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



Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

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FOREWORD

The Atomic Energy Commission, through Fuels and Materials, Directorate of Licensing, established a contract with the Battelle-Northwest Laboratory to conduct a study on "Considerations in the Assessment of the Consequences of Effluents from Mixed-Oxide Fuel Fabrication Plants." The results of this study are contained in this report.

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

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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

A plutonium fuels fabrication plant some 10 to 15 years in the future was characterized. The "reference facility" manufactures UO_2 - PuO_2 fuel for light water reactors and has a design capacity of 1 metric ton (MT) of fuel per day. The fuel contains 2-4 wt% PuO_2 in natural or depleted UO_2 . 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 Yankee 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.

The "reference plant" possesses design features consistent with the minimum criteria for new AEC plutonium facilities. In addition to the glovebox filters, the building final filter system has two stages of high efficiency filters.

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

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 ICRP 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 (TGLM).

Source terms are 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 5 μg . 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 was 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. They were derived by propagating accident occurrence and equipment failure rate statistics from fuel fabrication facilities and from related industries. Care was exercised in discerning the difference between the probability of events leading directly to releases and the probability of events which require the simultaneous occurrence of other events in order to cause a release.

In the final analysis, an attempt is made to quantify the risks a plutonium fuel fabrication facility imposes on the environment. Due to time limitations, the information on occurrences and failure rates were not exhaustive. For this reason and due to the nature of statistics, the results developed in this study should be viewed as preliminary. If these results are "cast in concrete" and the limited nature of their origins forgotten, they have dubious value.

The 50-year dose to the critical organs of an individual at 1000 meters from the facility was estimated for the 5 μg annual release rate of the mixture of plutonium and americium. Using the ICRP Publication 2 lung model and CAMS atmospheric dispersion model, the 50 year dose to the lung (insoluble) and to the bone (soluble) from inhalation of the mixture continuous 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. Using the ICRP Publication 2 lung Model and the USAEC Regulatory Guide 1.3 for atmospheric dispersion (8 hour 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} per year (insoluble) and 10^{-3} per 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 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 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.

The main chemical contaminants from the facility will 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.

III. SCOPE

Siting guides are needed for all of the facilities connected with the fuel cycle. With the emphasis that the Commission and the public are placing on plutonium, it was concluded that this study should deal 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 as a second phase would be relatively simple since much of the effort would overlap. The development of siting information for fuel reprocessing would also be aided by much of the information presented in this report.

The effects of effluent releases resulting from in-plant accidents as well as normal operation are considered. Both radioactive and nonradioactive effluents are reviewed, but the emphasis has been placed on plutonium because it appears to be the most limiting consideration in plant siting.

A wide variety of plutonium containing fuel types have been proposed. These include oxides, carbides, nitrides, carbonitrides, borides, sulfides, metal and various alloys. Each of these involve 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. It is felt that 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. We anticipate, however, that portions of the 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 for HTGR's will be in the form of $\text{PuO}_2\text{-ThO}_2$ or PuC-ThC . It is felt that plutonium fuel requirements for HTGR's will be minimal for at least the next decade. Additional credibility for these

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}Pu increases with fuel exposure, the specific activity and therefore the radiological hazard of a unit quantity of plutonium increases with fuel exposure.

Within the next 10 to 15 years, the bulk of the plutonium used for mixed oxide fuel fabrication will be obtained from reprocessed uranium fuel with small quantities coming from recycled plutonium. The recycled amounts are small mainly because of the rapid growth of the nuclear industry projected for the next several decades and the time lag between successive recycle. For this reason, the average plutonium isotopic mixture seen at the fabrication plant will be only slightly higher in heavier isotopes of plutonium than the plutonium isotopic mixture from an all uranium irradiation.

For the dose calculation in this study, a specific isotopic distribution of plutonium was chosen as the "reference mixture." It was obtained from a Yankee fuel sample discharged at about 35,000 MWd/MT.⁽¹⁾ 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}Pu released) in addition to the curves for the reference mixture which are 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.

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>
^{238}Pu	1.9
^{239}Pu	63.3
^{240}Pu	19.0
^{241}Pu	12.0
^{242}Pu	3.8

The plutonium will also contain some ^{241}Am (from the beta decay of the 13 year half-life ^{241}Pu). Approximately 5% of the ^{241}Pu will decay to ^{241}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 length of the cylinder is not limited and it 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 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. It is assumed that the plutonium will be received in the form of plutonium nitrate solutions that must be converted to PuO_2 . However, it is noted that the plutonium may be received in the form of 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 PuO_2 , followed by mechanical mixing with UO_2 to obtain $\text{PuO}_2\text{-UO}_2$ powder and (2) co-precipitation 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

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

	PuO ₂		FBR Fuel (d)		LWR Recycle Fuel (e)	
	Pu Mass	UO ₂ -PuO ₂ Mass	Pu Mass	UO ₂ -PuO ₂ Mass	Pu Mass	UO ₂ -PuO ₂ 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
Optimally 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% ²³⁹Pu. The presence of ²⁴⁰Pu will increase the safe masses (e.g., 20 wt% ²⁴⁰Pu will more than double the safe mass of optimally moderated LWR recycle fuel).
- c. Powder density = 5.6 g/cm³, pellets at theoretical density.
- d. Assumed to be 31 wt% PuO₂ - 69 wt% U_(nat)O₂.
- e. Assumed to be 4 wt% PuO₂ - 96 wt% U_(nat)O₂.

only for small batch fuel preparation. It is not certain how much difficulty will be involved in scaling up this process, therefore, it has not been considered in this analysis.

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

(3) Scrap Recovery. Whenever possible oxide scrap will be recycled through the process without chemical processing. For scrap and wastes where this is not possible, a typical recovery process is leaching, or dissolution, followed by reduction and ion exchange yielding $\text{Pu}(\text{NO}_3)_4$.

b. Reference Process

In order to allow detailed investigation in the study a specific process was chosen for examination. The process selected has been called the "reference process." It includes conversion of plutonium nitrate to PuO_2 , followed by pelletizing of the mechanically mixed $\text{PuO}_2\text{-UO}_2$. A flow diagram for the reference process is given in Figure 1. A tabular description of the characterized 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. There are several additional considerations 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.⁽⁴⁾ of a 1 MT/day.

