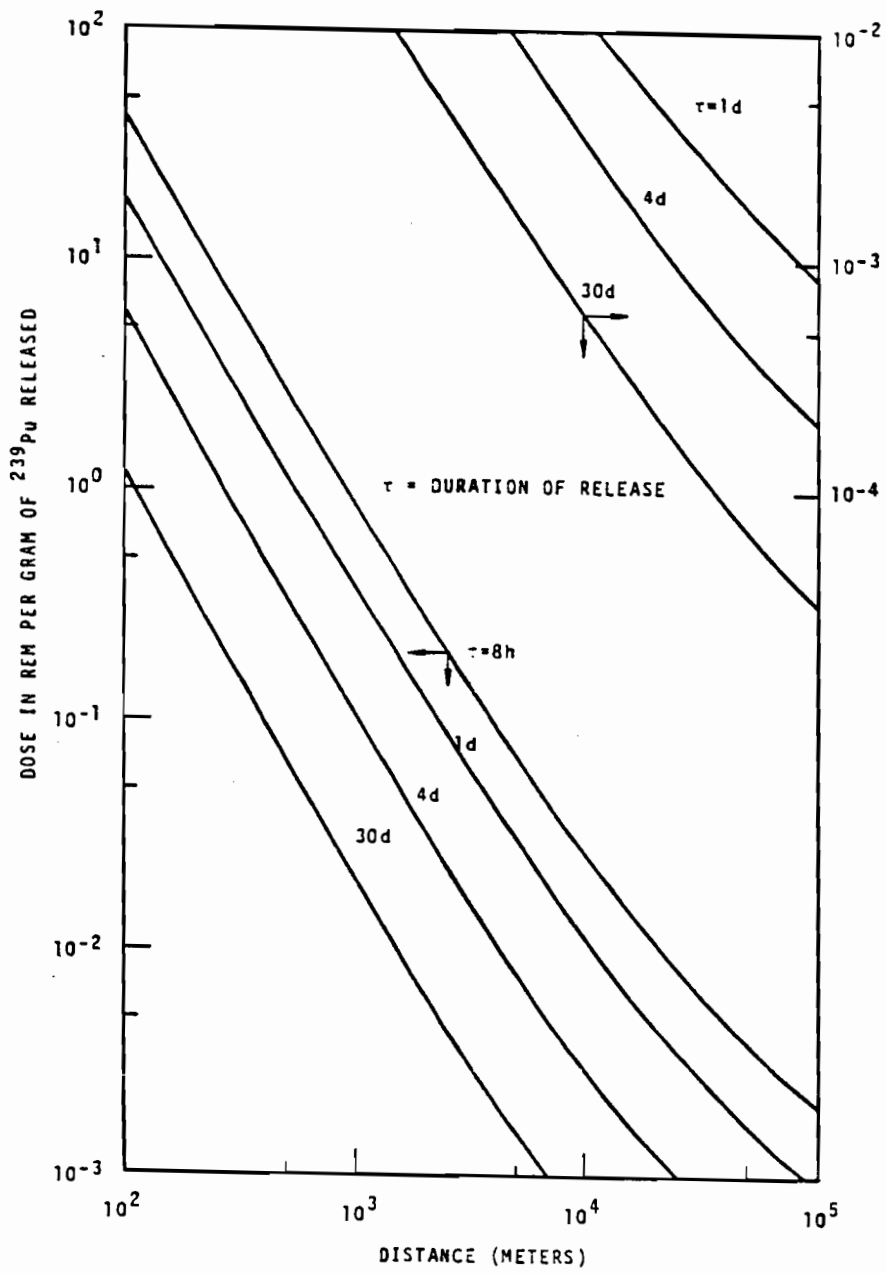


FIGURE B.12. Dose to Lung from Acute Inhalation of Plutonium-239, Ground Level Release

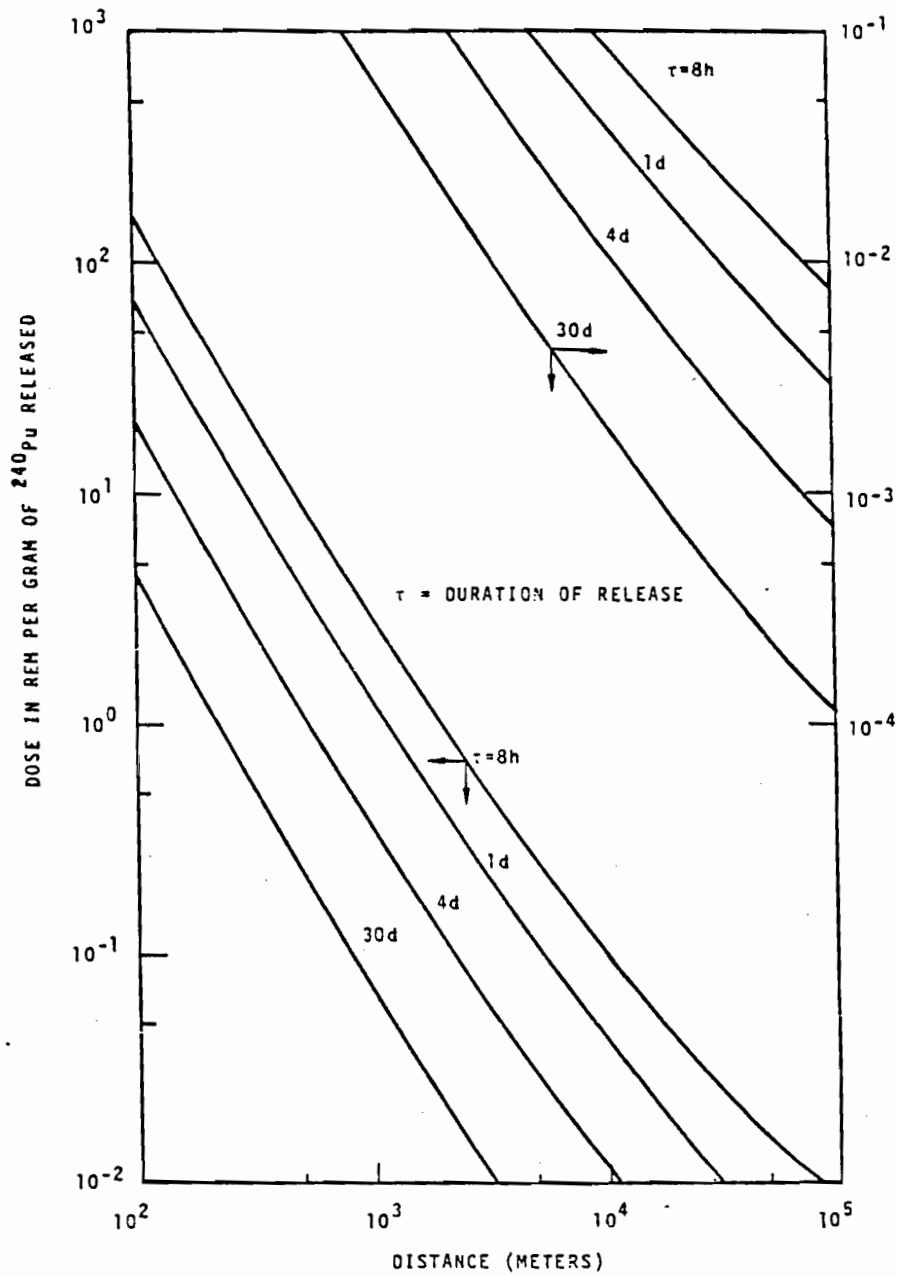
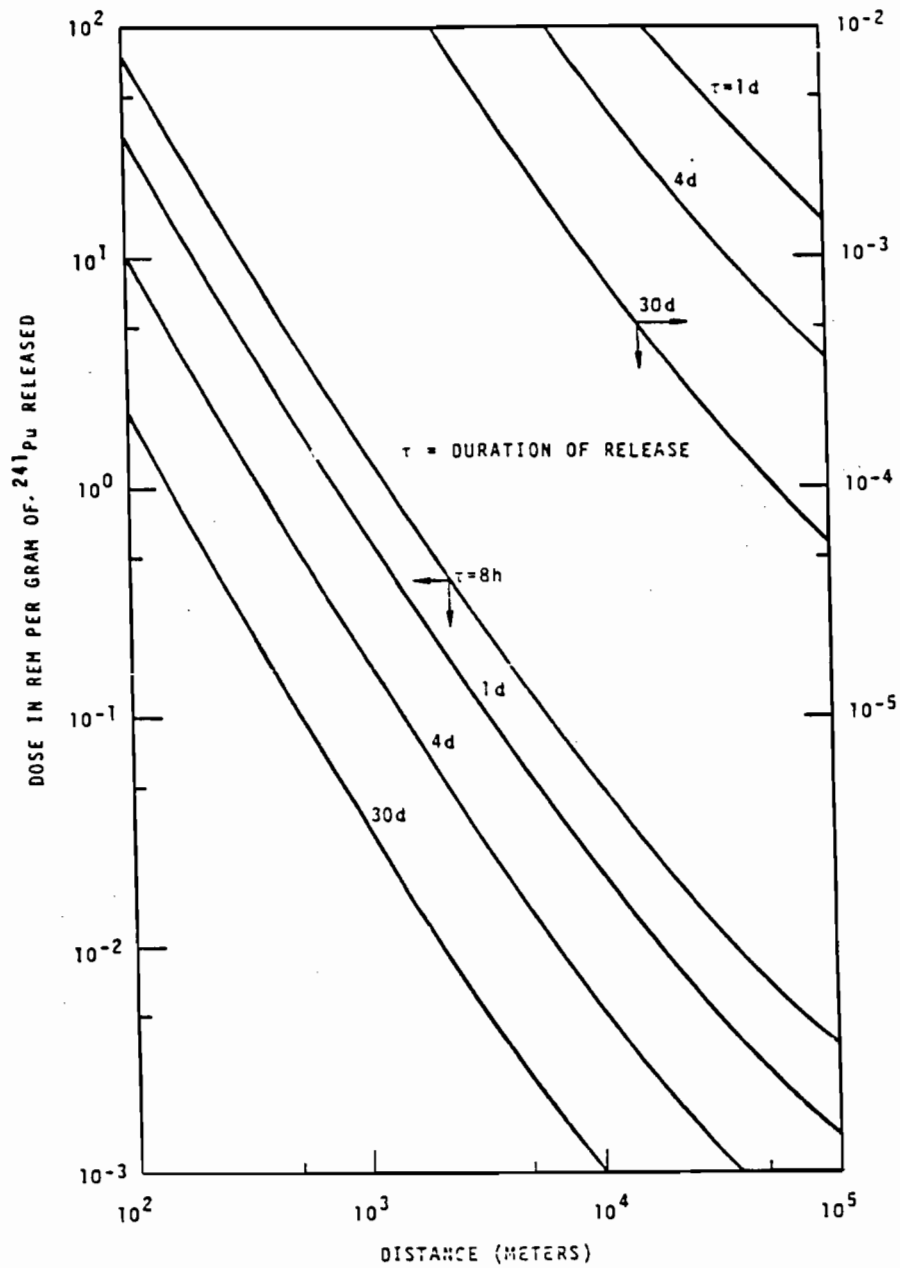


FIGURE B.13. Dose to Lung from Acute Inhalation of Plutonium-240, Ground Level Release



**FIGURE B.14.** Dose to Lung from Acute Inhalation of Plutonium-241, Ground Level Release

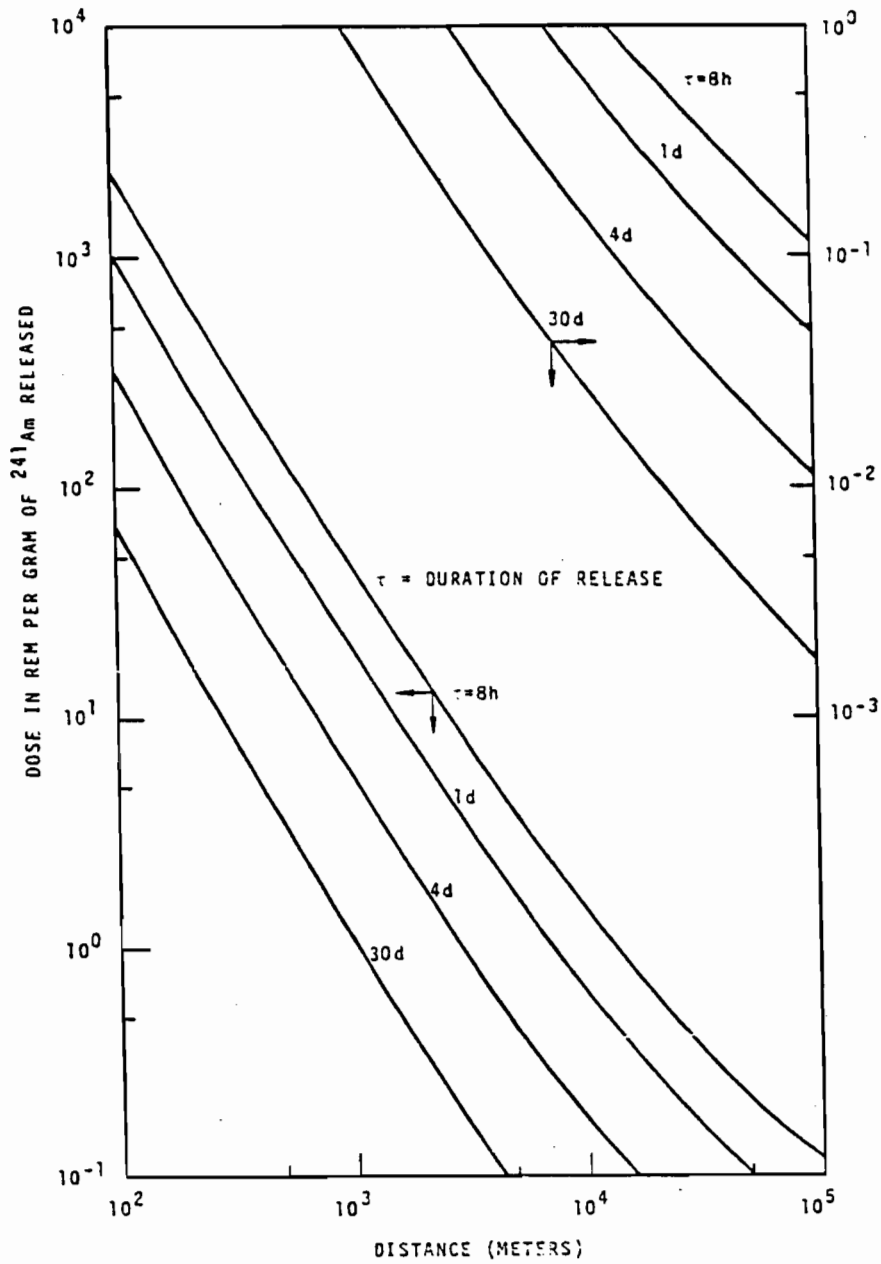


FIGURE B.15. Dose to Lung from Acute Inhalation of Americium-241, Ground Level Release

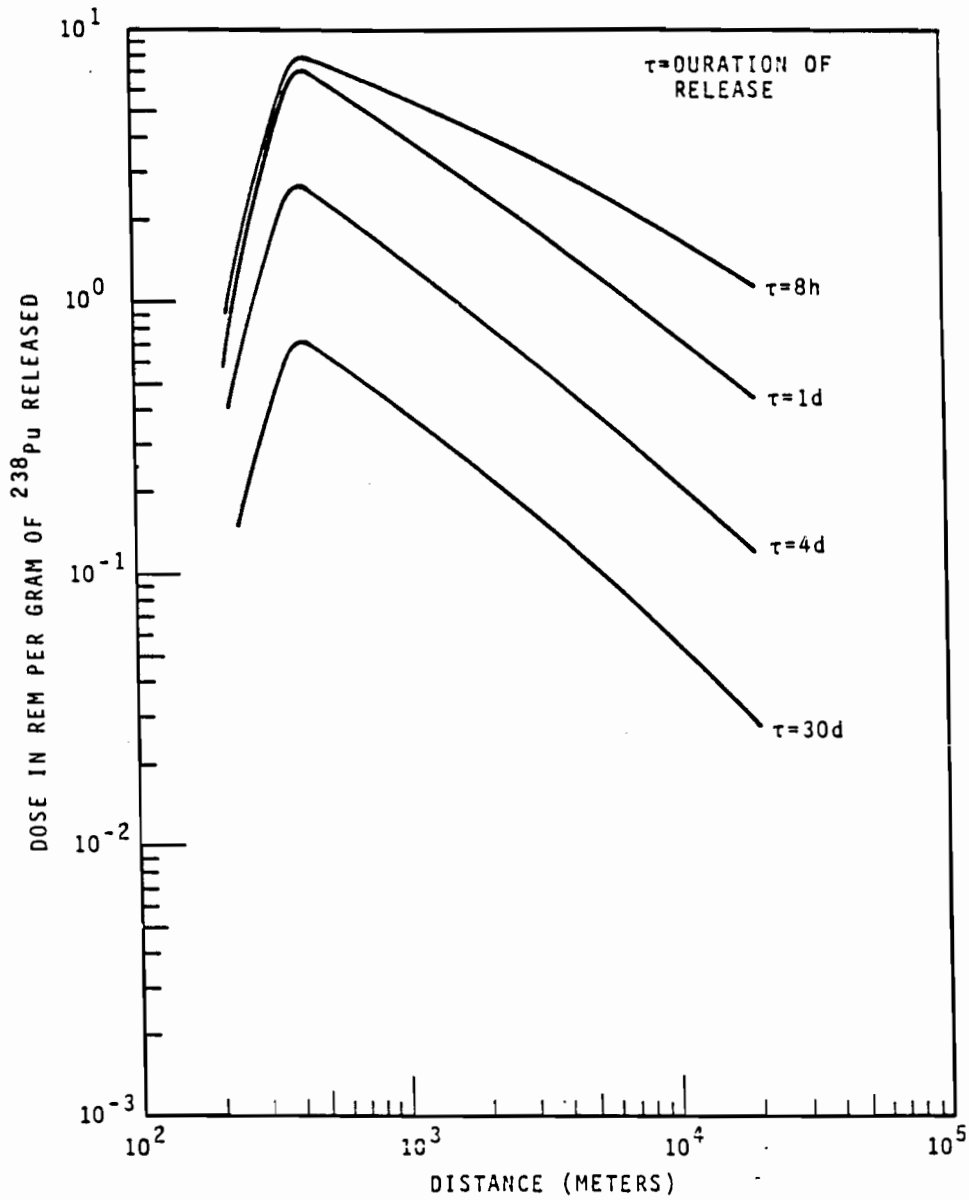


FIGURE B.16. Dose to Lung from Acute Inhalation of Plutonium-238, Release Height 100 Meters