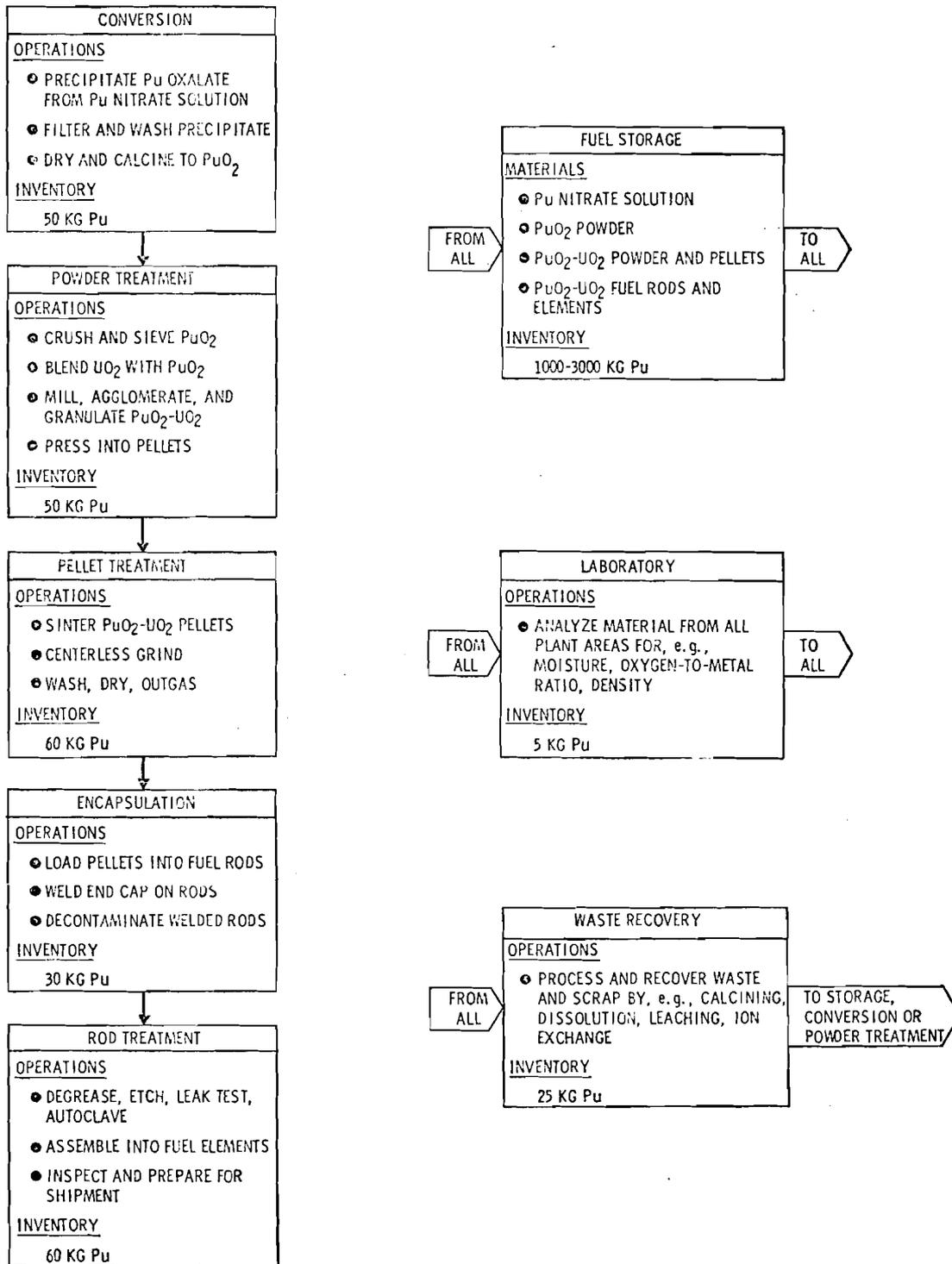


FIGURE 1. Area Flow Diagram for Model Process

PuO₂-UO₂ 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.⁽⁵⁾ 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.

A third possibility would be to employ totally remote, heavily shielded fabrication operations. If this method is used, personnel exposure considerations will 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

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, which for the purpose of this study shall be designated the primary barrier, is considered to include 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. Obviously this required system plays an important role in minimizing release of material to the environment.

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

Minimizing the fuel inventory which is at risk is another design feature. It includes decoupling various steps in the process relative to a given accident mechanism either by means of 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 feature is referred to as a secondary barrier.

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

1. The Primary Barrier

For purposes of this study, it is assumed that the tankage and piping systems 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 an 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.⁽⁶⁾ 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⁽⁷⁾ and therefore has a transmission factor of 5×10^{-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 5×10^{-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,⁽⁸⁾ 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.

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 1×10^{-7} .⁽⁹⁾ 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.

	<u>Removal Efficiency</u>	<u>Transmission Factors</u>	
		<u>Specific</u>	<u>Aggregate</u>
First Stage	99.9%	1×10^{-3}	1×10^{-3}
Second Stage	99%	1×10^{-2}	1×10^{-5}
Third Stage	94%	6×10^{-2}	6×10^{-7}
Fourth Stage	83%	1.7×10^{-1}	1.0×10^{-7}

For purposes of this study, the material present in the intact gloveboxes or within vented piping systems will be 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*	6×10^{-7}

The room air filtration system:

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

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

A characteristic of filter media which makes the foregoing transmission factors further 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 inch pressure drop for a 12 inch by 12 inch by 6 inch deep filter.⁽¹⁰⁾ Although a pressure drop of 3 inches is not exces-

sive in some installations, it is regarded as a nominal pressure drop which when exceeded would call for a filter change. It is likely that the efficiency is 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 inches 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 ($2000 \times 1 \times 10^{-5} \times 100$ filters). This would indicate that there is a realistic upper boundary to the amount of fuel material that can pass a HEPA filter.

While the above example cannot be totally supported, based on experience it appears to 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⁽¹¹⁾ (with one notable exception which is not a mixed oxide plant) are in the range of 1 to 75 $\mu\text{Ci}/\text{year}$.

Assuming a nominal flow rate of 10^5 cfm, the 75 μCi value is equivalent to an average release concentration of 5×10^{-14} $\mu\text{Ci}/\text{cm}^3$. The reporting of "less than" release values of 1×10^{-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, it can be inferred, within the present plutonium analytical capabilities of the industry, that 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.⁽¹²⁾ 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 those elements that are required for safe shutdown 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 that are required to remain functional for continued safe operation remain functional 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⁽¹²⁾ will be hardened to withstand the effects of the AEC Regulatory Model Tornado.⁽¹³⁾

c. Flood

The facility either is located such that water from the Probable Maximum Flood (PMF) 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.⁽¹⁴⁾

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⁽¹⁵⁾ the filters are required to withstand 180°F continuously and 700°F for 5 minutes.

Several tests have been conducted on the effects of elevated temperatures on high efficiency filters.^(15,16,17,18) 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.

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)								
	1-3	4-7	8-12	13-18	19-24	25-31	32-38	39+	ALL
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	0.8
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 $\bar{\chi}/Q'$ 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 $\bar{\chi}/Q'$ values that might occur at any site. Comparisons with independently calculated $\bar{\chi}/Q'$ values⁽²⁴⁾ has shown that the CAMS curves may be used as realistic but conservative estimates of $\bar{\chi}/Q'$ 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 $\bar{\chi}/Q'$ values.

In order to properly interpret the $\bar{\chi}/Q'$ 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.⁽²⁵⁾ For the United States

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.⁽²⁸⁾ 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/m².

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 10³ 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/m²/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 χ is the air concentration at time t , χ_0 is the initial air concentration, and Λ is the washout coefficient.⁽²⁹⁾

Experimental and theoretical evidence has shown the washout coefficient is about 10^{-4} to 10^{-3} sec^{-1} for uranium particles that have median mass diameter of between $6 \mu\text{m}$ and $15 \mu\text{m}$, respectively.⁽³⁰⁾ 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.^(30,31) 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.⁽³²⁾

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.

TABLE 9. Dose Parameters Used in Calculation Models

Nuclide	Half Life, Yr.	Activity Ci/g of Nuclide	Reference Mixture, wt%	Activity Ci/g of Mix	Activity in Mix (β, α), %	Dose Conversion Factor (b)		
						($\frac{\text{rem}}{\mu\text{Ci}\cdot\text{y}}$) BONE (c)	ILM (c)	($\frac{\text{rem}}{\mu\text{CT}}$) LUNG TGLM (d)
Pu-238	86.	17.5	1.9	0.33	2.4	750	500	2100
Pu-239	2.44×10^4	0.0616	63.	0.039	0.28	720	470	2000
Pu-240	6.58×10^3	0.227	19.	0.043	0.31	720	470	2000
Pu-241	13.2	113	12.	13.6	97.	37	0.47	1.8
Pu-242	3.79×10^5	0.00391	3.8	0.00015	0.0011	570	450	1900
Am-241	458.	3.25	0.60	0.020	0.14	750	500	2100
Reference Mixture				14. (α, β) 0.43 (α only)		59	16	55

- 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.

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

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:

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

$$P_s = b Q' T (E/Q) = b Q (E/Q)$$

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

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, $\text{rem per } \mu\text{Ci}$ in organ
 P_s - quantity inhaled, μCi
 λ_e - effective elimination rate constant, d^{-1}
 t - dose time, d
 b - ventilation rate for standard man, cm^3/sec
 $b = 350 \text{ cm}^3/\text{sec}$ (8 hour working rates)
 $= 230 \text{ cm}^3/\text{sec}$ (24 hour daily rate)
 Q' - atmospheric release rate, Ci/sec
 T - duration of release exposure, sec