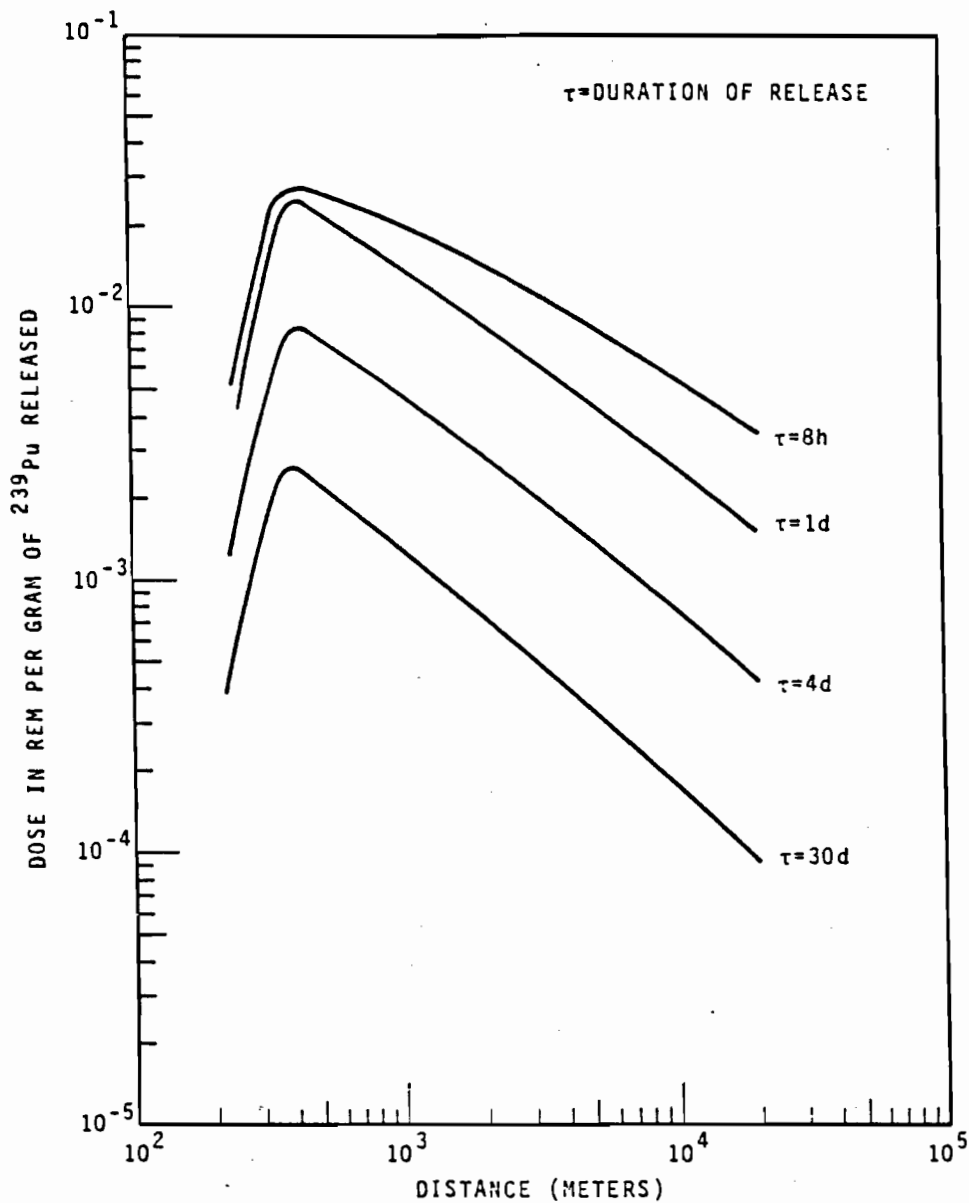
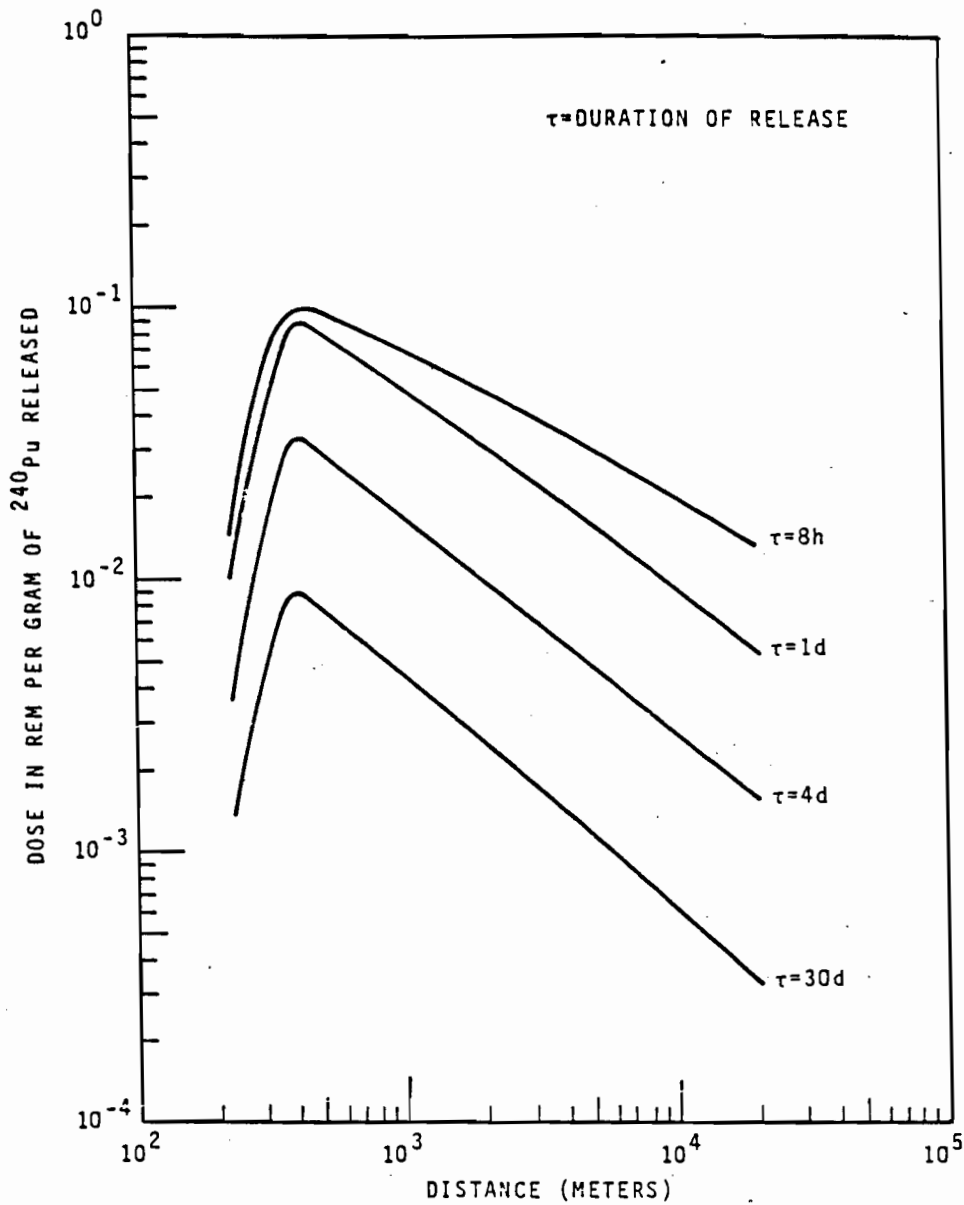


FIGURE B.17. Dose to Lung from Acute Inhalation of Plutonium-239, Release Height 100 Meters



**FIGURE B.18.** Dose to Lung from Acute Inhalation of Plutonium-240, Release Height 100 Meters



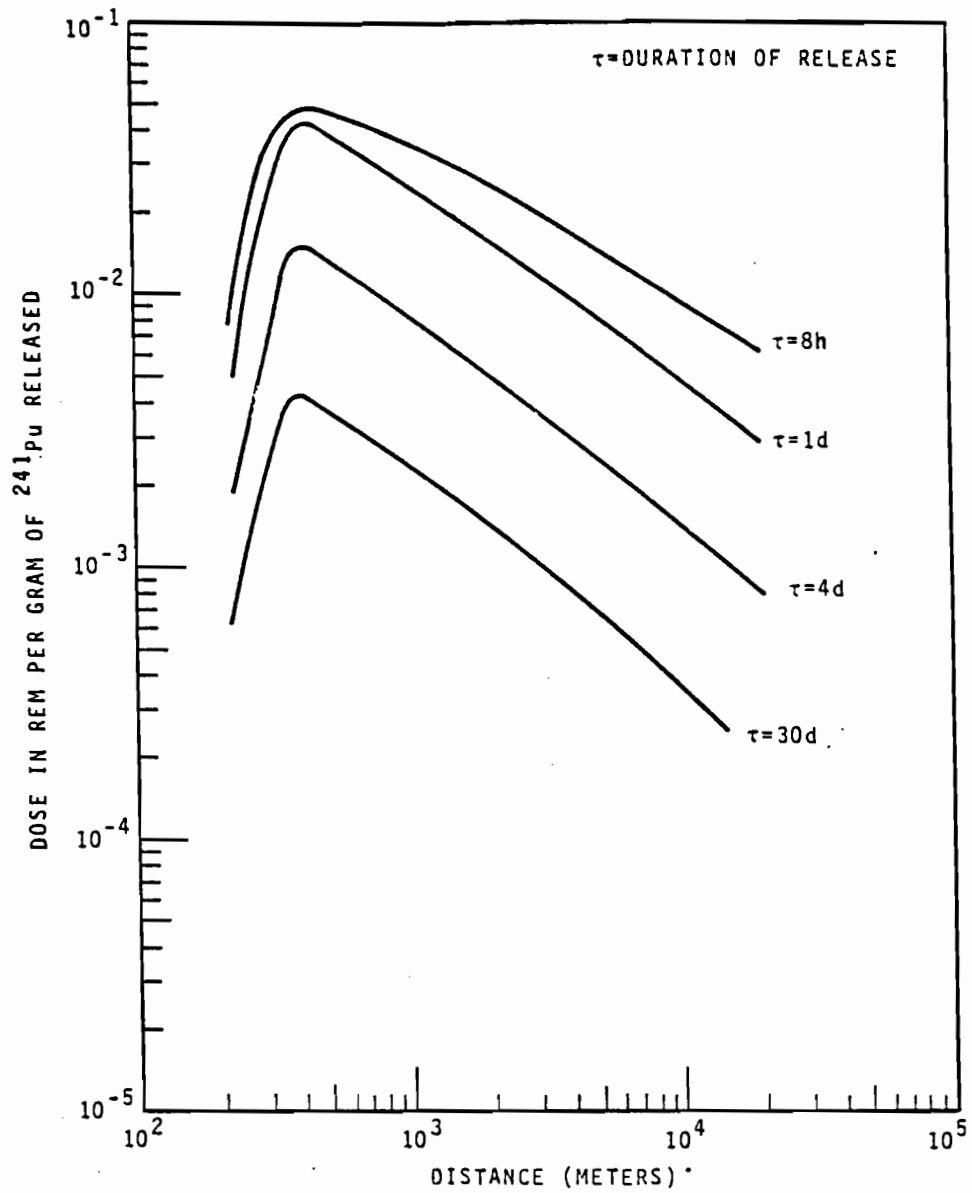


FIGURE B.19. Dose to Lung from Acute Inhalation of Plutonium-241, Release Height 100 Meters

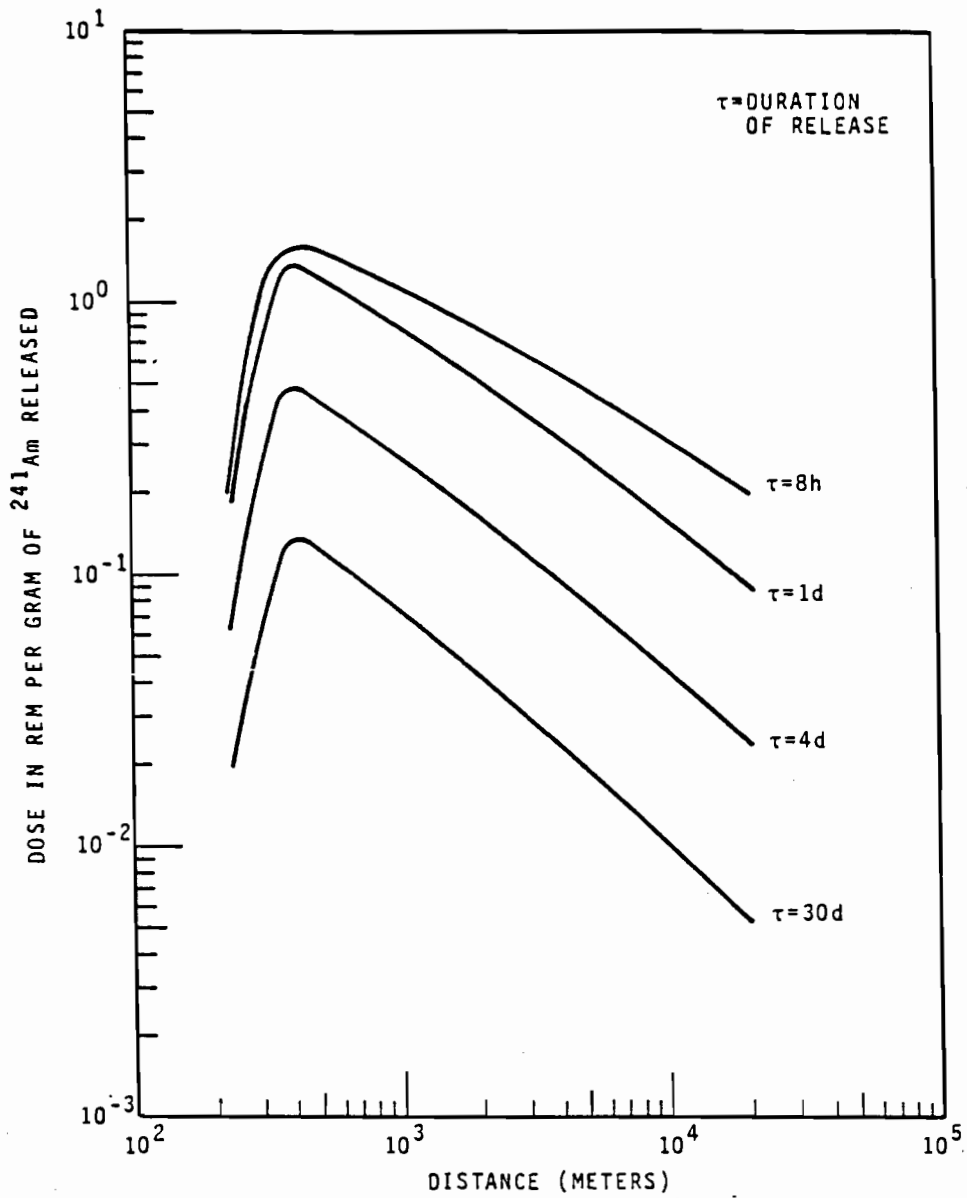


FIGURE B.20. Dose to Lung from Acute Inhalation of Americium-241, Release Height 100 Meters

APPENDIX C

PLUTONIUM ISOTOPIC COMPOSITIONS AND  
ASSOCIATED "SURFACE" DOSE RATES

## APPENDIX C

### PLUTONIUM ISOTOPIC COMPOSITIONS AND ASSOCIATED "SURFACE" DOSE RATES

#### PLUTONIUM ISOTOPIC COMPOSITIONS

The total quantities and the isotopic composition of the plutonium that the LWR industry will produce has been calculated. The calculation<sup>(1)</sup> includes the effects of varying exposure and of original enrichment as well as plutonium recycle so that the results are tabulated as "net plutonium available for recycle." The results are included in Tables C-1 through 3.

From the data presented in Table 2, the year 1980 was chosen as the "reference plutonium" because the  $^{238}\text{Pu}$  and  $^{241}\text{Pu}$  content of the plutonium would be approaching maximum concentrations. Also, the  $^{236}\text{Pu}$  appears to be at a maximum at that time but is still such a minor fraction of the total that it is negligible. The compositions given in Table 2 are the averages of the industry as known today and are subject to change. These changes could be brought about by planned plant startups, additions to or cancellations of existing plant orders, better fuel and/or mode of operation. In addition, since the isotopic compositions are average, the exact composition given in the table will probably never be encountered.

#### DOSES AND DOSE RATES

To determine the radiotoxicity of any isotopic composition of plutonium, one must know the time since last chemical separation and resultant in-growth of  $^{241}\text{Pu}$  daughters, namely  $^{237}\text{U}$  and  $^{241}\text{Am}$ . For the purposes of this study two years are assumed to have elapsed. Using the two-year age, the compositions for the years of 1975, 1980 and 1985, and bone as the reference organ, the one-year doses are given in Table C-4.

The photon "surface" dose rate from the same year's material and 2 years since chemical separation is given in Table C-5.

TABLE C-1. Plutonium Available for Fabrication  
(with self-produced plutonium recycle)

Year	Pu-236 Grams	Pu-238 Kg	Pu-239 Kg	Pu-240 Kg	Pu-241 Kg	Pu-242 Kg	Total Pu Kg	Total Fissile Pu Kg
1972	0.03	5.6	306	100	50	14	475.6	356
1973	0.04	6.9	442	137	62	16	663.9	504
1974	0.08	13.3	1,038	318	134	35	1,538.3	1,172
1975	0.13	23.4	1,719	571	238	70	2,621.4	1,957
1976	0.25	49.1	2,850	1,044	456	167	4,566.1	3,306
1977	0.47	91.3	5,170	1,927	841	299	8,328.3	6,011
1978	0.78	150.2	7,811	2,952	1,314	477	12,704.2	9,125
1979	1.11	219.1	9,440	3,778	1,756	692	15,885.1	11,196
1980	1.34	275.2	10,637	4,410	2,083	891	18,296.2	12,720
1981	1.62	349.5	13,414	5,624	2,665	1,198	23,250.5	16,079
1982	1.87	432.6	15,699	6,743	3,273	1,600	27,747.6	18,972
1983	2.14	525.4	18,217	8,033	3,949	2,065	32,789.4	22,166
1984	2.45	622.2	20,933	9,362	4,639	2,498	38,054.2	25,572
1985	2.8	716.8	23,767	10,714	5,314	2,899	43,410.8	29,081

TABLE C-2. Average Composition of Pu Available for Fabrication  
(weight fraction)

<u>Year</u>	<u>Pu-236</u>	<u>Pu-238</u>	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
1972	$6.3 \times 10^{-8}$	0.012	0.643	0.210	0.105	0.029
1973	$6.0 \times 10^{-8}$	0.010	0.666	0.206	0.093	0.024
1974	$5.2 \times 10^{-8}$	0.008	0.675	0.207	0.087	0.023
1975	$5.0 \times 10^{-8}$	0.009	0.656	0.218	0.090	0.027
1976	$5.5 \times 10^{-8}$	0.011	0.624	0.229	0.100	0.036
1977	$5.6 \times 10^{-8}$	0.011	0.621	0.231	0.101	0.036
1978	$6.1 \times 10^{-8}$	0.012	0.615	0.232	0.103	0.038
1979	$7.0 \times 10^{-8}$	0.014	0.594	0.238	0.110	0.044
1980	$7.3 \times 10^{-8}$	0.015	0.581	0.241	0.114	0.049
1981	$7.0 \times 10^{-8}$	0.015	0.577	0.242	0.115	0.051
1982	$6.7 \times 10^{-8}$	0.016	0.566	0.243	0.118	0.057
1983	$6.5 \times 10^{-8}$	0.016	0.555	0.245	0.120	0.063
1984	$6.4 \times 10^{-8}$	0.016	0.550	0.246	0.122	0.066
1985	$6.5 \times 10^{-8}$	0.016	0.547	0.247	0.122	0.067

TABLE C-3. Effect of Initial U-235 Enrichment and Fuel Exposure on Pu Composition

Reactor Type	Initial U-235 %	Exposure MWD/MT	Pu Composition (weight percent)						Fissile Pu, Kg/MT
			Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Pu-243	
PWR	2.75	10,000	0.002	0.794	0.145	0.054	0.005	3.90	
		15,000	0.003	0.720	0.183	0.081	0.012	4.90	
		20,000	0.006	0.657	0.212	0.130	0.022	5.57	
	3.00	10,000	0.002	0.805	0.137	0.051	0.004	3.86	
		15,000	0.003	0.733	0.175	0.078	0.010	4.88	
		20,000	0.005	0.672	0.204	0.100	0.019	5.58	
	3.25	10,000	0.002	0.815	0.131	0.048	0.004	3.81	
		15,000	0.003	0.745	0.168	0.075	0.009	4.86	
		20,000	0.005	0.685	0.196	0.096	0.018	5.60	
BWR	2.00	10,000	0.008	0.633	0.218	0.113	0.027	6.10	
		15,000	0.011	0.589	0.235	0.126	0.039	6.44	
		20,000	0.013	0.572	0.240	0.130	0.044	6.54	
	2.25	10,000	0.002	0.753	0.172	0.065	0.008	3.90	
		15,000	0.004	0.671	0.214	0.091	0.019	4.75	
		20,000	0.008	0.606	0.243	0.110	0.032	5.28	
	2.50	10,000	0.002	0.768	0.162	0.061	0.007	3.85	
		15,000	0.004	0.689	0.203	0.087	0.016	4.74	
		20,000	0.007	0.625	0.233	0.107	0.028	5.30	
2.75	10,000	0.011	0.572	0.254	0.121	0.042	5.67		
	15,000	0.014	0.561	0.254	0.125	0.046	5.89		
	20,000	0.014	0.561	0.254	0.125	0.046	5.89		

TABLE C-4. Total Doses, mrem/pCi Pu

<u>Year</u>	<u>Inhalation</u>	<u>Ingestion</u>	<u>Relative Hazard 1980=1 Inhalation</u>
1975	$1.96 \times 10^{-2}$	$4.84 \times 10^{-6}$	0.7
1980	$2.76 \times 10^{-2}$	$4.77 \times 10^{-6}$	1
1985	$2.9 \times 10^{-2}$	$7.12 \times 10^{-6}$	1.05

TABLE C-5. Photon Surface Dose Rate

<u>Year</u>	<u>Dose Rate, Rad/hr</u>
1975	8.1
1980	10.6
1985	11.3

The mixed oxide photon surface dose rate, natural uranium, is given in Figure C-1 as a function of PuO<sub>2</sub> enrichment. "Surface" indicates that the plutonium is contained or shielded by 50 mg/cm<sup>2</sup> thickness of plastic or some other low density material and the thickness of the source is  $\geq 1$  cm thick. The neutron dose rate is not nearly as straightforward.