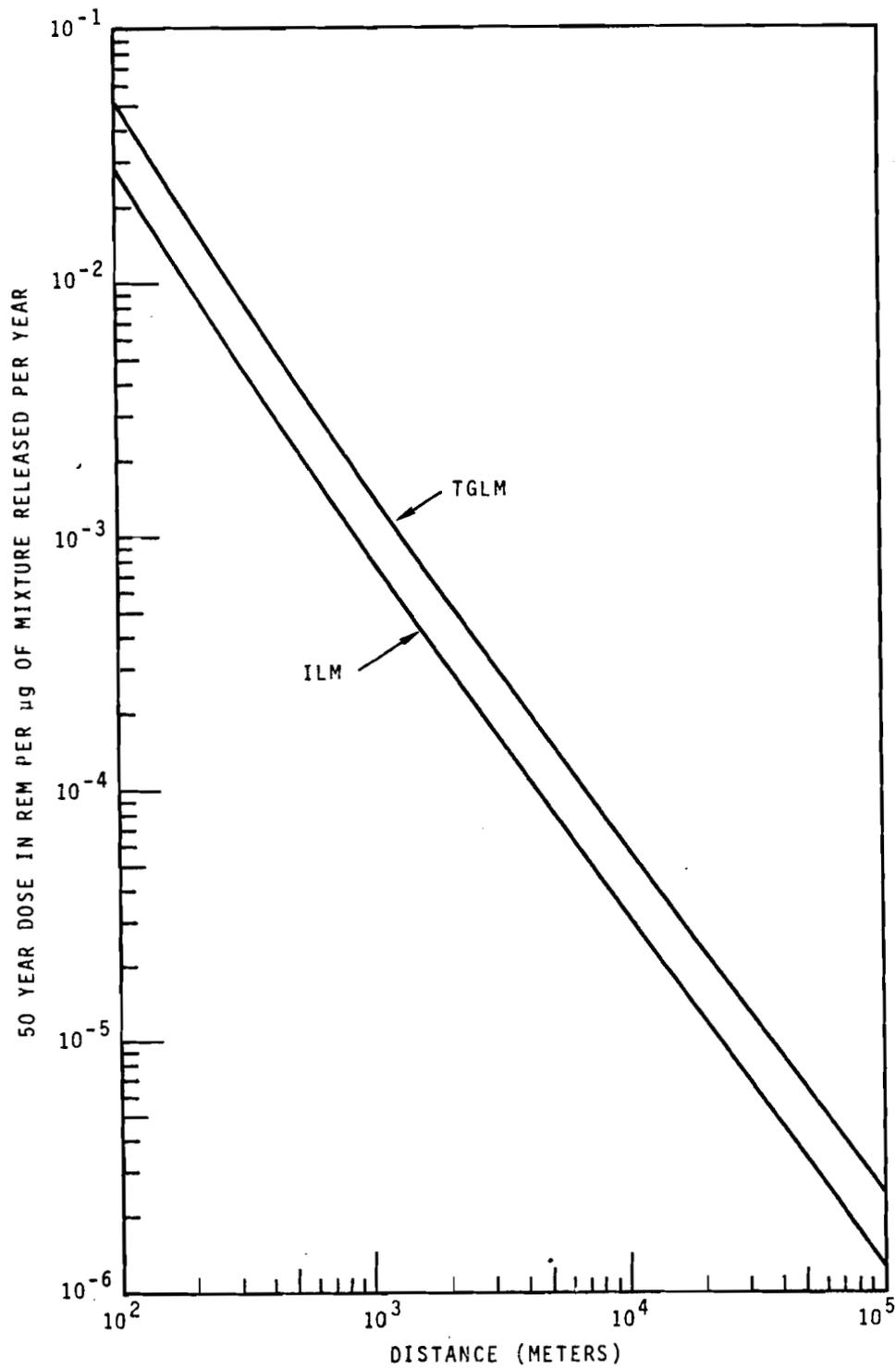


FIGURE 5. Comparison of the Dose to Bone from Inhalation of a Continuous Release of the Reference Mixture (Soluble) Using the Two ICRP Recommended Lung Models, Ground Level Release, Maximum Sector Curves

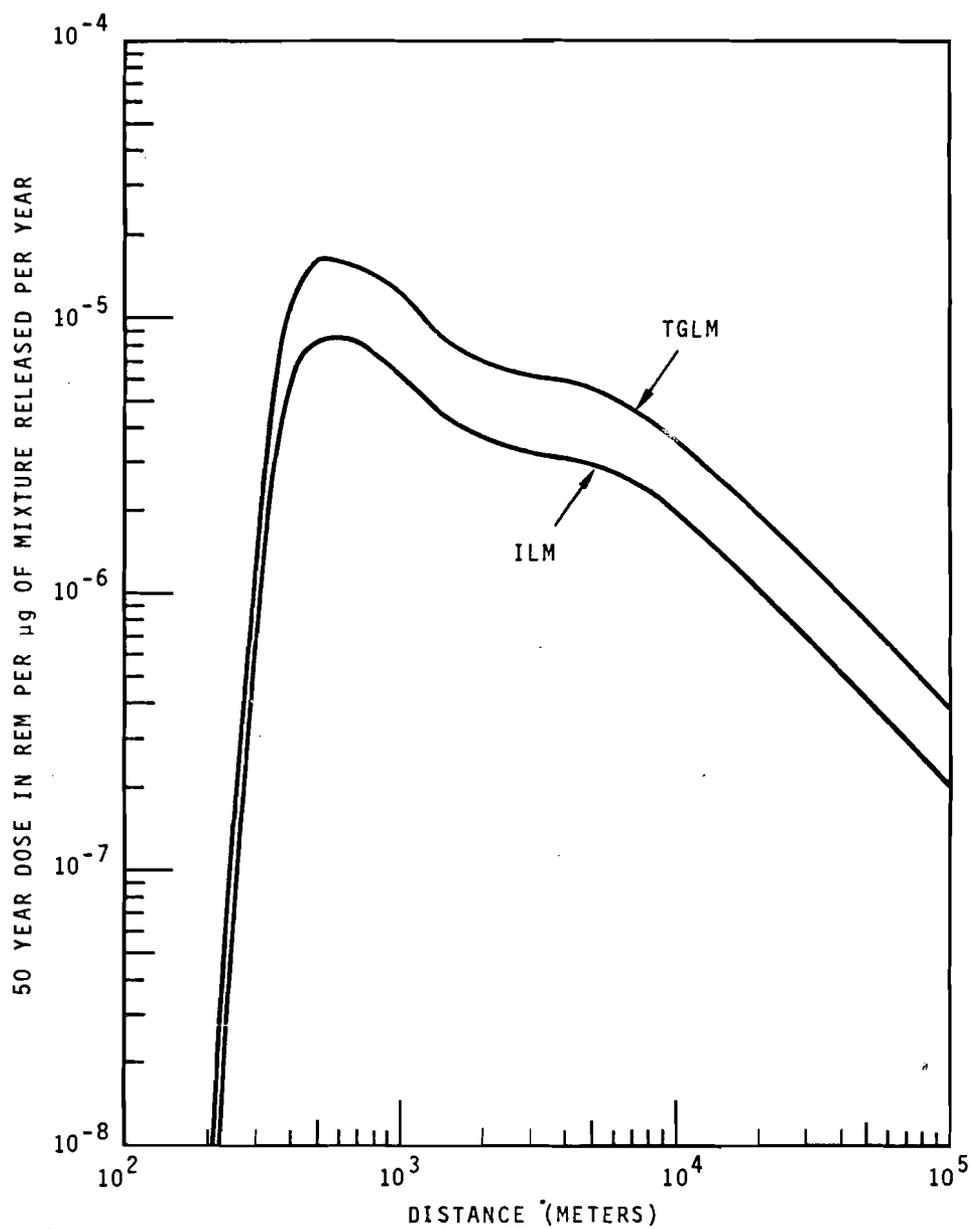


FIGURE 6. Comparison of the Dose to Bone from Inhalation of a Continuous Release of the Reference Mixture (Soluble) Using the Two ICRP Recommended Lung Models, Release Height 100 m, Maximum Sector Curves

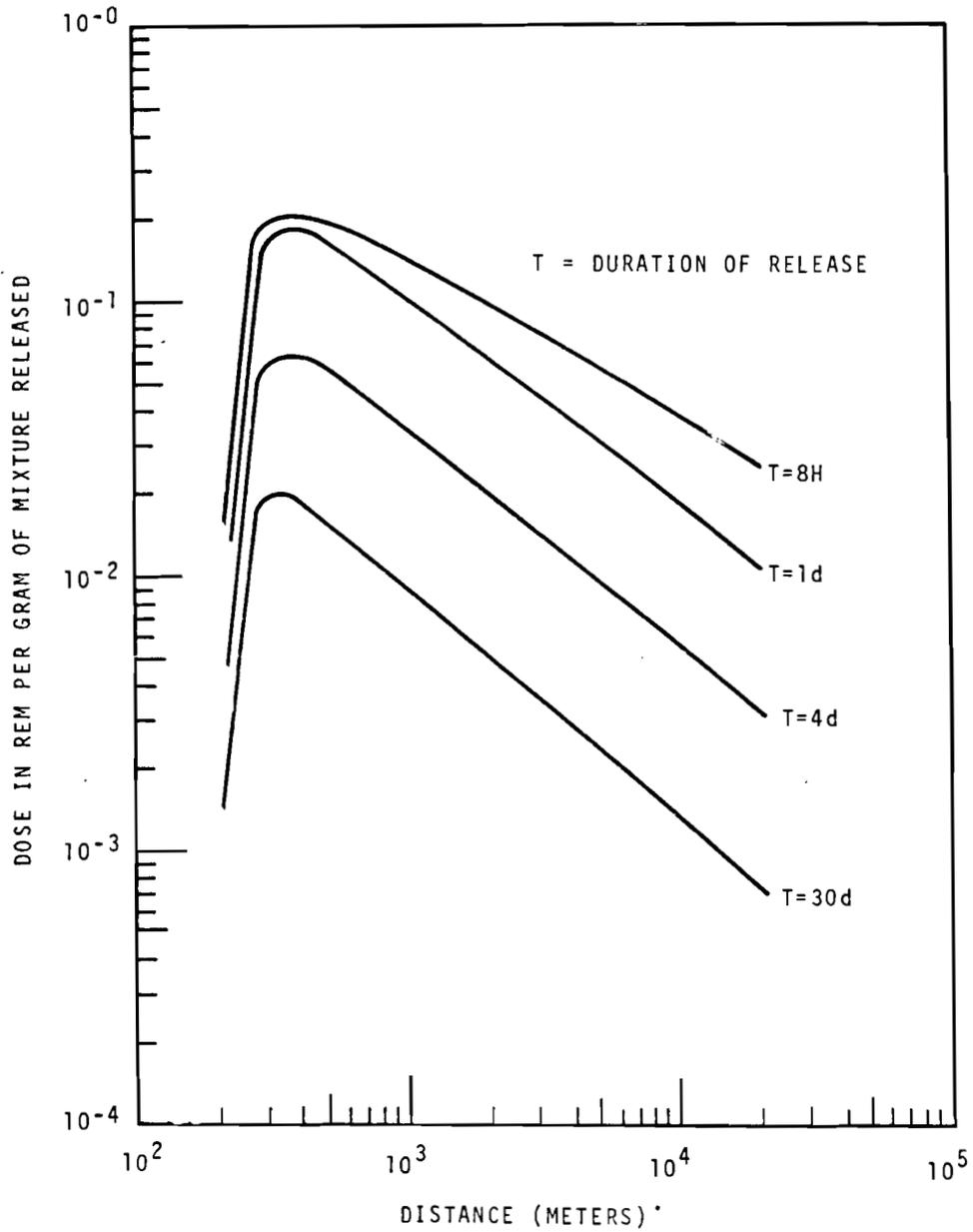


FIGURE 15. Dose to Lung from Acute Inhalation of the Reference Mixture of Plutonium and Americium, Release Height 100 m

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.

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.⁽⁵³⁾ 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.

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 10 summarizes these calculations. Even though experimental evidence⁽⁵⁴⁾ 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 2 was used for correcting the absorbed neutron dose to rem.^(55, 56, 57) 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.⁽⁵⁷⁾

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 6×10^6 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 10. Prompt Gamma and Neutron Dose to an Individual as a Function of Distance from a Criticality (10^{18} Fissions)

<u>Distance, m</u>	<u>Gamma Dose^(a) Rem</u>	<u>Neutron Dose Rem</u>	<u>Total Dose Rem</u>
10^2	4.6×10^{-1}	1.1	1.6
10^3	4.6×10^{-3}	1.1×10^{-2}	1.6×10^{-2}
10^4	4.6×10^{-5}	1.1×10^{-4}	1.6×10^{-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.⁽⁵⁸⁾ 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 - 3×10^{-5} g for two HEPA filters and 3×10^{-3} g for one HEPA filter, Particle Size -

TABLE 12. 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	1.1×10^1
10^3	3.1×10^{-2}	2.0×10^{-1}
10^4	2.2×10^{-4}	4.3×10^{-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.⁽⁶²⁾ 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.⁽⁶³⁾ By calculation, a total of 10^6 plutonium particles with an aerodynamic equivalent diameter (AED) of 10 μm 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 μm 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%.⁽⁵⁸⁾ 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.⁽⁶⁴⁾ 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 μm AED PuO_2 particle is formed by a 41 μm drop of nitrate solution of 150 g plutonium/liter concentration. Each particle contains approximately 2×10^{-10} g Pu. To generate an airborne concentration of 20 mg/m^3 , 10^8 particles/ 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%.⁽⁵⁸⁾ 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 PuO_2 powder prior to the 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 PuO₂ 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 PuO₂ 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.⁽⁶⁵⁾ 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.⁽⁶⁶⁾ Most of the plutonium powder for the operation is assumed to be in the respirable range (a PuO₂ 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 10⁴ 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 - 10⁻²g 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 13 and in Appendix C. 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 C. 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 13. 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 ^(a)
	Partial disruption of molten metal, airflow greater than 100 cm/sec	1.0 ^(a)
	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
•	<u>Explosion</u>	
	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 ³ after 10 minutes.	
•	<u>Other</u>	
	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 14. 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⁽⁷⁰⁾ and

TABLE 14. Frequencies of Major Accidents

<u>Event</u>	<u>Probable Frequency Of Occurrence Per Plant - Year</u>	<u>Range</u>
Tornado	6×10^{-4}	$4 \times 10^{-3} - 6 \times 10^{-6}$
Earthquake Intensity IX	2×10^{-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	2×10^{-4}	$4 \times 10^{-4} - 4 \times 10^{-5}$