To calculate the neutron dose rate of the reference material (as PuO<sub>2</sub>) a geometry and mass must be assumed. The geometry assumed corresponds to a standard 6M shipping container with an ID of about 13 cm. The mass of plutonium which can be shipped in a standard container is limited by a 10 watt heat generation rate,<sup>(2)</sup> which for the reference material is about 0.9 kg of PuO<sub>2</sub>. For a container with an  $\ell/D$  of 1 and a PuO<sub>2</sub> mass of 0.9 kg, the neutron dose rate at the surface and the mid-plane of the container will



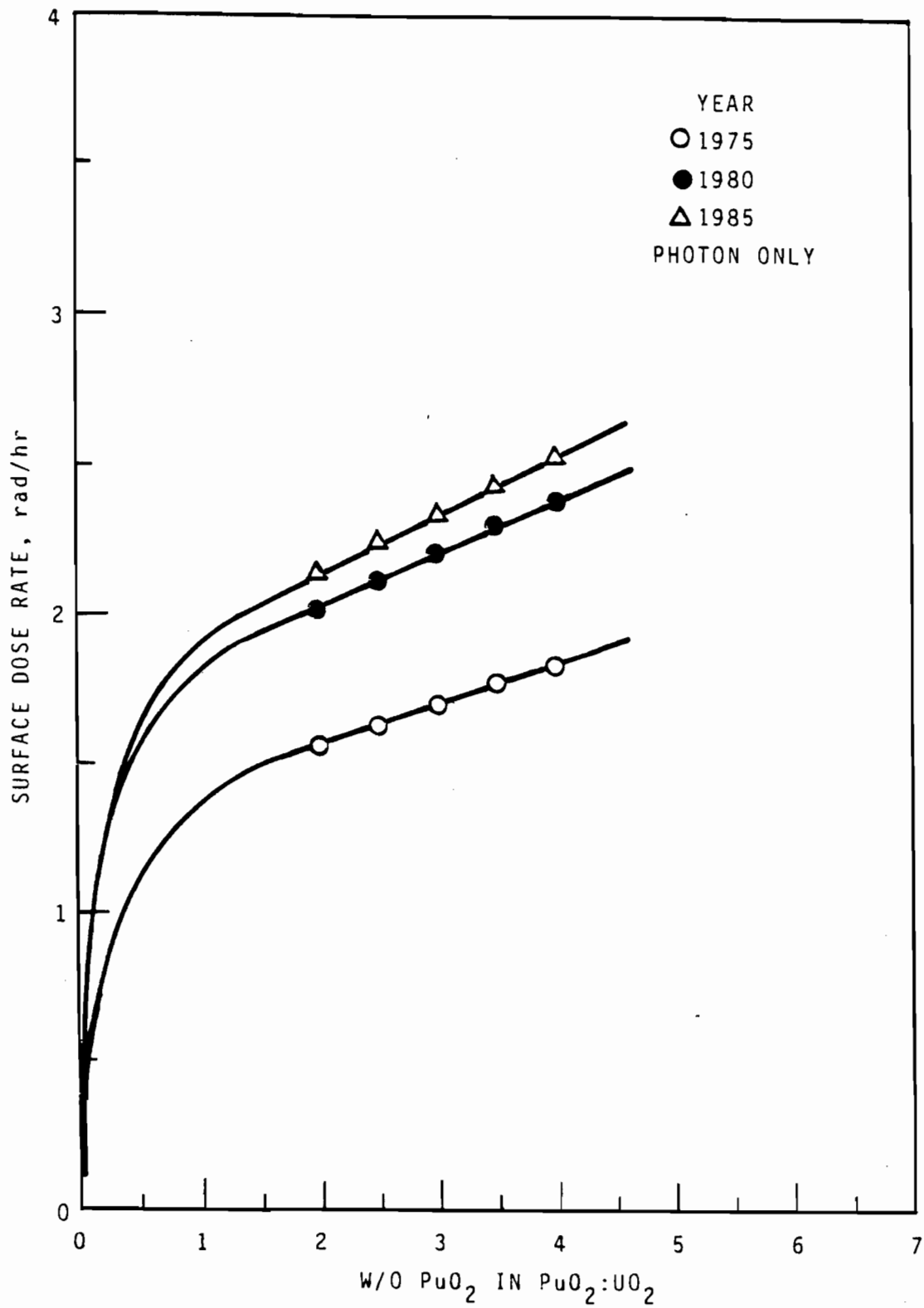


FIGURE C-1. Mixed Oxide Photon Surface Dose Rate

be about 450 mrem/hr. Table C-6 summarizes the total surface dose rates. The neutron contribution in the  $\text{MO}_2$  has not been evaluated because of uncertainties in masses, dimensions and densities of the  $\text{MO}_2$ .

TABLE C-6. Total Surface Dose Rates

<u>Year</u>	<u>Photon, rad/hr</u>	<u>Neutron, rem/hr</u>	<u>Total, rem/hr</u>
1975	8.1	0.34	8.4
1980	10.6	0.45	11.1
1985	11.3	0.49	11.8

REFERENCES

1. Deonigi, D. E., "Battelle Power Generation System, (BAPOGS) Data File, 9/1973.
2. 49CFR Transportation, Parts 100 to 199, Rev. 10/1/1973.

APPENDIX D  
DERIVATION OF THE CALCULATED ANNUAL  
MAXIMUM SECTOR CURVES

## APPENDIX D

### DERIVATION OF THE CALCULATED ANNUAL MAXIMUM SECTOR CURVES

#### INTRODUCTION

It is not generally economically practical to design nuclear facilities, including mixed oxide reprocessing plants, to operate without routine releases of radioactive gases or fine particulates to the atmosphere. Therefore, an estimate of downwind air concentrations is needed to evaluate the environmental impact of a proposed facility. In the initial stages of planning, the diffusion climatology may be unknown due to either lack of measurement at the exact site or lack of knowledge regarding the eventual location of the site. Hence, an estimate of the expected atmospheric dispersion from a "typical site" is needed.

The approach taken here makes use of available data from existing U.S. reactor sites. Maximum and average sector concentration curves as a function of distance derived from these data are representative of a variety of possible types of sites: river, lake, ocean and desert.

#### DATA

Twenty-six sites were used in the final analysis. Table D-1 contains the names of the sites along with additional information including the general type of location, height of wind measurements and the number of years of data. Other sites were eliminated for a variety of reasons, including incomplete data, questionable periods of data, and questionable instrument calibrations. Comments and recommendations on the appropriateness and accuracy of these data were given by the USAEC Division of Regulatory.

#### METHOD

The bivariate-normal model for atmospheric diffusion predicts the long-term sector average air concentration in a specified direction to be:

TABLE D-1. Reactor Sites Whose Micrometeorological Data Were Used

<u>Site</u>	<u>Location</u>	<u>Wind Speed Measurement Heights (ft.)</u>	<u>Number Years Of Data</u>
Arkansas	Rivershore	40	1
Arnold	Rivershore	35, 165	1
Bailly	Lakeshore	60, 150	1
Beaver Valley	Rivershore	50, 150	1
Browns Ferry	Rivershore	300	1.9
Brunswick	Seacoast	33, 350	1
Cook	Lakeshore	30, 200	1
Cooper	Rivershore	318	1
Davis-Besse	Lakeshore	100	1
Ginna	Lakeshore	50	2
Hanford 2	Rivershore	200	6.5
Hatch	Rivershore	75, 150	1
Humbolt Bay	Seacoast	250	2
Indian Point	Rivershore	100	1
Main Yankee	Seacoast	149	1
McGuire	Rivershore	131	1
Monticello	Rivershore	140	NA
Nine Mile Point	Lakeshore	31, 203	2
Oconee	Rivershore	150	2
Peach Bottom	Rivershore	30, 320	3
Pilgrim	Seacoast	300	1
Prairie Island	Rivershore	40	1
San Onofre	Seacoast	54	NA
Turkey Point	Seacoast	30	1
Watts Bar	Rivershore	30, 130	1
Zion	Lakeshore	35	1

$$\bar{x}_{x,h} = (2/\pi)^{1/2} Q' \sum_j \sum_i \frac{f_{i,j}}{(\sigma_z)_j (\bar{u}_h)_i w} \exp \left[ -h^2/2(\sigma_z)_j^2 \right] \quad (1)$$

where:

- $\bar{x}$  • long-term sector average air concentration at ground level in a specified direction,  $\mu\text{Ci}/\text{cm}^3$
- $Q'$  • average release rate from source, Ci/sec
- $f_{j,j}$  • fraction of time wind blows in sector during a specified stability  $j$ , at a given speed  $i$
- $(\sigma_z)_j$  • standard deviation of crosswind vertical dispersion of air concentration in cloud during stability condition  $j$ , m
- $(\bar{u}_h)_i$  • average wind speed in wind speed interval  $i$  at height of release, m/sec
- $h$  • height of release, m
- $w$  • arc width of sector, m. ( $w = 2\pi x/n$ )
- $x$  • downwind distance, m
- $n$  • number of sectors contained in 360 degrees (16 used in present study).

NOTE: Values for  $f_{i,j}$ ,  $Q'$ ,  $(\sigma_z)_j$ , and  $(\bar{u}_h)_i$  are long-term average values and the Pasquill-Gifford curves were used to evaluate the dispersion parameter  $\sigma_z$ .

The data for each site are first organized into stability class groupings (unstable, neutral, stable, very stable). Then for each stability class the persistences of the winds in each sector are given for individual wind speed classes. An illustrative example of this breakdown of data is shown in Table D-2 for one site. The values contained in

TABLE D-2. Wind Rose Frequency Percentage, Browns Ferry Nuclear Plant (February 11, 1967 to December 31, 1968)

Unstable Atmospheric Stability

Wind Direction	Wind Frequency (%) for Wind Speeds (mph)					
	0-3	4-7	8-12	13-18	19-24	>25
N	0	0.01	0.11	0.28	0	
NNE	0	0.07	0.18	0.11	0.01	
NE	0.01	0.04	0.12	0.01	0	
ENE	0.01	0.02	0.03	0.01	0.01	
E	0	0.03	0.04	0	0	
ESE	0	0.03	0.08	0.02	0.02	
SE	0.01	0.30	0.46	0.20	0.06	0.01
SSE	0.02	0.30	0.34	0.22	0.09	0.01
S	0.02	0.18	0.18	0.09	0.01	
SSW	0.01	0.09	0.15	0.15	0.06	
SW	0.01	0.07	0.09	0.13	0.03	
WSW	0.02	0.16	0.28	0.21	0.04	
W	0.02	0.17	0.25	0.24	0.13	0.02
WNW	0.01	0.21	0.58	0.47	0.16	0.08
NW	0.02	0.06	0.19	0.14	0.14	
NNW	0.01		0.14	0.19	0.07	0.01

Maximum Sector Value

Neutral Atmospheric Stability

Wind Direction	Wind Frequency (%) for Wind Speeds (mph)					
	0-3	4-7	8-12	13-18	19-24	>25
N	0.08	0.47	0.88	0.81	0.32	0.07
NNE	0.18	0.98	1.17	0.81	0.16	0.01
NE	0.31	1.29	0.66	0.06	0	0
ENE	0.16	0.55	0.51	0.11	0.01	0
E	0.11	0.45	0.43	0.23	0.02	0.02
ESE	0.15	0.94	1.23	0.83	0.25	0.05
SE	0.18	0.97	1.23	0.88	0.56	0.24
SSE	0.22	1.20	1.11	0.82	0.37	0.14
S	0.22	0.92	0.84	0.57	0.23	0.11
SSW	0.18	0.93	0.77	0.65	0.32	0.10
SW	0.22	0.88	0.76	0.61	0.32	0.05
WSW	0.22	1.02	0.83	0.80	0.27	0.04
W	0.19	0.99	0.82	0.56	0.17	0.09
WNW	0.14	0.98	1.12	0.80	0.44	0.19
NW	0.20	0.47	0.85	0.99	0.79	0.15
NNW	0.14	0.34	0.86	1.16	0.47	0.07

TABLE D-2. (Continued)

Moderately Stable Atmospheric Stability

Wind Direction	Wind Frequency (%) for Wind Speeds (mph)					
	0-3	4-7	8-12	13-18	19-24	>25
N	0.06	0.14	0.48	0.63	0.16	0
NNE	0.11	0.42	0.88	0.43	0.15	0
NE	0.14	0.84	0.66	0.09	0	0.01
ENE	0.12	0.25	0.34	0.30	0.06	0.01
E	0.07	0.23	0.40	0.25	0.04	0
ESE	0.11	0.38	0.61	0.68	0.14	0
SE	0.13	0.44	0.94	0.89	0.36	0.07
SSE	0.14	0.51	1.04	1.23	0.36	0.32
S	0.17	0.44	0.84	0.86	0.24	0.06
SSW	0.09	0.42	0.64	0.66	0.32	0.06
SW	0.12	0.44	0.63	0.68	0.16	0.05
WSW	0.12	0.37	0.55	0.44	0.07	0.01
W	0.11	0.35	0.50	0.26	0.05	0.03
WNW	0.18	0.31	0.45	0.21	0.04	0.01
NW	0.11	0.30	0.30	0.39	0.10	0.01
NNW	0.14	0.23	0.40	0.60	0.12	0

Very Stable Atmospheric Stability

Wind Direction	Wind Frequency (%) for Wind Speeds (mph)					
	0-3	4-7	8-12	13-18	19-24	>25
N	0.07	0.06	0.17	0.30	0.11	0.01
NNE	0.06	0.27	0.34	0.30	0.04	0
NE	0.14	0.59	0.51	0.14	0	0
ENE	0.08	0.20	0.34	0.48	0.11	0.02
E	0.07	0.20	0.40	0.21	0.14	0
ESE	0.17	0.47	0.65	0.36	0.01	0
SE	0.07	0.48	0.96	0.63	0.07	0
SSE	0.06	0.43	0.44	0.48	0.03	0
S	0.09	0.21	0.46	0.38	0.05	0
SSW	0.06	0.24	0.36	0.29	0.06	0
SW	0.08	0.14	0.20	0.13	0.03	0
WSW	0.04	0.07	0.15	0.06	0.01	0
W	0.04	0.14	0.06	0.04	0.01	0
WNW	0.13	0.09	0.06	0.04	0.01	0
NW	0.12	0.11	0.05	0.02	0.01	0
NNW	0.07	0.08	0.11	0.12	0.05	0



this table for persistence correspond to the values of  $f$  in Equation 1. An average sector concentration is calculated by summing over all these organized persistences in a sector as expressed by Equation 1.

Hence, the application of Equation 1 to a particular site requires the joint windspeed and direction frequencies for each stability class, based on a sufficiently long period so as to be representative of the diffusion climatology at the site. The wind speed frequency in each stability category for each direction is used to partition the total source in a functional manner, i.e., if the wind blows 10% of the time to the north from the point of release, then 10% of the releases are assumed to travel to the north. The average annual concentration for a given sector is proportional to the persistence in the sector; the greater the fraction, the greater the fraction of the total annual release that is assumed to be in this sector.

The available joint frequency data was inconsistent in another respect, i.e., the wind speed and direction measurements were not all taken at the same height above ground. Additionally, these measurements were not always taken at the height of potential release. Thus, a correction was made for release height by the following relationship:

$$\bar{u}_h = \left(\frac{h}{z}\right)^p \bar{u}_m \quad (2)$$

where:

- $\bar{u}_h$  • average wind speed at assumed height of release, m/sec
- $h$  • height of assumed release, m
- $z$  • height of wind speed and direction measurements, m
- $\bar{u}_m$  • mid-value of standard wind speed interval, m/sec
- $p$  • 0.25 for neutral and unstable conditions
- $p$  • 0.5 for stable conditions.

For a ground level release,  $h$  was assumed to be zero in Equation (1) and 10m in Equation (2).

The "average sector" was derived by averaging the overland sector frequencies within each stability category for each wind speed interval. These average frequencies were then used in Equation (1) to calculate the "average sector" annual air concentration.

#### MAXIMIZING PROCEDURE

A computational maximizing procedure was adopted for the  $(\bar{x}/Q')$  calculation at each site.

A calculation where the final value is the summation of a number of contributions may be maximized by selecting the largest values of each of the contributions. In the  $(\bar{x}/Q')$  calculation outlined above, the average annual  $(\bar{x}/Q')$  contribution for each component of the groupings in Table D-2 will depend directly on the persistence values (f). The estimates of a maximum annual average air concentration for a hypothetical sector were obtained by electing to use the largest values of the persistences that occurred in each wind speed class within each stability category disregarding direction. Hence, the actual procedure was to obtain a single set of maximum values for the wind speed groups, for each of the stability classes. That is, the maximum persistence value was taken for the 0 to 3 mph wind grouping, and then for the 4 to 7 mph, etc., for each stability category. This set of values represent the maximum persistence that occurred in each wind category regardless of wind direction, based on the site's data. The values used in the example given in Table D-2 have been circled. This procedure will maximize the calculated sector average annual air concentration by using the maximum persistences that occurred within the groupings inherent in the calculation.

Additional operations were necessary, depending on the form in which data were received. Not all data sets had the same wind speed class intervals. These data were converted to standard intervals by graphical means. Calm periods were included in the lowest wind speed class and were assumed to have the same distribution. For average sector values, the calm periods were equally divided between the sectors. For the maximum sector, a suitably higher percentage of the calm was used. Stability categories

were judged to be Pasquill-Class B for unstable, D for neutral, E for stable, and F for very stable. The values of  $\sigma_z$  that were used were based on values given by Pasquill and Gifford.

#### CALCULATED ANNUAL MAXIMUM SECTOR CURVES

The maximizing procedure was used to calculate a set of maximum sector curves as a function of distance for each of the twenty-six sites for a number of heights of release. Then for each release height, the maximum ( $\bar{x}/Q'$ ) values were taken at each of ten calculational distances to produce an upper limit curve based on all the sites. The set of curves for the different release heights is referred to as the CAMS (Calculated Annual Maximum Sector) curves. They are presented in Table D-3.

The onsite meteorological data from the reactor sites were also used to calculate mean values of sector average annual air concentrations. In this instance, the frequency distributions within each stability class and for each wind speed for all overland sectors were summed and then divided by the number of sectors. This result is a mean frequency distribution for a 22.5° sector. The mean meteorological condition for all of the sectors around a facility is represented by the mean sector value. Normalized average air concentrations were then calculated as a function of distance. The averages of these mean curves for all sites are referred to as the mean sector curves. A comparison is given in Figures D-1 and D-2 between the mean curves and CAMS as a function of distance for all sites for 10-meter and 100-meter, releases, respectively.

The CAMS curves were developed to be used as initial estimates of the annual average  $\bar{x}/Q'$  values that might occur at any site. Comparisons with independently calculated  $\bar{x}/Q'$  values have shown that the CAMS curves may be used as realistic but conservative estimates of  $\bar{x}/Q'$  in the absence of actual site climatological data.

Although the most conservative sector (CAMS) has been used in this study, credit should be given for local site characteristics in specific

TABLE 2. "Maximum Sector" Average Annual  
Normalized Air Concentration

( $\bar{x}/Q'$ ) as a function of distance

Distance (m)	Height (meters)							
	0	10	30	50	75	100	125	150
$1 \times 10^2$	2.86-3*	6.23-5	5.51-7	1.62-10	2.40-17	<<1-20	--	--
$2 \times 10^2$	7.07-4	1.07-4	4.11-6	5.10-7	1.02-8	4.58-11	4.56-14	7.87-18
$5 \times 10^2$	1.20-4	7.59-5	3.92-6	1.11-6	5.80-7	2.76-7	1.10-7	3.62-8
$1 \times 10^3$	3.49-5	2.97-5	3.34-6	9.26-7	3.48-7	2.10-7	1.68-7	1.32-7
$2 \times 10^3$	1.10-5	1.03-5	2.12-6	7.68-7	2.64-7	1.23-7	6.32-8	4.05-8
$5 \times 10^3$	2.56-6	2.50-6	7.48-7	3.92-7	1.85-7	9.99-8	5.62-8	3.33-8
$1 \times 10^4$	9.38-7	9.26-7	3.06-7	1.89-7	1.04-7	6.17-8	3.98-8	2.67-8
$2 \times 10^4$	3.66-7	3.63-7	1.26-7	8.47-8	5.30-8	3.31-8	2.24-8	1.60-8
$5 \times 10^4$	1.11-7	1.10-7	3.94-8	2.81-8	1.95-8	1.37-8	9.55-9	6.77-9
$1 \times 10^5$	4.59-8	4.58-8	1.66-8	1.21-8	8.81-9	6.53-9	4.83-9	3.57-9
$2 \times 10^5$	1.80-8	1.79-8	6.71-9	4.99-9	3.76-9	2.91-9	2.27-9	1.77-9
$5 \times 10^5$	5.46-9	5.46-9	2.07-9	1.57-9	1.22-9	9.86-10	8.08-10	6.65-10
$1 \times 10^6$	2.23-9	2.23-9	8.54-10	6.52-10	5.15-10	4.25-10	3.57-10	3.03-10

\*2.86-3 =  $2.86 \times 10^{-3}$

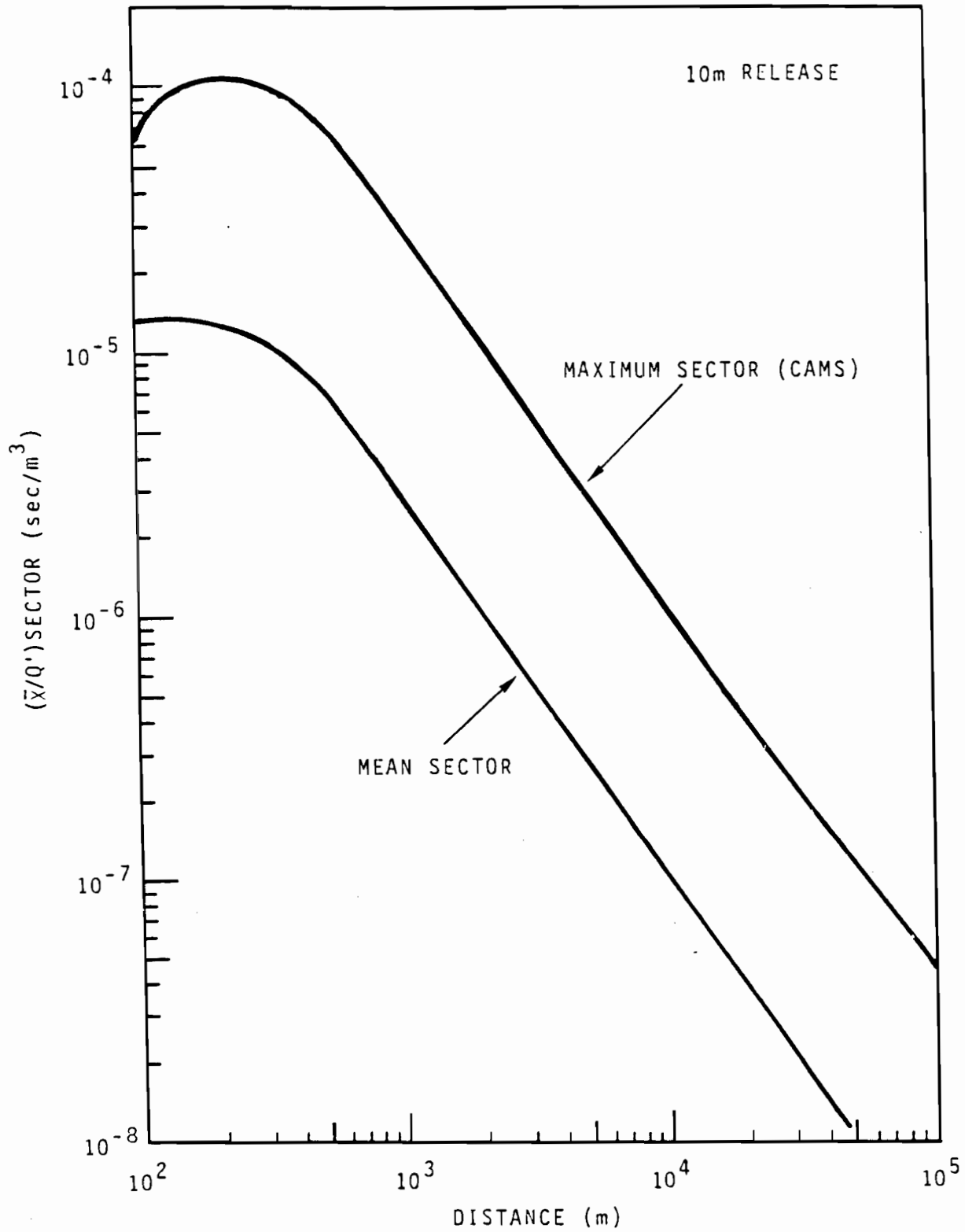
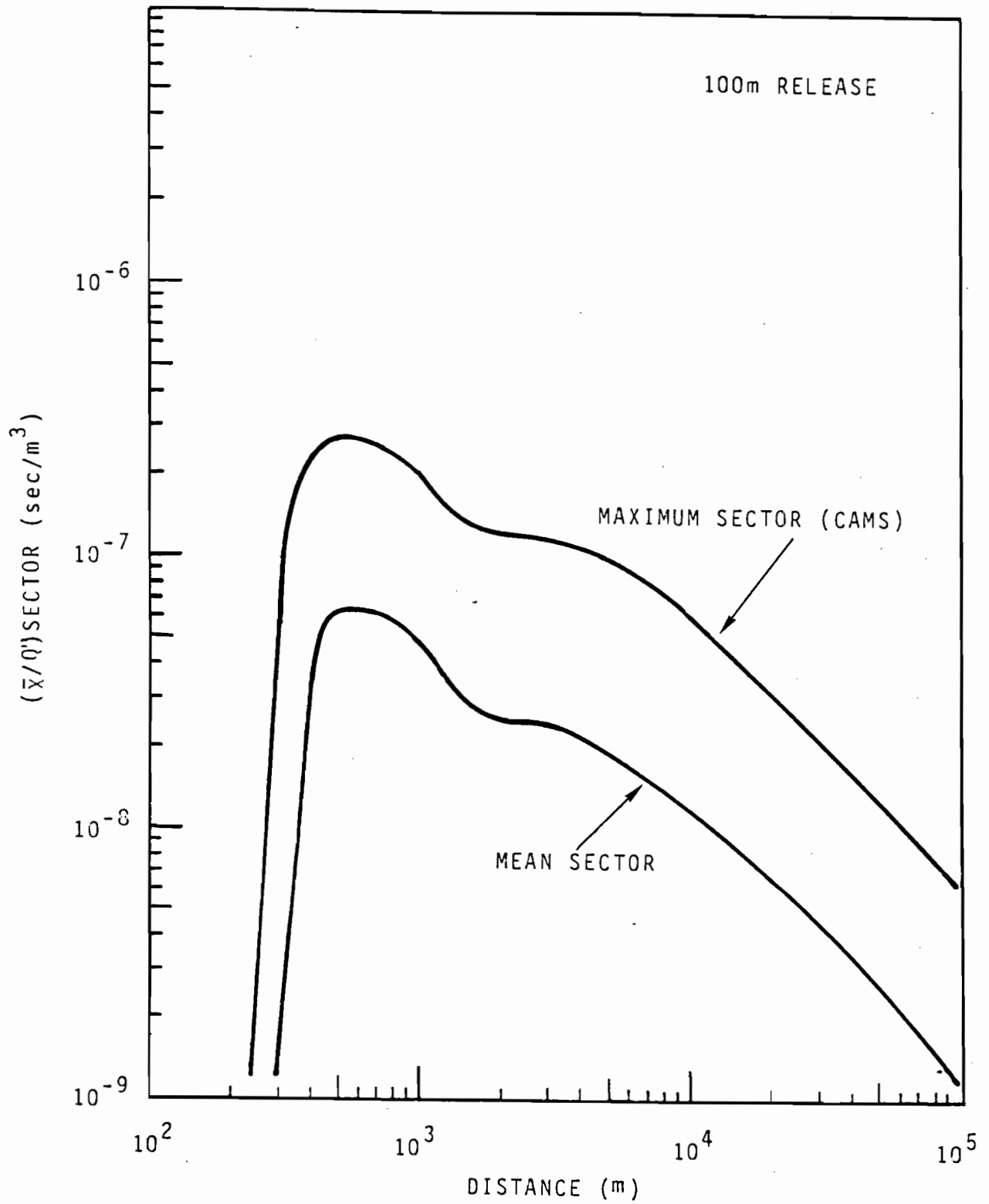


FIGURE D-1. Maximum (CAMS) and Mean  $\bar{x}/Q'$  Curves for 10 Meter Releases



**FIGURE D-2.** Maximum (CAMS) and Mean  $\bar{x}/Q'$  for 100 Meter Releases

applications. Conversely, even though CAMS has been shown to be conservative in this study, it is not expected, in a statistical sense, that the CAMS curves will enclose the population of all  $\bar{x}/Q'$  values.

In order to properly interpret the  $\bar{x}/Q'$  values calculated for the extreme distances presented herein, it must be understood that an upper limit has been placed on vertical diffusion. Such a limit does naturally occur and is discussed by Holzworth. For the United States this limit is around 1000 meters (2000 m has been used in this study). When no limit is placed on vertical diffusion, estimates of  $\bar{x}/Q'$  during Type A conditions are affected at distances beyond  $6 \times 10^3$  meters. These values will be underestimated less than an order of magnitude at  $10^5$  meters. Allowance may be made for the mixing depth in any particular region by use of maps published by Holzworth.<sup>(1)</sup>

The above method and the CAMS curves are not designed to replace in any way the need for onsite meteorological data collection programs. They are only for initial estimates for an undefined site for use in plant design. It is anticipated that once a particular site is chosen, onsite data will be collected and used to make a more accurate assessment of the diffusion climatology both for the routine as well as accidental release calculations.

#### REFERENCES

1. G. C. Holzworth, Mixing Heights, Windspeeds and Potential for Urban Air Pollution Throughout the Contiguous United States, Environmental Protection Agency, AP-101, January 1972.

APPENDIX E

EMPIRICAL METHOD FOR ESTIMATING THE TOTAL NUMBER  
OF FISSIONS FROM ACCIDENTAL CRITICALITY  
IN URANIUM OR PLUTONIUM SYSTEMS<sup>(1)</sup>



## APPENDIX E

### EMPIRICAL METHOD FOR ESTIMATING THE TOTAL NUMBER OF FISSIONS FROM ACCIDENTAL CRITICALITY IN URANIUM OR PLUTONIUM SYSTEMS<sup>(1)</sup>

#### INTRODUCTION

Each new plant that fabricates, processes, or otherwise handles fissionable materials undergoes a safety analysis prior to startup. One part of the safety analysis is to assess the potential consequences of a postulated worst credible criticality accident. To perform this assessment, the *total energy release*--expressed as "total number of fissions" from the criticality excursion--must be estimated or assumed.

In a plant where the assumed worst credible criticality accident is in a fissile solution system, the present industry practice is to assume a pre-established upper limit of  $4 \times 10^{19}$  fissions. This value is based somewhat on the past history of criticality accidents and the fact that the highest excursion to date resulted in an estimated  $4 \times 10^{19}$  fissions.

This appendix presents the results of a study to develop a model for predicting the total number of fissions from a criticality excursion in uranium or plutonium solution systems. Development of this model has been made possible by the recent criticality excursion experiments carried out in France by the Commissariat a l'Energie Atomique, referred to as the CRAC experiments.<sup>(2)</sup> These experiments have provided the first firm basis for developing an empirical model for predicting the total energy release from a criticality excursion in a given solution system.

#### SUMMARY

The criticality accident, as characterized by the model empirically derived from the experimental data provided by the CRAC experiments, is considered to be divided into 1) an initial fission burst followed by 2) a plateau period where the number of fissions/sec., ignoring oscillations, decreases with increasing time in the plateau.

The model for the initial burst relates the total fissions in the burst to a function of the volume (liters) of solution at the time of the burst. The empirically derived equation is

$$F_B = 2.95(10)^{15} V_B^{0.82}$$

The estimated 95% upper confidence level is given by

$$\log(F_B^U) = 15.47 + 0.82 \log V_B^0 + 0.23 \left[ 1.04 + \frac{(\log V_B^0 - 1.73)^2}{4.07} \right]^{1/2}$$

This relationship is assumed to hold for the vessel diameter range (11.8 to 31.5 inches) and solution addition rates used in the CRAC experiments.

Although the model developed for predicting the number of fissions in the initial burst is mainly applicable to highly enriched uranium systems, it is also applicable to plutonium and slightly enriched uranium systems. However, the energy release predicted will be conservatively high because of the presence of  $^{240}\text{Pu}$  and  $^{238}\text{U}$ , respectively, and other isotopes that undergo spontaneous fission. With a neutron source added for plutonium systems the fissions in the initial burst are estimated to be a factor of two or more lower than predicted by the model. A reduction factor has not been estimated for slightly enriched uranium.

The model for the number of fissions in the plateau is given by

$$F_p = 3.2(10)^{18} [1 - t^{-0.15}]$$

with the 95% upper confidence level given by

$$F_p^U = 4.6 \times 10^{19} [t^{0.02} - 1]$$

where  $t$  is the duration of the plateau in seconds and  $F_p$  is the number of fissions in the plateau. The model is developed from an empirically derived upper envelope fissions/sec plateau time plot and then integrating over the duration of the plateau ( $t$ ).

Thus, an estimate of the total number of fissions occurring during a criticality accident is obtained from  $F_T = F_B + F_p$ , the latter term generally being the main contributor.

Application of the model to find the potential energy release from a criticality accident for use in a plant safety analysis could be accomplished by listing the vessels to be considered, estimating a potential duration for the excursion based on past accidents, calculating for each vessel the total number of fissions using the model presented, and then selecting the highest number of fissions.

To test the model, the conditions of the seven past industrial criticality accidents<sup>(2)</sup> and three critical mass laboratory accidents were used to calculate initial burst and total energy release. The model is conservative in all but one instance.

## DISCUSSION

### Basic Approach

Even restricting attention to the CRAC experiments, considerable variation remains in the graphical plots of fissions/sec versus time of the individual criticality accident. However, the same basic pattern persists. Some empirical relationships, although not based on detailed physical processes, are useful in predicting the number of fissions for criticality accidents similar in nature to the CRAC experiments.

The criticality accident is divided into two parts: 1) an initial fission burst followed by 2) a plateau period where the average fission rate slowly, monotonically decreases. This is depicted in Figure E.1.

The fission burst at the onset of an accident is an immediate concern to personnel. Consequences depend on distance, the size of the burst, and the shielding between personnel and the event. A longer term concern is constituted by a release of fission products to the environment. The magnitude of a release is dependent upon the duration of the criticality, the fission product inventory (total fissions), and the adequacy of plant ventilation and filtering systems.

### CRAC Experiments

Development of the model is based on 41 criticality excursion experiments performed with highly enriched uranium at the Commissariat a l'Energie

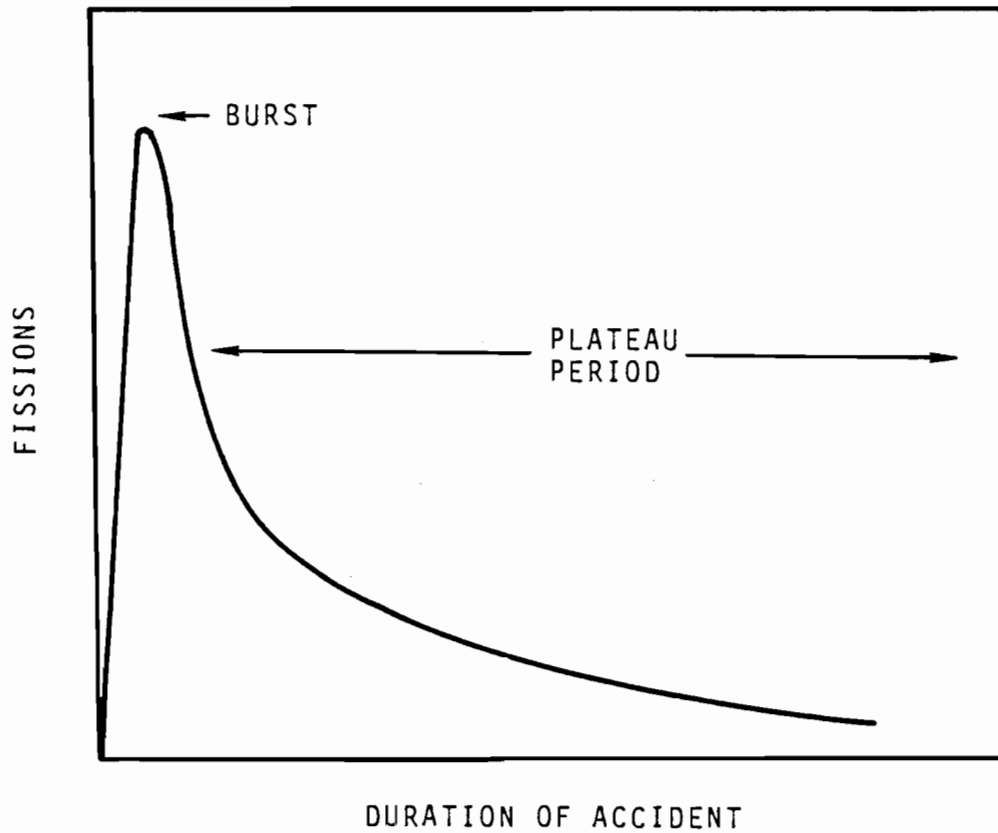


FIGURE E.1. Characteristic Energy Release from a Criticality Excursion

Atomique, France.<sup>(2)</sup> A summary is presented in Table E.1. The variables taken into consideration were as follows:

- Vessel Size (Two diameters were used: 11.8 in. ID and 31.5 in. ID)
- Neutron Source (Of the 41 experiments, 32 could be classified as having no initiating neutron source, four had a source by virtue of spray neutrons from the previous experiments.)
- Volume (Solution volumes at the time of the excursion ranged from 20 l to 250 l.)
- Excess Above Critical Height (Excess solution additions ranged from 4 cm to 185 cm above the critical height.)
- Mass (The total uranium involved in the excursions ranged from 1.6 kg up to 46 kg.)
- Solution Addition Rate
- Uranium Concentration (Ranged from 21 g U/l to 383 g U/l.)