Dunlap and Wirdner⁽⁷¹⁾ 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 3.62×10^{-3} . The probability of a tornado in the western United States, based on a study by Smith and Mirabella⁽⁷²⁾ is 1.3×10^{-6} /year. All locations in the U.S. fall within that range. The average for the contiguous 48 states is 6.0×10^{-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 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.⁽⁷²⁾ 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⁽⁷³⁾ analyzed the aircraft hazard for nuclear power plants. They determined that the probability of a strike per year varied from 1.4×10^{-5} for distances greater than 5 miles from an airport to 3.4×10^{-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 was 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.⁽⁷⁴⁾ 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.⁽⁷⁵⁾ This results in a probability of 4×10^{-4} major fires per plant per year. Recognizing the difference in industry characteristics, a lower range value of 2×10^{-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⁽⁷⁶⁾ has estimated the seismic risk in the various regions of the country. Gutenberg and Richter⁽⁷⁴⁾ 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⁽⁷⁸⁾ and a book by Wiegel.⁽⁷⁹⁾ 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 2×10^{-5} /year.

In California, the probability of a Richter magnitude 7.5 earthquake is 0.076/year.⁽⁷⁹⁾ 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⁽⁸⁰⁾ summarized current knowledge on the subject and concluded that the probability of a 2×10^7 kg meteorite impacting an area is $10^{-12}/\text{km}^2\text{-yr}$. 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 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.⁽⁸¹⁾ 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.⁽⁸²⁾ 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.⁽⁸³⁾ 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 15 and Table 16. A discussion of the development of some of these values follows. The estimated occurrence rate of a criticality in a fuel plant is also included in Table 15. 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.⁽⁸⁵⁾

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.⁽⁸⁶⁾ 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,⁽⁸⁷⁾ the frequency of a resin column fire and explosion can be developed.

Prior work,⁽⁸⁸⁾ 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 μ g) for this incident.

TABLE 15. Estimates of the Occurrence Frequencies and Failure Rates of Major Equipment

	<u>Failures or Events Per Year</u>
Resin Columns ⁽⁸¹⁾	<0.1
Dry Glovebox Operations ⁽⁸¹⁾	<0.1
Autoclave ⁽⁸¹⁾	<0.01
Sintering Furnaces ⁽⁸⁴⁾	<0.05
Ventilation Equipment ⁽⁸³⁾	
Direct Driven Fan	~0.01
Belt Driven Fan	0.4
Controls	0.08
Criticality	8.6×10^{-3}

TABLE 16. 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	1×10^{-5}

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

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 ²³⁸Pu 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_2 and UO_2 not actually in process inside the gloveboxes be kept inside metal containers.
- The dry operation involves PuO_2 and UO_2 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:

1. Incomplete removal of air (oxygen) from a cold furnace at startup before introducing the forming gas containing hydrogen.
2. In-leakage of air (oxygen into the furnace during operation with the furnace at temperature).
3. 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.⁽⁸⁹⁾ 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 17.

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 17. 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) ^(a)	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 18. ^(90 - 93)

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

TABLE 18. 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	16
	Total	432

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.

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:

- a. Obtain more accurate dates of plant startups
- b. Make allowance for reactivity of fuels processed.
- c. Obtain a better basis for quantities of fuel produced to date.
- d. 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.^(94, 95) Filters are judged defective for excessive penetration, filter medium failure, frame failure, gasket failure, and damage during shipment. About 35% to 45% of the defective filters have filter efficiencies less than design specifications.⁽⁹⁵⁾ The efficiency for the filters judged to be defective due to excessive penetration is in the 95% to 99% range.⁽⁹⁴⁾ Failure of the filter medium and frame are the next most important reject reasons.

Failures during installation due to improper seating or careless handling seem to occur at somewhat lower percentages, namely about 1 to 2%.⁽⁹⁶⁾ 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.

Because of these statistical defects, a fuel fabrication facility should have a pre-installation program for examining the HEPA filters to assume 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 Regulation. 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 19.

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 19. Occurrence Frequencies

	No. for 1970-71	Avg/yr	Est. Frequency, ^(a) No./plant/yr
• Personnel Exposure ^(b)	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, they 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 20.

TABLE 20. Occurrence Causes

	No. for 1970-71	Avg/yr	Est. Frequency, No./plant/yr
• 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 21.

TABLE 21. 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; for example, 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 tonne/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.