TABLE E.1 Summary of Criticality Excursion Experiments Performed in France (CRAC Experiments)

CRAC No.	Vessel Diameter, in.	Neutron Source Present, mci	Volume at Pulse Peak, l	Excess Above Critical Height, cm	Final Mass, Kg U	Concentration, g U <sub>2</sub>	Solution Addition Rate, l/min	Duration of Plateau, Min.	Average Fission Rate During Plateau, Fissions/sec.	Fissions in Initial Burst, Fissions	Total Fissions (x 10 <sup>17</sup> )
1	11.81	-	226	180.1	13.1	52.0	24.0	255	1.5 x 10 <sup>13</sup>	27.0	5.0
2	11.81	-	173	72.5	10.3	55.0 (mik)	27.7	21	3.8 x 10 <sup>14</sup>	18.0	6.6
3	11.81	-	186	185.3	13.0	52.2	8.7	32	1.25 x 10 <sup>14</sup>	17.0	4.0
4	11.81	-	225	153.4	12.2	51.7	28.3	255	1.2 x 10 <sup>13</sup>	22.0	4.0
5	11.81	-	56.0	31.4	4.2	61.1	24.7	19	2.8 x 10 <sup>14</sup>	6.3	3.8
6	11.81	-	56.2	32.0	4.2	61.1	25.5	20	2.4 x 10 <sup>14</sup>	6.7	3.6
7	11.81	-	20.5	5.8	4.6	202	27.9	10	4.5 x 10 <sup>14</sup>	4.0	3.1
8	11.81	-	20.3	5.6	4.6	202	27.0	10	3.2 x 10 <sup>14</sup>	8.0	2.7
9	11.81	-	32.3	12.4	3.0	78.3	26.6	6	6.8 x 10 <sup>14</sup>	4.4	2.9
10	11.81	-	30.2	23.7	3.5	78.3	8.2	13	6.9 x 10 <sup>14</sup>	4.3	5.8
11	11.81	-	24	5.9	9.2	383.0	25.1	16	4.6 x 10 <sup>14</sup>	3.7	4.8
12	11.81	-	31.8	19.3	3.3	77.9	1.7	7	9.8 x 10 <sup>14</sup>	5.2	4.6
13	11.81	-	36.5	20.4	3.7	77.9	23.8	13	6.9 x 10 <sup>14</sup>	4.0	5.8
14	11.81	100 Residual (a)	31.1	24.7	3.6	77.9	10.3	5	1.7 x 10 <sup>15</sup>	3.7	5.5
15	11.81	Residual	23.9	24.0	3.7	82.3	24.8	5	1.1 x 10 <sup>15</sup>	3.6	9.0
16	11.81	Residual	30.3	40.1	4.6	82.3	7.8	13	1.3 x 10 <sup>15</sup>	3.7	4.9
17	11.81	Residual	30.4	20.2	3.4	79.6	8.3	6	2.3 x 10 <sup>14</sup>	3.7	50.0
18	11.81	Residual	30.1	60.4	5.5	79.6	1.7	360	1.5 x 10 <sup>15</sup>	3.5	4.9
19	11.81	-	30.8	21.6	3.5	82.0	8.5	5	7.5 x 10 <sup>14</sup>	2.0	2.0
20.1	11.81	200	19.7	4.3	4.7	218.0	24.4	4	1.4 x 10 <sup>15</sup>	2.1	0.22
20.2	11.81	200	19.7	5.9	5.0	218.0	21.5	3	1.1 x 10 <sup>15</sup>	2.0	0.61
20.3	11.81	200	19.8	3.1	4.6	218.0	25.0	0.25	1.4 x 10 <sup>15</sup>	2.2	2.7
20.4	11.81	-	20.2	3.5	4.6	218.0	24.8	0.25	1.5 x 10 <sup>15</sup>	3.1	4.9
20.5	11.81	(30 min. after 20.4)	19.7	5.9	5.0	218.0	22.3	3	1.5 x 10 <sup>15</sup>	4.2	5.3
21	11.81	-	31.1	21.5	3.5	80.2	8.4	5	1.0 x 10 <sup>15</sup>	3.9	0.39
22	11.81	-	20.0	6.4	4.8	207.0	17.8	3	1.4 x 10 <sup>15</sup>	3.9	3.0
23	11.81	-	27.2	21.9	3.7	91.8	23.0	7	2.7 x 10 <sup>15</sup>	3.5	2.8
24	11.81	-	39	28.7	6.0	153.2	-	-	1.6 x 10 <sup>15</sup>	3.7	5.1
25	11.81	-	22.2	5.5	7.3	320.0	31.1	-	2.2 x 10 <sup>15</sup>	3.3	4.3
26	11.81	-	23.1	12.2	4.6	166.0	25.3	3	8.1 x 10 <sup>15</sup>	34.0	32.0
27	11.81	-	28.4	23.6	3.8	89.0	25.6	1.5	1.2 x 10 <sup>15</sup>	32.0	27.0
28	11.81	-	29.1	21.3	3.6	65.0	18.4	3	7.9 x 10 <sup>15</sup>	16.0	16.0
29	11.81	-	30.8	22.2	3.6	81.0	8.4	2	8.3 x 10 <sup>15</sup>	13.0	10.8
37	31.5	-	230.8	7.1	5.6	21.8	7.5	5.8	5.2 x 10 <sup>15</sup>	8.0	10.2
38	31.5	-	241.2	5.8	5.5	21.4	21.9	3.3	4.8 x 10 <sup>15</sup>	17.0	4.6
39	31.5	-	138.6	1.7	4.4	30.6	23.3	3	5.1 x 10 <sup>15</sup>	13.0	10.5
40	31.5	-	92.9	.7	5.6	58.7	13.8	2	2.5 x 10 <sup>16</sup>	19.0	17.0
41	31.5	-	92.9	.6	5.6	59.3	1.6	3			
42	31.5	-	72.5	.9	15.5	203.0	21.3	1			
43	31.5	-	72.0	1.1	15.5	202.0	23.5	3			
44	31.5	-	246	-	9.2	(b)	46	1			

(a) Neutrons present from previous burst.

(b) 46 ± 200 g U<sub>2</sub> into 200 ± 2N HNO<sub>3</sub>.

Development of Initial Burst Model (Source not present)

The first model examines the number of fissions in the initial burst  $F_B$ , and relationship with volume at time of burst  $V_B$  and rate of solution addition  $A_R$ .

Ignoring the addition rate for the moment, there is a definite relationship between volume and fissions in burst (Figure E.2). In Figure E.2, a cluster of points at  $V_B = 19.7$  can be attributed to the presence of a neutron source during the criticality experiment. At  $V_B \approx 30$  another cluster of points is not as readily explained. Careful examination, however, reveals that some of these data points result from experiments with neutron sources present and that if the solution addition rate is taken into consideration, then the number of fissions in the burst is reduced for these experiments (as expected).

A second major point of interest is the definite difference in the experiments conducted in the two different diameter size vessels. The same basic relationship between volume and fissions in the burst remains the same except for a shift in position. A similar phenomenon occurs in the relationship between volume at burst and concentration (Figure E.3).

By restricting attention to those experiments with the same vessel diameter size, and excluding any experiments with an external neutron source, a specific empirical model can be derived based on the data contained in Table E.2. An empirical model of the form

$$\log(F_B) = A + B \log(V_B) + C \log A_R$$

was fit by least squares to give

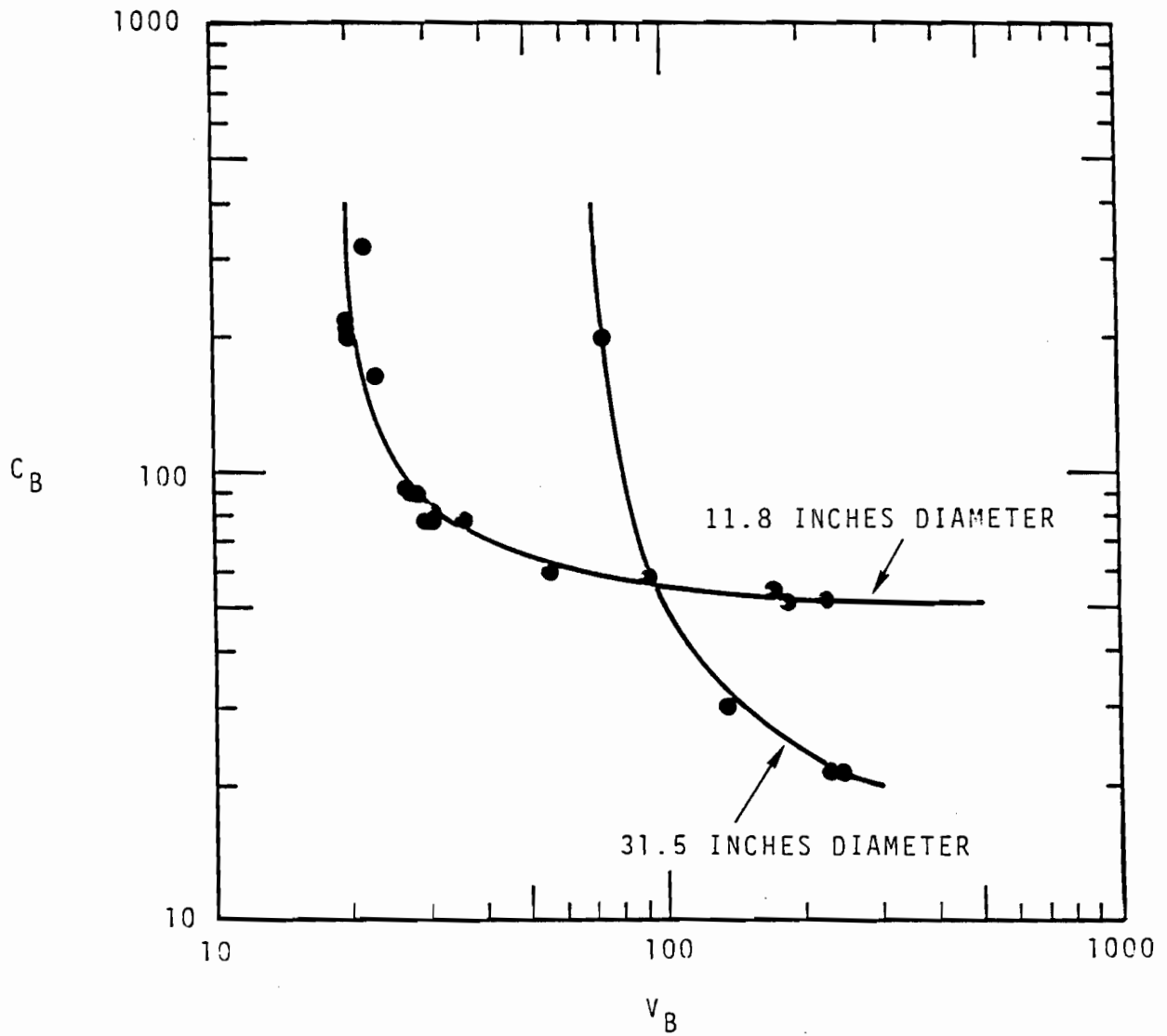
$$\log(F_B) = 15.36 + 0.72 \log(V_B) + 0.20 \log A_R \quad (\text{Model I})$$

or in the terms of the original variables

$$F_B = 2.29 \times 10^{15} V_B^{0.72} A_R^{0.20}$$







**FIGURE E.3.** Empirical Relationship Between Volume at Pulse Peak and Concentration of Uranium Solution for CRAC Experiments. (Note the Shift in the Relationship for the 11.8 in. and 31.5 in. Vessel Diameter Experiments.)

TABLE E.2. Summary of Fissions in Burst as Related to Volume at Peak Power and Solution Addition Rate

<u>CRAC</u> <u>Exp. No.</u>	<u>Volume at Peak</u> <u>Power(<math>V_B</math>), liters</u>	<u>Solution Addition Rate (<math>A_R</math>),</u> <u>liters/min</u>	<u>Fissions in Burst (<math>F_B</math>),</u> <u>x 10<sup>16</sup></u>
<u>11.81 in. Diameter Vessel</u>			
20.2	19.7 <sup>a</sup>	21.5	2.1
20.1	19.7 <sup>a</sup>	24.4	2.0
20.5	19.7 <sup>a</sup>	22.3	2.2
20.3	19.8 <sup>a</sup>	25.0	2.0
22	20.0	17.8	4.2
20.4	20.2	24.8	5.9
8	20.3	27.0	8.0
7	20.5	27.9	4.0
25	22.2	31.1	3.9
26	23.1	25.3	3.9
23	27.2	23.0	4.2
27	28.4	25.6	3.5
28	29.1	18.4	3.7
15	29.9 <sup>a</sup>	24.8	3.7
18	30.1 <sup>a</sup>	1.7	3.7
10	30.2	8.2	4.3
16	30.3 <sup>3</sup>	7.8	3.6
17	30.4 <sup>a</sup>	8.3	3.7
19	30.8	8.5	3.5
29	30.8	8.4	3.3
21	31.1	8.4	3.1
14	31.1 <sup>a</sup>	10.3	4.0
12	31.8	1.7	3.7
19	32.3	26.6	4.4
13	36.5	23.8	5.2
5	56.0	24.7	6.3
6	56.2	25.5	6.7
2	173.0	27.7	18.0
3	186.0	8.7	17.0
4	225.0	28.3	22.0
1	226.0	24.0	27.0
<u>31.5 in. Diameter Vessel</u>			
43	72.0	23.5	13.0
42	72.5	21.3	17.0
41	92.9	1.6	8.0
40	92.9	13.8	13.0
39	138.6	23.3	18.0
37	230.8	7.5	34.0
38	241.0	21.9	32.0

a) A source of neutrons was present during experiment. For other experiments no special source was present.

Hence, an increase in either the solution addition rate  $A_R$  or the solution volume of time of burst  $V_B$  leads to an increase in the estimated number of fissions in the burst.

An inspection of the fit indicates that there are two possible outliers, CRAC numbers 8 and 20.4. These points appear to be valid; however, both experiments were with high concentrations which might lead to a great variability in the fissions in the burst.

An estimated 95% upper confidence bound on a predicted value of  $\log(F_B)$  corresponding to a particular volume  $V_B^0$  and addition rate  $A_R^0$  is given by:

$$\log(F_B) = 15.36 + 0.72 \log(V_B^0) + 0.20 \log A_R^0 + 0.20 \left\{ 1 + [1 \log V_B^0 \log A_R^0] \begin{bmatrix} 1.71 & -0.58 & -0.59 \\ -0.58 & 0.38 & -0.03 \\ -0.59 & -0.03 & 0.51 \end{bmatrix} \begin{bmatrix} 1 \\ \log V_B^0 \\ \log A_R^0 \end{bmatrix} \right\}^{1/2}$$

For example, the predicted value for CRAC #1 is  $\hat{F}_B = 2.2 \times 10^{17}$  and the upper bound is  $3.6 \times 10^{17}$ .

Similar results can be obtained for the larger diameter size vessel, but the limited number of experiments for the larger size restricts the usefulness of such analyses. However, these experiments will be incorporated in the following analysis.

A reasonable empirical procedure for combining the different vessel diameter size experiments is based on the relationship between volume and concentrations shown in Figure E.3 and the corresponding plots in Figure E.2. As the diameter of the vessel increases, with the resulting shift in the volume versus concentration curve in Figure E.3, the fissions in burst versus volume relationship will be expected to remain the same, but the range of volume will be shifted upward. The suspected increase in variability at the high concentrations previously noted also is present for the large diameter

vessel, and this is postulated as being present for the intermediate diameter size vessels. Hence, if all the diameter sizes were included in a single analysis, then the same empirical relationship as derived from a single diameter size would hold, but with an increased variability. In terms of estimating an upper bound on fissions in a burst, the larger variability would result in a higher estimated bound.

Combining the data from all the CRAC experiments without external neutron sources results in

$$\log(F_B) = 15.24 + 0.83 \log(V_B) + 0.18 \log A_R \quad (\text{Model II})$$

or in the original variables

$$F_B = 1.74 \times 10^{15} V_B^{0.83} A_R^{0.18} .$$

An estimated 95% upper confidence bound on a predicted value of  $\log(F_B)$  for a given  $V_B^0$  and  $A_R^0$  is

$$\log(F_B) = 15.24 + 0.83 \log(V_B^0) + 0.18 \log A_R^0 + 0.21 \left\{ 1 + \begin{bmatrix} 1 & \log V_B^0 & \log A_R^0 \end{bmatrix} \begin{bmatrix} 1.29 & -0.44 & -0.41 \\ -0.44 & 0.25 & 0.02 \\ -0.41 & 0.02 & 0.32 \end{bmatrix} \begin{bmatrix} 1 \\ \log V_B^0 \\ \log A_R^0 \end{bmatrix} \right\}^{1/2}$$

Using the same example as above, CRAC #1, the predicted number of fissions in burst is  $2.8 \times 10^{17}$  with an upper bound of  $4.7 \times 10^{17}$ ; compared to  $2.2 \times 10^{17}$  and  $3.6 \times 10^{17}$ , respectively.

Although the solution addition rate is a significant variable in the empirical relationship, in a particular application it may be necessary to estimate the number of fissions based on the volume at the time of burst alone. The estimated relationship without a term for the addition rate is

$$\log(F_B) = 15.47 + 0.82 \log V_B \quad (\text{Model III})$$

based on the same data as Model II, with an estimated 95% upper confidence of

$$\log(F_B) = 15.47 + 0.82 \log V_B^0 + 0.23 \left\{ 1.04 + \frac{(\log V_B^0 - 1.73)^2}{4.07} \right\}^{1/2}$$

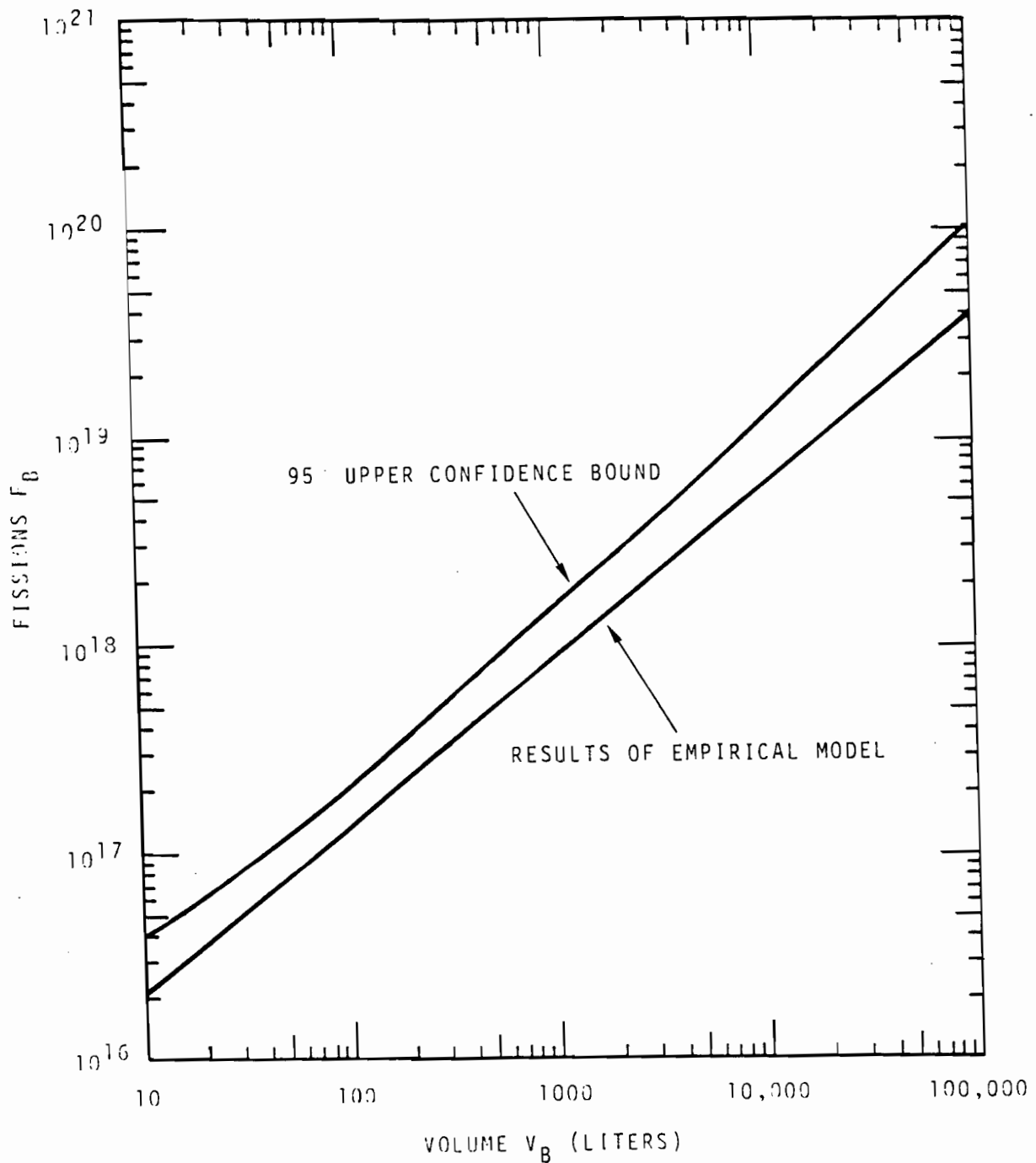
Again using CRAC #1 as an example, the predicted value of  $F_B$  is  $2.5 \times 10^{17}$  with an upper bound of  $4.4 \times 10^{17}$ .

Table E.3 gives the estimated number of fissions in initial burst and the 95% upper confidence limit for selected volume sizes using Model III. This is graphically displayed in Figure E.4.

TABLE E.3. Number of Fissions in Initial Burst ( $F_B$ ) as a Function of Vessel Volume (based on Model III)

Volume ( $V_B$ ) liters	Estimated		95% Upper Confidence	
	$\log(F_B)$	$F_B$	$\log(F_B)$	$F_B$
50	16.86	$7.2 \times 10^{16}$	17.10	$1.2 \times 10^{17}$
100	17.11	$1.3 \times 10^{17}$	17.35	$2.2 \times 10^{17}$
500	17.68	$4.8 \times 10^{17}$	17.94	$8.7 \times 10^{17}$
1,000	17.93	$8.5 \times 10^{17}$	18.21	$1.6 \times 10^{18}$
5,000	18.50	$3.2 \times 10^{18}$	18.82	$6.7 \times 10^{18}$
10,000	18.75	$5.6 \times 10^{18}$	19.10	$1.2 \times 10^{19}$
50,000	19.32	$2.1 \times 10^{19}$	19.73	$5.4 \times 10^{19}$
100,000	19.57	$3.7 \times 10^{19}$	20.01	$1.0 \times 10^{20}$

In the above models, all CRAC experiments with an external neutron source were excluded from the analysis. CRAC experiments have shown that when an external source is present, these models will overpredict the number of fissions in the burst.



**FIGURE E.4.** Least Squares Estimated Relationship Between the Number of Fissions in Initial Burst and the Volume at Pulse Peak Using Model III. (Based on CRAC Experiments Without External Neutron Sources. Upper Curve Gives the 95% Upper Confidence Bound.)

In summarizing the analysis of the initial burst portion of the criticality accidents, it is emphasized that the models are empirically derived and are restricted to simple functional forms. More complicated and realistic models can undoubtedly be formulated based on the detailed physical characteristics of the individual CRAC experiments. The empirical models developed here should be adequate for estimating upper bounds on the number of fissions in the initial burst from criticality accidents of a similar nature to the CRAC experiments. A choice of which model can be made depending on the level of information available. If the volume and solution addition rate are known, but the vessel diameter size is only known to be between 11 to 32 inches, then Model II can be used.

#### Development of Plateau Model

An empirical method for estimating the number of fissions in the plateau region of the criticality accident can be derived based on the average fission rate during the plateau and the time spent in the plateau. A comparison of different CRAC experiment time plots of the number of fissions/sec shows that although the plots have similar characteristics, a common plot does not apply to all experiments.

If the average fission rates  $F_R$ , based on plateau length of  $t$  seconds, are plotted at time  $t/2$  seconds, then the resulting plot resembles a time plot of fissions/sec for a single experiment. More importantly, if the individual CRAC experiments are plotted on the same graph, then the  $F_R$  versus  $t/2$  plot forms an upper envelope for the individual plateau plots. An estimate of the total fissions during the plateau period based on this upper envelope should give a reasonable upper bound when applied to a particular criticality accident.

The time during the plateau for the CRAC experiments is artificial in the sense that the experiments were manually terminated. The use of this time variable in the model is to construct the upper envelope plateau function and is not used directly to estimate the total fissions in the plateau. From the evidence available, the artificial termination of the experiments does not seem to invalidate the approach taken.

Figure E.5 gives the plot of average fission rate  $F_R$  as a function of time. For an  $F_R$  based on  $t$  seconds, the  $F_R$  value is plotted at  $t/2$ . This denotes that  $F_R$  is an estimate of the fissions/sec at time  $t/2$  for the upper envelope plateau function. The plot suggests the simple functional form

$$\log F_R = A - B \log t.$$

The parameters  $A$  and  $B$  estimated by least squares from the data in Table E.4 (excluding the \* data point) gives

$$\log F_R = 17.68 - 1.15 \log t$$

$$\text{or } F_R = \frac{4.8 \times 10^{17}}{t^{1.15}},$$

where  $t$  is in seconds and  $F_R$  in fissions/sec.

A 95% upper confidence bound on an estimated value of  $F_R$  at a given point in time  $t$  is

$$\log F_R = 17.68 - 1.15t + .51 \left\{ 1.03 + \frac{(\log t - 2.33)^2}{9.14} \right\}^{1/2}.$$

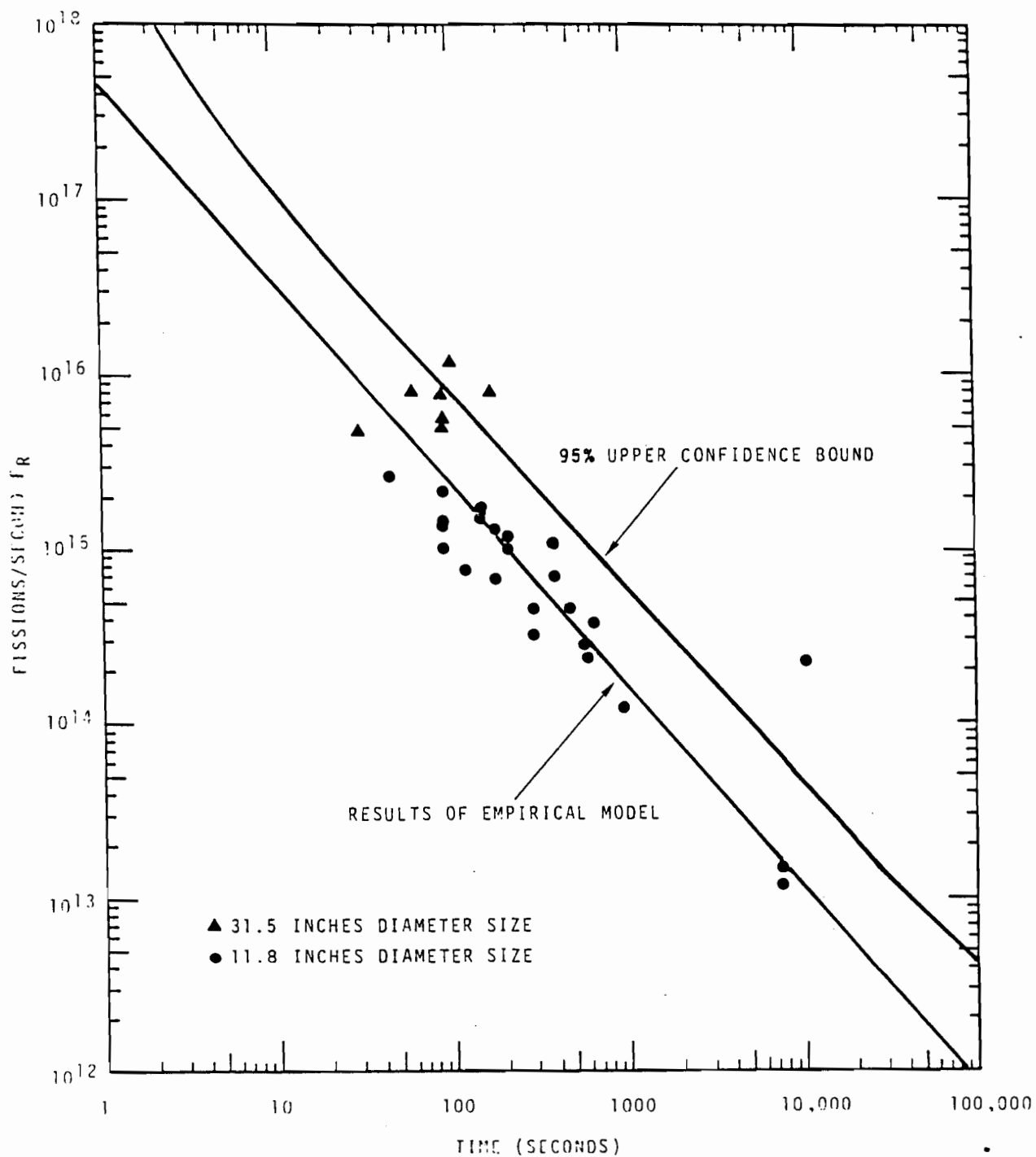
An estimate of the number of fissions in plateau during time  $t_0$  to  $t_1$  is obtained by integrating

$$F_p = \int_{t_0}^{t_1} A' t^{-B} dt = \frac{A' [t_1^{1-B} - t_0^{1-B}]}{1-B}.$$

If  $B = 1$ , then the above is indeterminate and the correct form is

$$F_p = A' [\ln t_1 - \ln t_0].$$





**FIGURE E.5.** Average Fission Rate in Plateau and One-half the Duration of Plateau for CRAC Experiments. (Lower and Upper Curves are the Regression and 95% Upper Confidence Limit for the Upper Envelope Fission/sec. Plateau Function.)

TABLE E.4. Average Fission Rate as a Function of Time

<u>CRAC Expt. No.</u>	<u>Average Fission Rate (<math>F_R</math>), Fissions/sec x <math>10^{13}</math></u>	<u>Time (t), seconds</u>
42	480(a)	30
27	270	45
40	830(a)	60
43	510(a)	90
41	520(a)	90
39	780(a)	90
29	220	90
26	140	90
22	100	90
20.5	140	90
20.2	140	90
38	1200(a)	99
20.1	75	120
28	160	150
21	150	150
19	150	150
15	170	150
37	810(a)	150
9	68	174
17	130	180
23	120	180
13	98	210
7	45	210
8	32	300
10	69	300
14	69	390
16	110	390
12	46	390
5	28	480
6	24	570
2	38	600
3	12.5	630
1	1.5	960
4	1.2	7,650
18	23.0(a)	7,650
		10,800

a. Measurements from 31.5 in. diameter vessel experiments

Also if B is less than or equal to 1, then as  $t_1$  increases with  $t_0$  constant  $F_p$  increases to infinity. When B is greater than 1,  $F_p$  still increases but reaches the asymptotic value  $A' t_0^{1-B}/(B-1)$ .

It is assumed in the remainder of the analysis that  $t_0 = 1$  sec., which will result in  $F_p$  estimating the total number of fissions in a plateau that lasts  $t_1$  seconds. From the empirical model IV this gives

$$F_p = 3.2 \times 10^{18} \left[ 1 - \frac{1}{t_1^{0.15}} \right].$$

An approximate 95% upper bound for  $F_p$  is given by

$$F_p^U = 4.6 \times 10^{19} [t_1^{0.02} - 1].$$

In the development of Model IV, the starred data point in Table E.3 is obviously different from the remainder of the data and hence was excluded from the analysis. This point is from CRAC experiment 18 and multiple excursions occurred during the plateau. Also, the experiments using the large diameter vessel are grouped together in Figure E.5 which suggests that they may behave differently from the other experiments. Furthermore, any judgment concerning the behavior of other intermediate diameter size experiments is difficult.

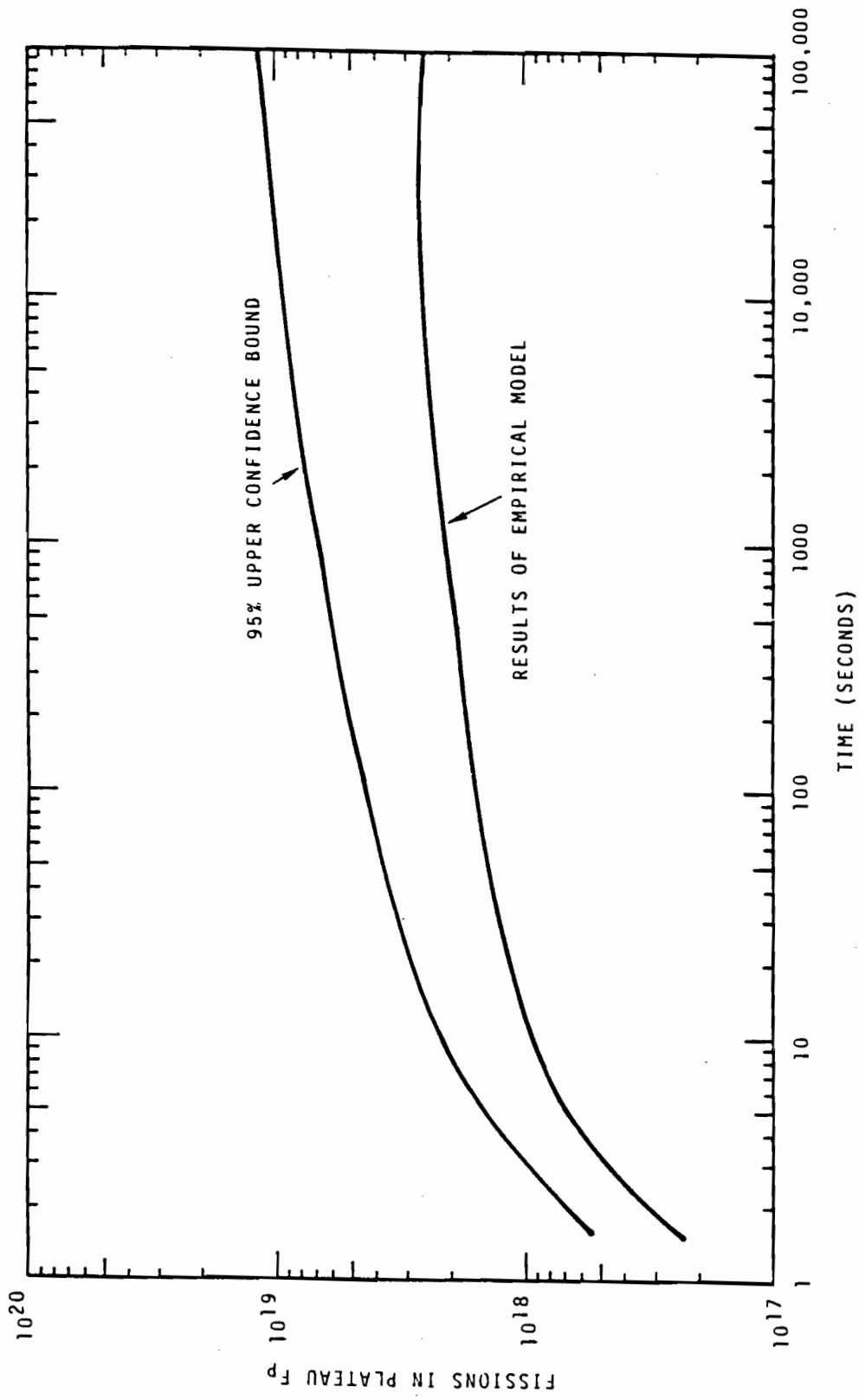
Summary information concerning the analysis of the plateau section of the criticality safety analysis is presented in Tables E.5 and E.6 and graphically in Figures E.5 and E.6.

#### Calculation of Total Number of Fissions

An estimate and upper bound for the total number of fissions  $F_T$  during a criticality can be constructed as  $F_T = F_B + F_p$ . This combines the information from the two different portions of the criticality but does not account for any interdependence between the portions.

**TABLE E.5.** Estimates of Upper Envelope Plateau Function ( $F_R$ ) as a Function of Time

Time (t), seconds	Estimate		95% Upper Confidence Bound	
	$\log(F_R)$	$F_R$ , fissions/sec.	$\log(F_R)$	$F_R$ , fissions/sec.
1	17.68	$4.8 \times 10^{17}$	18.33	$2.1 \times 10^{18}$
5	16.88	$7.6 \times 10^{16}$	17.47	$2.9 \times 10^{17}$
10	16.53	$3.4 \times 10^{16}$	17.10	$1.2 \times 10^{17}$
50	15.73	$5.4 \times 10^{15}$	16.26	$1.8 \times 10^{16}$
100	15.38	$2.4 \times 10^{15}$	15.90	$8.0 \times 10^{15}$
500	14.58	$3.8 \times 10^{14}$	15.10	$1.3 \times 10^{15}$
1,000	14.23	$1.7 \times 10^{14}$	14.76	$5.8 \times 10^{14}$
5,000	13.43	$2.7 \times 10^{13}$	14.00	$1.0 \times 10^{14}$
10,000	13.08	$1.2 \times 10^{13}$	13.67	$4.6 \times 10^{13}$
50,000	12.28	$1.9 \times 10^{12}$	12.94	$8.6 \times 10^{12}$
100,000	11.93	$8.5 \times 10^{11}$	12.61	$4.1 \times 10^{12}$



**FIGURE E.6.** Estimated Total Number of Fissions Occurring in Plateau Region of an Excursion as a Function of the Duration of Plateau. (The 95% Upper Confidence Bound is Presented as the Upper Curve.)

TABLE E.6. Total Number of Fissions in Plateau ( $F_p$ )  
as a Function of Time

<u>Time,</u> <u>seconds</u>	<u>Estimate</u> <u><math>F_p</math></u>	<u>95% Upper Confidence Bound</u> <u><math>F_p^U</math></u>
5	$6.9 \times 10^{17}$	$1.5 \times 10^{18}$
10	$9.3 \times 10^{17}$	$2.2 \times 10^{18}$
50	$1.4 \times 10^{18}$	$3.7 \times 10^{18}$
100	$1.6 \times 10^{18}$	$4.4 \times 10^{18}$
500	$1.9 \times 10^{18}$	$6.1 \times 10^{18}$
1,000	$2.1 \times 10^{18}$	$6.8 \times 10^{18}$
5,000	$2.3 \times 10^{18}$	$8.5 \times 10^{18}$
10,000	$2.4 \times 10^{18}$	$9.3 \times 10^{18}$
50,000	$2.6 \times 10^{18}$	$1.1 \times 10^{19}$
100,000	$2.6 \times 10^{18}$	$1.2 \times 10^{19}$

COMPARISON OF MODEL TO ACTUAL CRITICALITY EXCURSIONS

To test the model, the conditions of nine past criticality accidents were compared to predictions by the model (Table E.7). The accidents included in this test were P-1 through P-7 and SE-2, SE-3 and SE-5 from Table E.8.

Conclusions are as follows:

1. The model over predicts the energy release in all ten instances except for P-3. This excursion occurred at the Reactor Test Station, Idaho Falls. The discrepancy could be due to the special characteristics of the system that reached criticality or to the uncertainty in the analysis of the accident.
2. For the uranium excursions, the model over predicts the total number of fissions by a factor of 2 to 30, with the one exception noted above.
3. For the plutonium excursions, the model over predicts by a factor of 3 to 20. In one instance (P-7), the prediction is 1600 times higher, which could be due to uncertainties in the analysis of the accident or to the unusual characteristics of the plutonium system.

TABLE E.7. Comparison of Total Energy Release Predicted by Model and Actual Energy Release of Past Criticality Accidents

No.	Location	Form	Characteristics of Fissile Material System		Volume, $\ell$	Duration of Excursion Min.	Number of Fissions ( $\times 10^{17}$ )		
			Mass, Kg Fissile	Mass, Kg Fissile			Actual Excursion Initial Burst	Total	Predicted By BNW Model Initial Burst
P-1	Y12	UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> (a)	2.5	56	13	~ 0.1	13.0	0.8	21.0
P-2	LASL	Pu/Organic	3.3	168	< 1	1.5	1.5	2.0	16.7
P-3	IF	UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> (a)	34.5	800	20	1.0	400.	7.1	28.1
P-4	IF	UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> (a)	8.0	40	< 1	~ 0.6	6.0	0.6	15.3
P-5	Hanford	Pu Complex	1.5	~ 60	2220	~ 0.1	8.0	0.9	27.4
P-6	Wood River	UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> (a)	2.6	~ 70	< 1	1.1	1.3	1.0	15.7
P-7	Windscale	Pu/Organic	2.5	~ 100	< 1	0.01	0.01	1.3	16.0
SE-2	ORNL	Pu(NO <sub>3</sub> ) <sub>4</sub>	1.15	64	< 1	~ 0.8	0.8	0.9	15.6
SE-3	ORNL	UO <sub>2</sub> F <sub>2</sub>	18.3	55	< 1	0.5	0.5	0.8	15.5
SE-5	ORNL	<sup>233</sup> UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub>	~ 1.0	5.8	< 1	0.11	0.11	0.1	14.8

(a) Uranium Enrichment ~ 93 wt% <sup>235</sup>U

TABLE E.8. Summary of Known Accidental Criticality Excursions Since 1945

No.	Date	Location	Title For References	Fissionable Material	Arrangement	Energy Release			Cause	Physical Damage
						Initial Prompt Critical BURST, Fissions	Duration of Criticality	Total Fissions		
SE 1	December 1949	LASL, New Mexico	Water Boiler	U(93)O <sub>2</sub> (W <sub>2</sub> ) <sub>2</sub> (-1 kg <sup>235</sup> U; 13.6 g)	Sphere, granite-reflected	~3x10 <sup>15</sup> (barely over prompt critical)	Not known	3-4x10 <sup>16</sup>	Control rods withdrawn too fast	None
SE 2	November 16, 1951	Hanford Works, Richland, Washington	P-11	PuO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> (1.15 kg Pu; 63.8 g)	Sphere, 92% full reflected	8x10 <sup>16</sup>	Single burst	8x10 <sup>16</sup>	Too high fuel addition	None
SE 3	May 26, 1954	ORNL, Tennessee	Spider	UO <sub>2</sub> F <sub>2</sub> (18.3 kg <sup>235</sup> U; 55.4 g)	Cylindrical annulus unreflected	5x10 <sup>16</sup>	Not known	1x10 <sup>17</sup>	Shift of neutron	None
SE 4	February 1, 1956	ORNL, Tennessee	Scram blade	UO <sub>2</sub> F <sub>2</sub> (27.7 kg <sup>235</sup> U; 58.9 g)	Cylinder unreflected	1.6x10 <sup>17</sup>	Single burst	1.6x10 <sup>17</sup>	Geometry change	Warping of bottom of cylinder (gevser).
SE 5	January 30, 1968	ORNL, Tennessee	U-233	UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> (1 kg <sup>233</sup> U; 5.8 g)	Sphere water-reflected	1.1x10 <sup>16</sup>	Single burst	1.1x10 <sup>16</sup>	Air in line	None
P 1	June 16, 1958	ORNL, Tennessee, Y-12 Processing Plant	Y-12	UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> (2.5 kg <sup>235</sup> U; 56 g)	Cylinder concrete reflected below	~1x10 <sup>16</sup>	13 min.	1.3x10 <sup>18</sup>	Valve leaked or left open	None (loss: \$1,000)
P 2	December 30, 1958	LASL, New Mexico Pu Processing Plant	Agitator	PuO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> (3.27 kg Pu; 168 g)	Cylinder water reflected below	1.5x10 <sup>17</sup>	Single burst	1.5x10 <sup>17</sup>	Procedure not followed	None
P 3	October 16, 1959	Idaho Reactor Testing Area, Chemical Processing Plant	IF-1 (siphon)	UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> (34.5 kg <sup>235</sup> U; .800 g)	Cylinder concrete reflected below	~10 <sup>17</sup>	Not known	~4x10 <sup>19</sup>	Sparre gauge damaged	None (loss: \$62,000)
P 4	January 25, 1961	Idaho Reactor Testing Area, Chemical Processing Plant	IF-II (air lift)	UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> (8 kg <sup>235</sup> U; 40 g)	Cylinder	No estimate	Not known	6x10 <sup>17</sup>	Instructions misinterpreted	None (loss: \$6,000)
P 5	April 7, 1962	Hanford Works, Richland, Washington	Recuplex	Pu complex (1.5 kg Pu)	Cylinder unreflected	~10 <sup>16</sup>	37 hrs.	8x10 <sup>17</sup>	Valve leaked or opened	None (loss: \$1,000)
P 6	July 24, 1964	Wood River Junction, R.I., scrap recovery facility	Wood River	UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> (2.64 kg <sup>235</sup> U)	Cylinder unreflected	1.1x10 <sup>17</sup>	Not known	1.3x10 <sup>17</sup>	Procedure not followed	None
P 7	August 24, 1970	Winnscale Works, England	Winnscale	Pu complex (2.5 kg Pu; .100 g)	Cylinder	No estimate	5-10 sec.	1x10 <sup>15</sup>	Pu accumulated in organic	None



4. If 95% upper confidence limits are applied, the predictions are 2.5 to 4.0 times higher than predicted by the basic model.