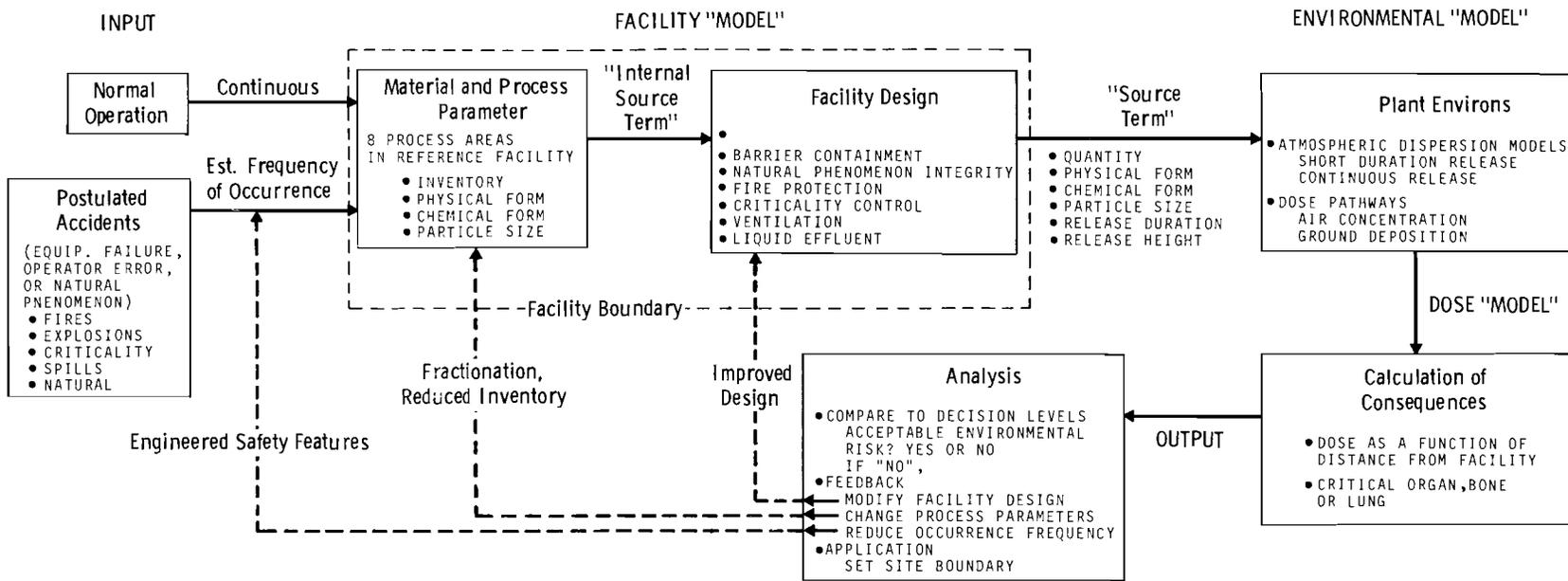


FIGURE 16. Safety Analysis Logic for a Plutonium Fuel Fabrication Facility

A summary of the risk analysis for the reference facility is presented in Table 22. 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 probable dose that an individual will be committed to on an annual basis. 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 22, 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 22. 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 meter, mrem	Probability of Release, yr ⁻¹	Annual "Dose Commitment" Risk, mrem(e)
Normal Operation	5 x 10 ⁻⁶ (S) (I)	Pu Mix (b)	bone lung	4/50 yr (d) 0.06/50 yr	1 1	4 0.06
Criticality	3 x 10 ⁻⁵ (S)	Pu Mix direct radiation Noble gases and Halogens iodine	bone	28/50 yr	8.6 x 10 ⁻³	0.24
			whole body	16	8.6 x 10 ⁻³	0.14
			whole body	31	8.6 x 10 ⁻³	0.27
			thyroid	200	8.6 x 10 ⁻³	1.7
Explosion	10 ⁻² (S)	Pu Mix	bone	9400/50 yr	≈10 ⁻³	9.4
Fire	1.1 x 10 ⁻² (S)	Pu Mix	lung	70/2 yr	<10 ⁻²	<0.70
Glovebox Damage	10 ⁻² (I)	Pu Mix	lung	60/2 yr	<10 ⁻²	<0.60
Facility Fire	10 ⁻² (I)	Pu Mix	lung	60/2 yr	2 x 10 ⁻⁴	0.01
Resin Column Fire	3 x 10 ⁻⁵ (S)	Pu Mix	bone	28/50 yr	<1 x 10 ⁻¹	<2.8
Sintering Furnace Explosion	10 ⁻⁵ (I)	Pu Mix	lung	0.06/yr	<5 x 10 ⁻²	<<0.01
TOTALS:						Bone <16 mrem Lung <1.5 mrem Thyroid 1.7 mrem Whole Body 0.4 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.

X. 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⁽⁹⁷⁾ 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 Figure 17. The definition of "contaminated" was chosen as 10 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 Table 22. Mixture III is the mixture discussed in Section V-A. The Plutonium Surface Contamination Decision Levels shown in Figure 17 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 Table 24.