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1. R. L. Hooper, T. Olsen, V.O. Uotinen and C. L. Brown, Empirical Method for Estimating the Total Number of Fissions from Accidental Criticality in Uranium or Plutonium Systems, BNWL-1840, Battelle, Pacific Northwest Laboratories, Richland, WA, July 1974.
2. P. Lécorché and R. L. Seale, "A Review of Experiments Performed to Determine the Radiological Consequences of a Criticality Accident," Y-CDC-12, Criticality Data Center, November, 1973.
3. W. R. Stratton, "A Review of Criticality Accidents," LASL Report LA-3611, 1967.

APPENDIX F  
DATA USEFUL IN THE EVALUATION OF AIRBORNE PLUTONIUM  
FROM POSTULATED ACCIDENT SITUATIONS

## APPENDIX F

### DATA USEFUL IN THE EVALUATION OF AIRBORNE PLUTONIUM FROM POSTULATED ACCIDENT SITUATIONS

Information on the behavior of plutonium that may be useful in evaluating the fractional airborne release of plutonium from postulated accidents has been abstracted. The data have been collected from a variety of sources--from experiments, calculated, accident reviews, etc.--and encompass a variety of circumstances. The quantity and characteristics of the material airborne in an accident are the result of a complex interrelationship between mechanisms, airflow, and material at the point and time of release. Each accident is a unique and complex event, and conditions surrounding the event are usually ill-defined. Discrete properties of the materials of concern are seldom known in detail. Thus, considerable judgment is required to apply these data drawn from experimental studies to large scale fires, explosions, etc. In the absence of better-defined data, one must use the available data conservatively, selecting release values obtained under laboratory conditions as much like those occurring in an accident as possible. Effort to obtain exact correspondence between accident conditions and experimental conditions for which release data are available will be unrewarding and only approximate release factors can be selected. Knowing this, one must acknowledge the rather large uncertainties in predicting airborne concentrations following an accident.

Much of the basic information on the chemical and physical properties of plutonium and compounds has recently been gathered into a single source.<sup>(1)</sup> Significant quantities of plutonium do not vaporize under conditions postulated for industrial accidents.<sup>(2)</sup> Information pertinent to the airborne release of plutonium published prior to 1964 (almost exclusively on metals) has been reviewed.<sup>(3,4,5)</sup> Release data published since that time are summarized below.

## CHEMICAL AND PHYSICAL PROPERTIES OF PLUTONIUM

### Metal

Freshly cleaned plutonium is bright and has the appearance of nickel. Six allotropes of plutonium exist between room temperature and 640°C, the melting point of the metal. Plutonium oxidizes slowly in any air, but relatively rapidly in a moist atmosphere.<sup>(1)</sup>

Stewart found the oxidation rate and release of oxide to be continuous for the pure metal. Repeated cycling through the  $\alpha \rightleftharpoons \beta$  phase transition appeared to cause a marked increase in the oxidation rate.<sup>(6)</sup> Chatfield has provided a series of graphs for order of magnitude estimates of aerosol generation from an oxidizing plutonium surface where there is no disturbance except airflow. Both unalloyed and delta stabilized metal are considered in dry and moisture-saturated air at temperatures from ambient to 100°C. The values are given in  $\mu\text{Ci}/\text{cm}^2/\text{sec}$ . If one assumes a specific activity of  $6.17 \times 10^{-2} \text{ Ci/g}$  ( $^{239}\text{Pu}$ ), quantities of particles less than 10  $\mu\text{m}$  (AED) generated ranged from  $10^{-6}$  to  $10^{-5} \text{ g}/\text{cm}^2/\text{sec}$  in dry air and  $6 \times 10^{-12}$  to  $8 \times 10^{-6} \text{ g}/\text{cm}^2/\text{sec}$  in saturated air. In moist air, unalloyed metal appears to generate an order of magnitude greater quantity of particles in this size range than alloyed metal.<sup>(7)</sup>

Mishima found very little plutonium airborne during the oxidation in dry air of ignited, small specimens of unalloyed metal. From  $3 \times 10^{-6}$  to  $5 \times 10^{-5}\%$  of the plutonium was found in air passing through the quartz tube furnace at velocities ranging from 3.3 to 5 cm/sec. As much as 0.03% of the residue was composed of particles less than 15  $\mu\text{m}$  Aerodynamic Equivalent Diameter (AED).<sup>(8)</sup> Greater amounts of plutonium were found airborne during the oxidation of larger, ignited pieces of metal (unalloyed and delta-stabilized)--up to 0.032%. During some of these experiments, considerable quantities of molten metal leaked from large cracks in the oxide coat. The Mass Median Diameter of the particles airborne was 4.2  $\mu\text{m}$ .<sup>(9)</sup> Carter and Stewart measured the airborne release from ignited and melting metal and from falling droplets of molten metal. A value at the 95% confidence level of 0.01% from ignited and molten metal and 1% from droplets as an aerosol

of particles less than  $10\ \mu\text{m}$  AED is suggested.<sup>(10)</sup> Thus under static conditions (only disturbance is airflow under 80 cm/sec) very small quantities of aerosol in the respirable size range are generated--less than  $10^{-4}\%$ . As the amount of disturbance increases (higher airflow and flow of molten metal), the quantity of aerosol increased to 0.01 to 0.05%. If droplets are formed and molten metal falls through space spattering on impact, airborne release can increase to 1%.

### Plutonium Compounds, Powders

#### Plutonium Dioxide

Plutonium dioxide is formed when plutonium and some compounds are heated in air. The dioxide is stable and a very refractory material, particularly when prepared at high temperatures. Normally green, the color is a function of purity and particle size and varies with method of preparation.<sup>(1)</sup> Very little of the material becomes airborne in static heating-- $5 \times 10^{-6}\%$  of a powder composed of particles, 15 to  $150\ \mu\text{m}$  AED during an hour at  $900^\circ\text{C}$  by an upsweep of air at 10 cm/sec. Increasing the air velocity to 117 cm/sec increased the quantity airborne to as much as 0.56% in the apparatus used. As much as 70% of a powder placed on the stainless steel planchet could be suspended with air drawn tangentially across and upwards.<sup>(11)</sup> Mishima and Schwendiman have used a ball-milled uranium dioxide powder (30 to 70% less than  $10\ \mu\text{m}$  AED) as a simulant for plutonium dioxide in a series of experiments. From 6.8 to 38% of the uranium incorporated in various flammable materials was airborne during its burning at the base of a chimney with air at a velocity in excess of 100 cm/sec. (Configuration similar to forced draft incinerator.)<sup>(12)</sup> Under static conditions (materials made airborne by naturally occurring phenomena), only 0.05% was found airborne.<sup>(13)</sup> Approximately 80% of the uranium airborne was associated with particles less than  $10\ \mu\text{m}$  AED.

Fractional releases of dioxide powder was also measured from various surfaces at two air velocities (2.5 and 20 mph) with and without a gasoline fire. From bare, sandy soil, 0.003% of the freshly deposited dioxide powder was entrained in a 24-hr period by air having a velocity of 2.5 mph.

Approximately 50% of the material airborne was associated with particles less than 10  $\mu\text{m}$  AED. Involving the powder in a gasoline fire under these conditions increased the quantity of material airborne during the course of the fire (1 to 2 hr) to 0.5% of which 70% was associated with particles less than 10  $\mu\text{m}$  AED. An equal quantity could be entrained from the residue after the fire in a 24-hr period. Only 20% of this material was under 10  $\mu\text{m}$  AED.

Increasing the wind speed under these conditions significantly increased the quantity airborne. At 20 mph, up to 15% of freshly deposited oxide powder was entrained (40% material under 10  $\mu\text{m}$  AED) during a 24-hr period. During the course of a gasoline fire (20 to 45 min) engulfing the source material on sandy soil, 0.5% was airborne of which 60% of the material was associated with particles less than 10  $\mu\text{m}$  AED. As much as 1.0% could be entrained from the solid residue remaining from the fire in a 24-hr period.

The presence of vegetation decreased the aerodynamic entrainment of the dioxide powder to 0.005 and 1.0% at 2.5 and 20 mph respectively over a 24-hr period. Engulfing the source material in a gasoline fire released 0.01 and 3.1% at the two air velocities.

Dioxide powders are more readily entrained from smooth, hard surfaces (stainless steel)--0.1% in 24 hr at 2.5 mph (85% less than 10  $\mu\text{m}$  AED) and 5.4% in 24 hr at 20 mph (14% less than 10  $\mu\text{m}$  AED). As noted before, as much as 70% of the powder on a stainless steel surface could be made airborne by a jet of air directed into a mound of material. Burning gasoline over the surface (15 to 60 min) released 0.12 and 0.5% of the dioxide powder at the two air velocities. At the lower wind speed, 60% of the dioxide was associated with particles less than 10  $\mu\text{m}$  AED while only 7% was in this size range at 20 mph. Approximately 0.1% of the residue remaining after the fire could be made airborne in 5 hr at 2.5 mph (85% less than 10  $\mu\text{m}$  AED) and 0.7% from the residue in 24 hr at 20 mph (45% less than 10  $\mu\text{m}$  AED).

Thus very little of a finely divided, preformed, oxide powder is made airborne under static conditions (less than  $10^{-5}\%$ ). As the velocity of the

air passing over the powder increases, the quantity of powder airborne increases, although the surface from which the powder is entrained plays some part. Fractional releases ranged from 0.004% in a 24-hr period from sandy soil or soil with a vegetation cover to 0.1 to 0.5% from vegetation covered soil to 15% from sandy soil to 5.4% from stainless steel. At the lower velocities, entrainment is initially high with a gradual reduction of rate. Entrainment appears to be complete in 18 to 20 hr.<sup>(12)</sup> At the higher velocity entrainment appears to be essentially complete in 2 hr.<sup>(12)</sup> If air at a velocity of greater than 10 mph is directed upon the powder, as much as 70% can be made airborne.

The presence of heat (fire) only influences the quantity of material airborne by its effect on the surface that the dioxide powder is deposited on. Heating the dioxide powder under static conditions does not significantly change the fractional airborne release. Engulfing the dioxide powder in a gasoline fire increased the fractional releases at 2.5 mph to 0.5 and 0.01% from soil and soil with a vegetation cover. The fractional release from a stainless steel surface did not change significantly. At an air velocity of 20 mph, the fractional releases during a gasoline fire were slightly higher than the values for aerodynamic entrainment from soil, with and without a vegetation cover, but were reduced to 0.5% from a stainless steel surface.

When the dioxide powder was involved with flammable materials in fire, 0.05% was airborne under static conditions, 1% was airborne at low airflows and up to 40% could be made airborne using "forced draft."

#### Plutonium Oxalate Powder

Plutonium (IV) oxalate hexahydrate,  $\text{Pu}(\text{C}_2\text{O}_4)_2 \cdot 6\text{H}_2\text{O}$ , is a yellow-green powder which decomposes to the dioxide when heated in air.<sup>(1)</sup> The particle size of the oxide produced by calcination depends upon the conditions which the precursor and oxide are prepared.<sup>(14)</sup> Although the size of the basic oxide particles produced appear to increase with temperature,<sup>(15,13)</sup> the size of the aggregates showed no correlation to calcination temperature and indicated a Mass Median Diameter of 8 to 12  $\mu\text{m}$ .<sup>(13)</sup> Oxide particles

were 26 to 68% of the size of their precursor.<sup>(13)</sup> These measurements were made under laboratory conditions and the oxide produced under accident conditions will probably be coarser.

Mishima, Schwendiman and Radasch report fractional releases up to 1% of the source when heating either the oxalate or partially oxidized oxalate in an upswep of air.<sup>(11)</sup> The oxalate used was a moist powder with the color, consistency, and physical appearance of brown sugar. The MMD of the powder was determined microscopically to be 50  $\mu\text{m}$ . The partially oxidized oxalate was a tan, finely divided, free flowing powder composed of spheres with a MMD of 32  $\mu\text{m}$  determined microscopically. As much as 0.9% of the plutonium used in the source was made airborne during a 1-hr period at temperatures up to 1000°C and air velocities up to 100 cm/sec. Fractional releases tended to increase with temperature and air velocity. The partially oxidized material was more readily airborne with significant fractional releases under less rigorous conditions than required for fresh oxalate. Less than 0.004% of either material was airborne in the absence of heat. Both materials produced residues which could be dispersed when heated to 400°C or less, but hard cakes were formed when heated to 700°C or more.

#### Plutonium Fluoride Powder

Plutonium fluoride is stable in moist air up to about 300°C with complete conversion to the dioxide around 600°C.<sup>(1)</sup>

Mishima, Schwendiman, and Radasch reported fractional airborne releases up to 0.056% during the heating of fluoride powders to temperatures up to 1000°C in an upswep of air up to 100 cm/sec.<sup>(11)</sup> The source material was a green, finely divided free-flowing powder composed of spheres and agglomerates with a MMD of 38  $\mu\text{m}$ . Measurable fractional releases were only obtained at elevated temperatures and an air velocity of 100 cm/sec. Values measured ranged from 0.007% at 400°C to 0.056% at 1000°C. Easily dispersed residues were produced at all temperatures.

#### Plutonium Nitrate Solutions

Plutonium nitrate pentahydrate,  $\text{Pu}(\text{NO}_3)_4 \cdot 5\text{H}_2\text{O}$ , can be prepared by evaporation of plutonium nitrate solutions at room temperature and is



relatively stable at that temperature in dry or moist air. Decomposition begins around 40°C and conversion to the dioxide is rapid and complete above 250°C. (1)

Mishima, Schwendiman, and Radasch have measured fractional airborne releases during 1) low temperature air drying of shallow pools of concentrated plutonium nitrate solutions, 2) evaporation of water from dilute plutonium nitrate solutions at different heating rates, and 3) heating of the solid residues from the evaporation of shallow pools of concentrated plutonium nitrate solutions. (16) Fractional releases ranged from less than  $10^{-8}\%$  during drying at room temperature with an airflow of 100 cm/sec, to  $3 \times 10^{-3}\%$  during drying at about 100°C with the same airflow. Sampling periods ranged from 1.5 to 24 hr. Less than  $10^{-6}\%$  was airborne from the solid residues during a 24-hr period in a 100 cm/sec air stream. As much as 0.18% of the plutonium in a dilute nitrate solution was made airborne during the evaporation of approximately 90% of the water in a deep-form beaker at a rolling boil. An air sweep of 2.9 cm/sec was used to carry the airborne material from the volume of air near the top of the vessel. Lesser amounts (as low as  $3 \times 10^{-6}\%$ ) were airborne at lower heating rates. From 0.001 to 0.15% of the plutonium in the solid residues from the low temperature air drying experiments was airborne when the residues were heated to temperatures up to 1000°C in air flows up to 100 cm/sec.

Fractional releases have also been measured during the heating/burning of materials contaminated with plutonium nitrate solution. (12,17) Some experiments were conducted to determine the amount of plutonium release during the heating of sand bearing dried plutonium nitrate. Small quantities of materials were indirectly heated in stainless steel cups at temperatures up to 1000°C for 1-hr periods. Using completely dried material--sand surface was completely undisturbed after the run--up to 0.002% was found in air drawn up and around the sample at 100 cm/sec. If moisture were present, sand particles were dislodged and some of these particles entrained in the air stream. Releases under these conditions ranged from 0.0053 to 0.028%.

Higher fractional releases were found during the combustion of flammable materials contaminated with nitrate solution. From 0.005 to 0.015% was released during the burning of waste cartons containing uranium nitrate contaminated flammable materials under static conditions. Greater than 80% of the uranium airborne was associated with particles less than 10  $\mu\text{m}$  AED.<sup>(13)</sup> As much as 1% of the plutonium incorporated was made airborne during the burning of small quantities of contaminated flammables at a nominal velocity of 5.6 cm/sec.<sup>(12)</sup> When air at a higher velocity was drawn through the burning mass (in a manner similar to a forced draft furnace), the fractional release was again increased. Releases range from  $3.4 \times 10^{-4}$  to 0.1% at a nominal velocity of 50 cm/sec and 0.05 to 8% at 100 cm/sec.<sup>(12)</sup>

Fractional airborne releases of uranium nitrate (as a simulant for plutonium) were also measured from various surfaces at two air velocities, with and without a gasoline fire.<sup>(12)</sup> Aerodynamic entrainment was low from all surfaces for the nitrate solution or its solid residues at a wind speed of 2.5 mph. Values ranged from 0.005% (75% of the material airborne associated with particles less than 10  $\mu\text{m}$  AED) from sandy soil to 0.06% from an asphalt/gravel mixture of which 50% was associated with particles less than 10  $\mu\text{m}$ . Increasing the wind speed to 20 mph did not significantly alter the amount entrained from the liquid but increased the entrainment of solid residues to 1.0% from the soil (22% less than 10  $\mu\text{m}$  AED) and 2.6% from a stainless steel surface (30% less than 10  $\mu\text{m}$  AED).

Involving the nitrate solution in a fire increased the fractional airborne release. As much as 0.04% (18% less than 10  $\mu\text{m}$ ) from sandy soil, 0.1% from vegetation covered soil (80% less than 10  $\mu\text{m}$  AED), 0.24% (68% less than 10  $\mu\text{m}$  AED) from an asphalt/gravel mixture and 1.1% (40% less than 10  $\mu\text{m}$  AED) from a stainless steel surface during a gasoline fire by air with a velocity of 2.5 mph. Increasing the wind speed to 20 mph increased the fractional release from sandy soil to 0.12% (32% less than 10  $\mu\text{m}$  AED) and from stainless steel to 11.4% (35% less than 10  $\mu\text{m}$  AED).

The information on fractional releases from plutonium is not easily summarized. Some general observations can be made:

As long as the nitrate remains in an aqueous solution, fractional releases are small--less than 0.003% during evaporation during moderate temperature and airflow and less than 0.2% with the aqua boiling.

Several factors influence the fractional release of the solid residues. Very little (less than  $10^{-6}\%$ ) is released from residues at moderate temperature and airflow. If the residues are incorporated in nonflammable material like soil and surface remains intact (wind speeds of less than 2.5 mph) the release values remain low (0.005%). At high wind speeds (20 mph) fractional releases increase to around 1%. If the surface is flammable and wind speed low, fractional release remains moderate (less than 0.1%). If air is drawn through the burning mass of contaminated flammable material, fractional releases can be as high as 8 to 10%.

#### PLUTONIUM AEROSOL BEHAVIOR

##### Airborne "Mass Concentration"

The quantity of material which becomes airborne in a given volume is not necessarily the mass concentration of interest. For instance, a large solid piece of material can become airborne but will soon be deposited on the ground. It is the quantity per unit volume of plutonium as particles in the respirable size range that persist over a period of time during which they may be inhaled by individuals or transported to such locations is the concentration of interest.

Rodebush indicates the mass density of an aerosol cloud cannot differ greatly from that of air<sup>(18)</sup> ( $1170 \text{ g/m}^3$ ). Large mass concentrations encountered are due entirely to large particles that settle out rapidly and thus heavy aerosol concentrations cannot persist. Particle population as a function of time has been investigated and although initial concentrations may be high, concentrations are reduced to less than  $10^6$  particles/cm<sup>3</sup> in a few minutes. Particle populations of less than  $10^5$  particles/cm<sup>3</sup> are found even in dusty operations.<sup>(19)</sup>

Making a conservative assumption that all particles are of the largest size in the respirable size range, 10  $\mu\text{m}$  AED, and an aerosol containing  $10^6$  particles/ $\text{cm}^3$  will persist, the mass of plutonium airborne as oxide would be  $2 \text{ g/m}^3$ . Generating a monodispersed aerosol is a difficult task.<sup>(20)</sup> Some authors have offered experimental confirmation of the tendency of environmental aerosols for a "self-preserving size distribution."<sup>(21,22)</sup> The physical laws controlling the formation of particles tend to form a preferential size and distribution. Thus, a monodispersed aerosol is not possible for industrial dusts.

Swain and Haberman<sup>(23)</sup> calculated the mass density of the plutonium aerosol anticipated from a plutonium metal fire. Assuming a log-normal size distribution from 0.04 to 10  $\mu\text{m}$  plutonium dioxide with a density of 2 (due to agglomeration) with a population of  $10^6$  particles/ $\text{cm}^3$ ; they estimated the mass concentration would be  $33 \text{ mg Pu/m}^3$ . Some corroboration for a mass concentration in this range is obtained by making a rough calculation of potential airborne concentration during the Rocky Flats fire (see Loss of Control Incidents Involving Plutonium or Its Compound Section). If we accept the assumption that as much as 0.5% of the plutonium involved could have been airborne during the fire and an air space 400 x 200 x 40 ft ( $90,613 \text{ m}^3$ ) the airborne concentration is in the order of  $45 \text{ mg Pu/m}^3$ . The value would undoubtedly be high since a significant portion of the plutonium included is probably inert debris and the air volume is greater than used since the exhaust system was operative during the incident.

Castleman, et al.,<sup>(24)</sup> vaporized  $\text{PuO}_2$  and  $\text{UO}_2$  in a  $0.8 \text{ m}^3$  vessel and measured the mass concentration and size distribution of the particles of the aerosol generated. The material was vaporized in the chamber into a flowing gas stream. The geometric mean radius of the log-normal distribution ranged from 0.057 to 0.11  $\mu\text{m}$  5 min after cessation of vaporization with standard deviations ranging from 1.5 to 2.1. Theoretical mass concentration (quantity of material vaporized per volume of vessel) ranged from 0.041 to  $0.375 \text{ g/m}^3$  but measured mass concentrations range from 0.0105 to  $0.071 \text{ g/m}^3$  (from 15 to 35% of the quantity airborne). An aerosol composed of larger particles would increase the mass concentration but fewer particles would

persist due to increased agglomeration and settling velocity. Particles greater than 10  $\mu\text{m}$  AED are not respirable and do not constitute an inhalation hazard. Thus a maximum mass concentration in the neighborhood of 100 mg Pu/m<sup>3</sup> seems reasonable.

#### "Models of Mass Concentration and Particle Size Distribution Changes of Aerosol with Time"

Fish, et al.,<sup>(25)</sup> have recently reviewed theoretically based models and empirical equations by Schikarski, Davis, Koontz and Castleman that describe the reduction of airborne particulate plutonium through agglomeration and settling. All have been compared with experimental results. Models from Atomics International (Koontz) and Brookhaven National Laboratory (Castleman) used measured initial particle radii, geometric standard deviation and mass concentration plus height of vessel used and good agreement was obtained with experimental results.

Fish found all models agree that the initial high number density of particles decays rapidly during the first few hours and less rapidly for a day or so. Both the Atomics International and Brookhaven models include terms for addition of particles and removal by Brownian motion, gravitational agglomeration and settling and wall plating. Although such rigorous treatment may not be possible for accident situations under consideration here, such models are available if desired.

#### Loss of Control Incidents Involving Plutonium and Its Compounds

Various incidents have occurred involving plutonium and its compounds ranging from spread of contamination to major fires.<sup>(26)</sup> In no case have hazardous quantities of plutonium been released to the environment. Three of the incidents were very serious in nature and involved different forms of plutonium.

In November 1959, an estimated 500 mg of plutonium was blown through the open door and operating holes of a cell during decontamination of an evaporator. The explosion was attributed to inadvertently using nitrate organic cleaning agent. Although nearby buildings, vehicles, the roadway

and ground were contaminated, air samplers in the area did not indicate air concentrations above acceptable limits. Thus, although air concentrations near the contamination may have exceeded limits for a short period of time, air concentrations exceeding established limits could not leave the site boundaries even under these rigorous conditions.

Detonation of nitrated exchange resin initiated a fire that destroyed a plutonium purification facility in Richland, Washington in November 1963. Many kilograms of plutonium as nitrate were involved. The integrity of the vessel and glove box were destroyed and material could escape through a partially open door. Although alpha contamination was widespread throughout the facility, air samplers located at the site boundaries within one-quarter mile did not indicate air concentrations exceeding established limits. The alpha contamination in this instance appeared to be associated with soot and the great number of soot particles generated in this incident may have effectively removed a large part of any plutonium which was airborne.

The most serious and significant incident involving plutonium to date was the fire in a major plutonium fabrication facility at Rocky Flats, Colorado in May 1969. Products of a fire in one area clogged the exhaust filters of one of three exhaust systems. Flammable vapors passed into other areas. Ultimately, a significant portion of the facility was involved. The supply fans operated during the initial phase of the fire and loss in negative pressure allowed back diffusion into office areas. Hundreds of kilograms of plutonium as metal and compounds was involved with a significant quantity in unknown form involved with the equipment (Material Unaccounted For). Only 200  $\mu\text{Ci}$  of airborne material (0.003 g) was released through a damaged exhaust system.<sup>(27)</sup> Based on the author's personal observation and data, a maximum of 0.5% of the plutonium may have been airborne within the facility. This value was derived by making the highly conservative assumption that all contamination measured on the ceiling, walls and floor of all contaminated areas of the facility and all surfaces outside the enclosure was due to airborne material. The estimate does not include the negligible amounts of plutonium found in the water collected from extinguishment nor the unknown quantities in the exhaust system. The

vast majority of the plutonium used to obtain this estimate was measured as floor contamination in the immediate fire area and is probably debris which fell or was washed from the enclosure during extinguishment.

### Resuspension

Once into the atmosphere, the plutonium is acted upon by climatological phenomena and ultimately approaches the surface. If the plutonium particles are deposited on a surface, they still can constitute an inhalation hazard. Although gross resuspension is the primary concern, Healy enumerates other modes (direct inhalation) by which deposited plutonium can be an inhalation hazard.<sup>(28)</sup> As part of the hazards evaluation for plutonium release from a weapon involved in an accident, a value of  $100 \mu\text{g Pu/m}^2$  was calculated for safe, lifetime occupancy. On the basis of measurements obtained from experiments involving nonnuclear detonations of weapons, a value of  $1000 \mu\text{g Pu/m}^2$  was established as safe for lifetime occupancy (weather being the sole resuspension force) although a value of  $3500 \mu\text{g/m}^2$  was determined as safe.<sup>(29)</sup> Resuspension factors in the range of  $7 \times 10^{-7}$  to  $7 \times 10^{-5} \text{ m}^{-1}$  were found.<sup>(30)</sup> Resuspension factors in the order of  $10^{-13} \text{ m}^{-1}$  were reported for the thoroughly weathered material.<sup>(31)</sup> Most recently, Sehmel measured resuspension of 0.001 to 1% of particles freshly deposited on an asphalt road per pass using a car or 3/4 ton truck. Higher resuspension corresponded to higher vehicle speed. A marked decrease of resuspension was observed with weathering.<sup>(32)</sup>

The resuspension of plutonium particles has also been measured. Jones and Pond<sup>(33)</sup> measured the resuspension from paper covered, plastic covered and waxed linoleum surfaces. Particles ranged from 0.4 to 60  $\mu\text{m}$ . Higher resuspension factors were obtained from surfaces contaminated by oxide than nitrate. When the plutonium was applied as a solution, absorbent paper surfaces gave lower resuspension factors than sound impermeable surfaces. Higher resuspension factors correlated with increased activity. Resuspension factors in the order of  $2 \times 10^{-8} \text{ m}^{-1}$  were found with no movement and  $10^{-6}$  to  $10^{-5}$  with walking.

Glauber<sup>(34)</sup> measured resuspension of aged material from concrete surface in an old production facility. Factors for plutonium oxide ranged

from  $10^{-6}$  to  $10^{-7} \text{ m}^{-1}$  for no movement before the test,  $10^{-3} \text{ m}^{-1}$  for no movement after the test,  $10^{-3} \text{ m}^{-1}$  with air circulation over the surface, to  $10^{-2} \text{ m}^{-1}$  when a dolly was run over the surface with air circulation.

Thus in resuspension, the form of plutonium deposited, the surface, the entraining force and age of the material all play a role. Particles entrain more readily than solutions. Greater amounts can be resuspended from hard, impermeable surfaces. Direct transfer of momentum is a more effective mechanism for resuspension than aerodynamic forces. Freshly deposited materials are more readily entrained than weathered material.



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APPENDIX G

PLUTONIUM DECISION LEVELS

## APPENDIX G

### PLUTONIUM DECISION LEVELS

There are several studies, reports, etc., which characterize the accident or accident parameters for reactors. Additionally, recognized authorities such as FRC, ICRP, NCRP, etc., have developed population dose limits which apply to the emission from reactor facilities. These have been applied in the reactor siting guide.

Similar guidance does not appear to be available for plutonium or plutonium fuel fabrication plants. In fact, Wright Langham noted<sup>(1)</sup> that guides need to be developed for minimum acceptable action in the event of plutonium accidents.

#### A. SURFACE CONTAMINATION

In an effort to arrive at some useful numbers in the absence of any established levels, an attempt has been made to orient existing limits, guides, detection levels, etc., for surface contamination to determine if meaningful relationships exist. Proposed plutonium surface contamination decision levels and their relationships to existing guidance are shown in FigureG-1. The definition of "contaminated" was chosen as  $10 \text{ nCi/m}^2$ . This is equivalent to approximately  $200 \text{ d/m}/100 \text{ cm}^2$  or "detectable" limits for alpha contamination survey instrumentation. A comparison of various parameters for four isotopic mixtures is shown in TableG-1. Mixture III is the mixture discussed in Section V-A. The Plutonium Surface Contamination Decision Levels shown in FigureG-1 are appropriate for all mixtures.

#### B. POPULATION DOSE DECISION LEVELS

Several references were reviewed to characterize the annual dose received from natural background radiation. A summary of values found is shown in TableG-2.

Published Guidance

Proposed Decision Levels

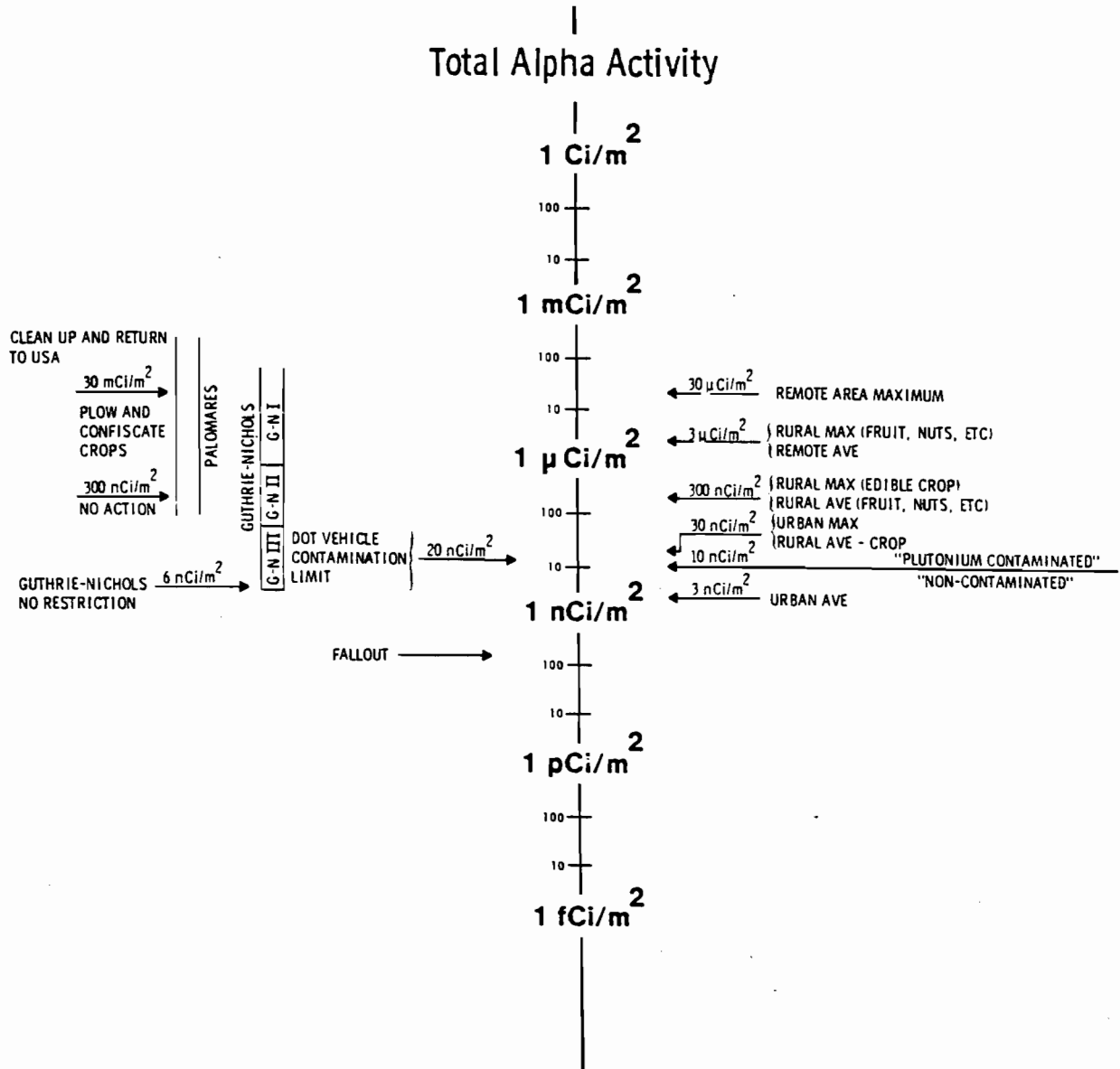


FIGURE G-1. Plutonium Surface Contamination Levels

TABLE G-1. Comparison of Isotopic Mixtures

Isotope	wt%	Ci/g Isotope	Ci/g Mixture	Activity %	MPC <sub>A</sub> Mixture μCi/cm <sup>3</sup>
<u>MIXTURE I</u>					
Pu-239	95	0.061	0.06	86	(S 6x10 <sup>-13</sup> I 1x10 <sup>-11</sup> )
Pu-240	5	0.227	0.01 0.07	14	
<u>MIXTURE II</u>					
Pu-239	2.85	0.061	0.002	87	(S 6x10 <sup>-13</sup> I 1x10 <sup>-11</sup> )
Pu-240	0.15	0.227	0.0003	13	
U-238	96.3	3.33x10 <sup>-7</sup>	3.21x10 <sup>-7</sup>		
U-235	0.7	2.14x10 <sup>-6</sup>	1.49x10 <sup>-8</sup> 0.0023		
<u>MIXTURE III</u>					
Pu-238	1.9	16.8	0.319	2.3	(S 3x10 <sup>-11</sup> I 1x10 <sup>-8</sup> )
Pu-239	63	0.061	0.038	0.3	
Pu-240	19	0.227	0.043	0.3	
Pu-241	12	114.0	13.7	97.1	
Pu-242	3.8	0.004	0.0002 14.1	---	
<u>MIXTURE IV</u>					
Pu-238	0.06	16.8	0.010	2.4	(S 3x10 <sup>-11</sup> I 1x10 <sup>-8</sup> )
Pu-239	1.9	0.061	0.0012	0.28	
Pu-240	0.57	0.227	0.0013	0.31	
Pu-241	0.36	114	0.4104	97	
Pu-242	0.11	0.004	4.4x10 <sup>-6</sup>	---	
U-238	96.3	3.33x10 <sup>-7</sup>	3.2x10 <sup>-7</sup>	---	
U-235	0.7	2.14x10 <sup>-6</sup>	1.5x10 <sup>-8</sup> 0.423	---	
S - Soluble					
I - Insoluble					

TABLE G-2. Annual Average Whole Body Dose  
from Natural Background Radiation  
(mrem)

	<u>Beir</u> (2)	<u>Unsear</u> (3)	<u>Table II-4</u> <u>ORP/CSD 72-1</u> (4)
Cosmic-External	44	31	45
Terrestrial External	40	44	60
Terrestrial Internal	<u>18</u>	<u>18</u>	<u>25</u>
Total	102	93	130

Based on the values presented in ORP/CSD 72-1, the annual average dose to the bone and to the lung from natural background radiation would be as shown in Table G-3.

TABLE G-3. Average Annual Bone and Lung Dose  
from Natural Background Radiation

	<u>Bone</u> <u>(Endosteal Cells)</u>	<u>Lung</u>
Cosmic External	45	45
Terrestrial External	60	60
Terrestrial Internal	<u>47</u>	<u>25</u>
Total	152 mrem/yr	130 mrem/yr

On the assumption that the exposure limits appropriate to the general population would be in proportion to natural background radiation dose to the various organs of interest, 130 mrem/yr to whole body and lung, and 152 mrem/yr to the bone, the general population exposure limits for man-made radiation other than medical would be as shown in Table G-4.



TABLE G-4. Derived Population Exposure Limits for Manmade Radiation(a)

<u>Gen. Pop. Limit</u>	<u>Whole Body</u>	<u>Derived Organ Limits</u>	
		<u>Bone</u>	<u>Lung</u>
Maximum Individual (mrem/yr)	500	585	500
Average Individual (mrem/yr)	170	200	170
Average Individual (rem/50 yrs)	or 8.5	10	8.5

a. excluding medical exposure

The assumption is generally consistent with the position taken by the NCRP<sup>(5)</sup> in their report number 39, wherein they say:

"To limit the radiation-induced incidence of leukemia and other serious consequences in the whole population, a dose limit comparable with that for the genetic case is desirable. The two limits have been numerically equated here by extending the organs of interest from the gonads to the whole body. It is expected that this will be a practical simplification, essentially requiring only one calculation of average population dose in most circumstances. It is also expected that the dose limit of 0.5 rem (500 mrem) per year for any critical organ of an individual member of the public, combined with the average population dose limit of 0.17 rem (170 mrem) per year for the critical organs, will have the effect of controlling the actual population exposures well below the stipulated limits. No specific evidence can be established that would seem to warrant further reduction of average or individual dose limits for members of the public, at this time. The low dose and low dose rate of the radiation exposure of the population still provide adequate safety factors. The idealized objective of having public exposure, in addition to that from natural radiation, as close to zero as is reasonably possible, of course, remains. In this connection, it must be pointed out again that although these limits do not include the contributions of radiation from the healing arts, there is a clear intention to encourage their reduction to the lowest practicable levels."

The derived "limits" above are for all sources other than natural background and medical exposure. For the purpose of this study, it is assumed that on the average no more than 1/10 of this 5-year dose commitment should result from fuel cycle activities such as fuel fabrication; therefore, the average person in the population in the vicinity of a fuel fabrication plant should incur a dose commitment no greater than the Population Dose Decision Levels shown in Table G-5.

TABLE G-5. Population Dose Decision Levels

	<u>50-year Dose Commitment (rem)</u>	<u>Incremental Change in Dose Rate from Sustained Exposure (mrem/yr)</u>
Bone (soluble)	1	0.8
Lung (insoluble)	0.85	0.7

The proposed decision levels are about 1/750 of the dose which would be received by the bone of an individual exposed for 50 years to soluble material at the occupational MPC, and about 1/880 of the dose which would be received by the lung of an individual exposed for 50 years to insoluble material at the occupational MPC.

On this basis, the MPC's appropriate to the long-term exposure of the general population to the isotopic mixtures assumed in Table G-6.

TABLE G-6. Derived MPC<sub>a</sub>'s

<u>Mixture</u>	<u>Soluble Material MPC<sub>a</sub> (bone) in <math>\mu\text{Ci}/\text{cm}^3</math></u>	<u>Insoluble Material MPC<sub>a</sub> (lung) in <math>\mu\text{Ci}/\text{cm}^3</math></u>
I LWR Pu	$8 \times 10^{-16}$	$1 \times 10^{-14}$
II LWR Fuel 3% Pu	$8 \times 10^{-16}$	$1 \times 10^{-14}$
III Hi Exposure LWR Pu	$4 \times 10^{-14}$	$1 \times 10^{-11}$
IV Hi Exposure LWR Fuel 3% Pu	$4 \times 10^{-14}$	$1 \times 10^{-11}$

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