Published Guidance

Proposed Decision Levels

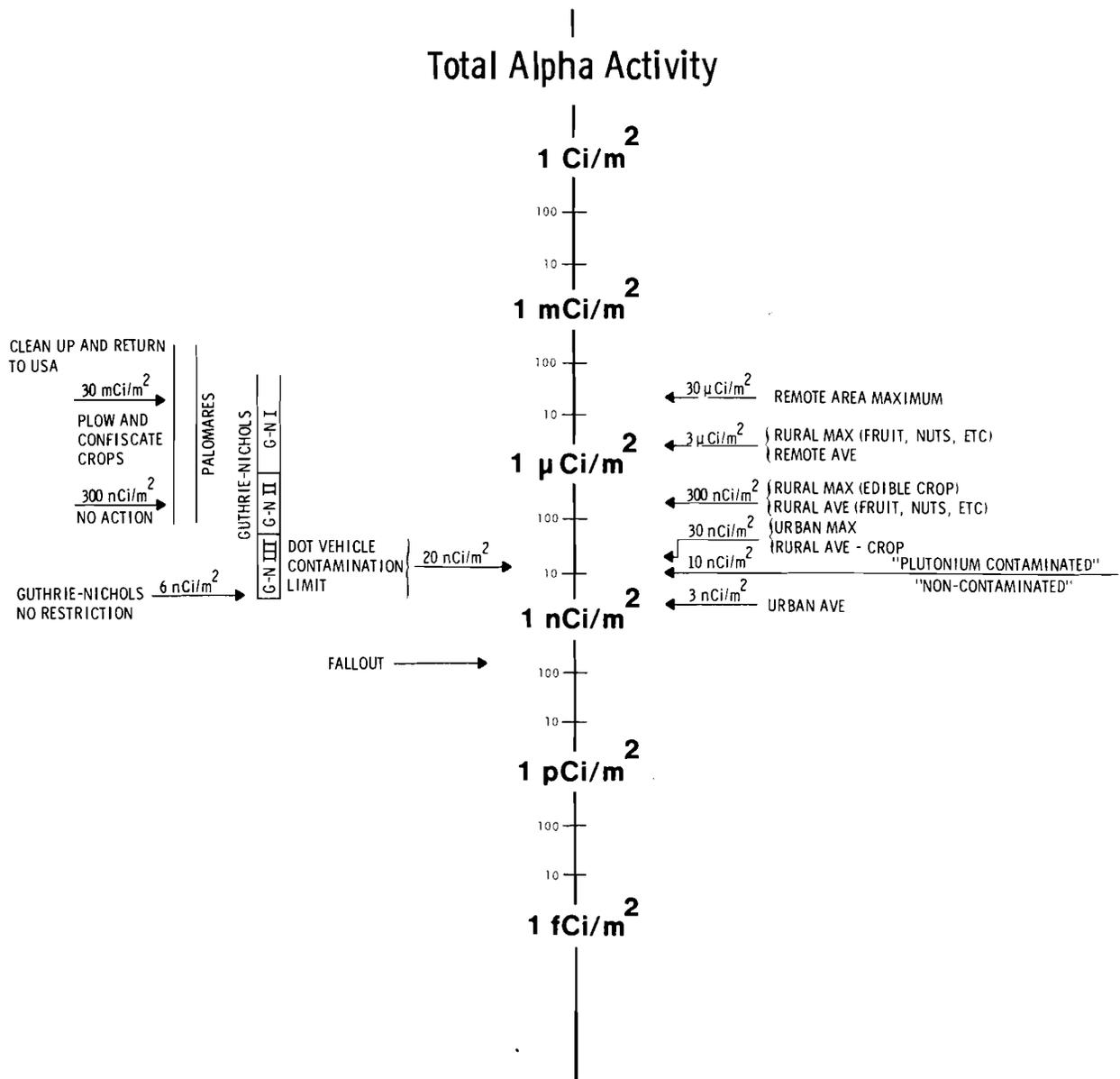


FIGURE 17. Plutonium Surface Contamination Levels

TABLE 23. Comparison of Isotopic Mixtures

<u>Isotope</u>	<u>wt%</u>	<u>Ci/g Isotope</u>	<u>Ci/g Mixture</u>	<u>Activity %</u>	<u>MPC_A Mixture μCi/cm³</u>
Pu-239	95	0.061	<u>MIXTURE I</u> 0.06 0.01 <u>0.07</u>	86 14	(S 6x10 ⁻¹³ I 1x10 ⁻¹¹)
Pu-240	5	0.227			
Pu-239	2.85	0.061	<u>MIXTURE II</u> 0.002 0.0003 3.21x10 ⁻⁷ 1.49x10 ⁻⁸ <u>0.0023</u>	87 13	(S 6x10 ⁻¹³ I 1x10 ⁻¹¹)
Pu-240	0.15	0.227			
U-238	96.3	3.33x10 ⁻⁷			
U-235	0.7	2.14x10 ⁻⁶			
Pu-238	1.9	16.8	<u>MIXTURE III</u> 0.319 0.038 0.043 13.7 0.0002 <u>14.1</u>	2.3 0.3 0.3 97.1 ---	(S 3x10 ⁻¹¹ I 1x10 ⁻⁸)
Pu-239	63	0.061			
Pu-240	19	0.227			
Pu-241	12	114.0			
Pu-242	3.8	0.004			
Pu-238	0.06	16.8	<u>MIXTURE IV</u> 0.010 0.0012 0.0013 0.4104 4.4x10 ⁻⁶ 3.2x10 ⁻⁷ 1.5x10 ⁻⁸ <u>0.423</u>	2.4 0.28 0.31 97 --- --- ---	(S 3x10 ⁻¹¹ I 1x10 ⁻⁸)
Pu-239	1.9	0.061			
Pu-240	0.57	0.227			
Pu-241	0.36	114			
Pu-242	0.11	0.004			
U-238	96.3	3.33x10 ⁻⁷			
U-235	0.7	2.14x10 ⁻⁶			

S - Soluble
I - Insoluble

TABLE 24. Annual Average Whole Body Dose
from Natural Background Radiation
(mrem)

	<u>Beir</u> ⁽⁹⁸⁾	<u>Unsear</u> ⁽⁹⁹⁾	<u>Table II-4</u> <u>ORP/CSD 72-1</u> ⁽¹⁰⁰⁾
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 25.

TABLE 25. 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 1952 mrem/yr to the bone, the general population exposure limits for man-made radiation other than medical would be as shown in Table 26.

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 27.

TABLE 27. 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 28.

TABLE 28. Derived MPC_a's

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

TABLE 26. 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⁽¹⁰¹⁾ 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."

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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 (NO₃)₄, soluble
PuO₂, insoluble
PuO₂-UO₂, insoluble
Small quantities of miscellaneous soluble Pu compounds

Physical Forms: PuO₂ powders, dispersible, particle size 0.1-80 μm
PuO₂-UO₂ powders, dispersible, particle size 0.1-1400 μm
PuO₂-UO₂ pellets, essentially nondispersible
Solutions (primarily nitrate) containing soluble Pu, dispersible
Slurries containing insoluble Pu, dispersible
PuO₂-UO₂ 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.

A.1

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 μm . The larger particles are loosely bound agglomerates. Approximately 95% of the particles are larger than 1 μm and the average particle size is 6-10 μm .

Inventory: 50 kg Pu

Chemical Forms: Pu oxalate, soluble
 PuO_2 , insoluble

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 PuO₂-UO₂ (60 kg Pu) in pellets
1.6 kg Pu in solution or slurry

Chemical Forms: PuO₂-UO₂, insoluble

Physical Forms: PuO₂-UO₂ green (not easily dispersed) and sintered (essentially nondispersible) pellets

PuO₂-UO₂ powder in solution or slurry, dispersible

Comments: The centerless grinding operation yields PuO₂-UO₂ dust or slurry depending on whether the operation is performed wet (the usual case) or dry. The washing operation yields a PuO₂-UO₂ solution or slurry.

PROCESS AREA V - PELLET LOADING AND FUEL ROD ENCAPSULATION

Typical Operations: The out-gassed PuO₂-UO₂ pellets are loaded into fuel rods. The rods are then welded closed and decontaminated.

Inventory: 850 kg PuO₂-UO₂ (30 kg Pu)

Chemical Forms: PuO₂-UO₂, insoluble

Physical Forms: Sintered PuO₂-UO₂ pellets, essentially nondispersible

Fuel rods containing sintered PuO₂-UO₂ pellets, nondispersible

Physical Forms: Pu oxalate slurry and wet powder, dispersible
dry PuO₂ 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 PuO₂ powder is crushed and screened to obtain particles of a few micrometers and then blended with UO₂ powder. The blended PuO₂-UO₂ 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% PuO₂-96 wt% UO₂) with UO₂

Chemical Forms: PuO₂, insoluble
UO₂, insoluble

Physical Forms: PuO₂ powder, dispersible
UO₂ powder, dispersible
PuO₂-UO₂ powder, dispersible
Green PuO₂-UO₂ 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 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 PuO₂-UO₂ (60 kg Pu)

Chemical Forms: PuO₂-UO₂, 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 UO₂
 ≤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 PuO₂-UO₂ and PuO₂ 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 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 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

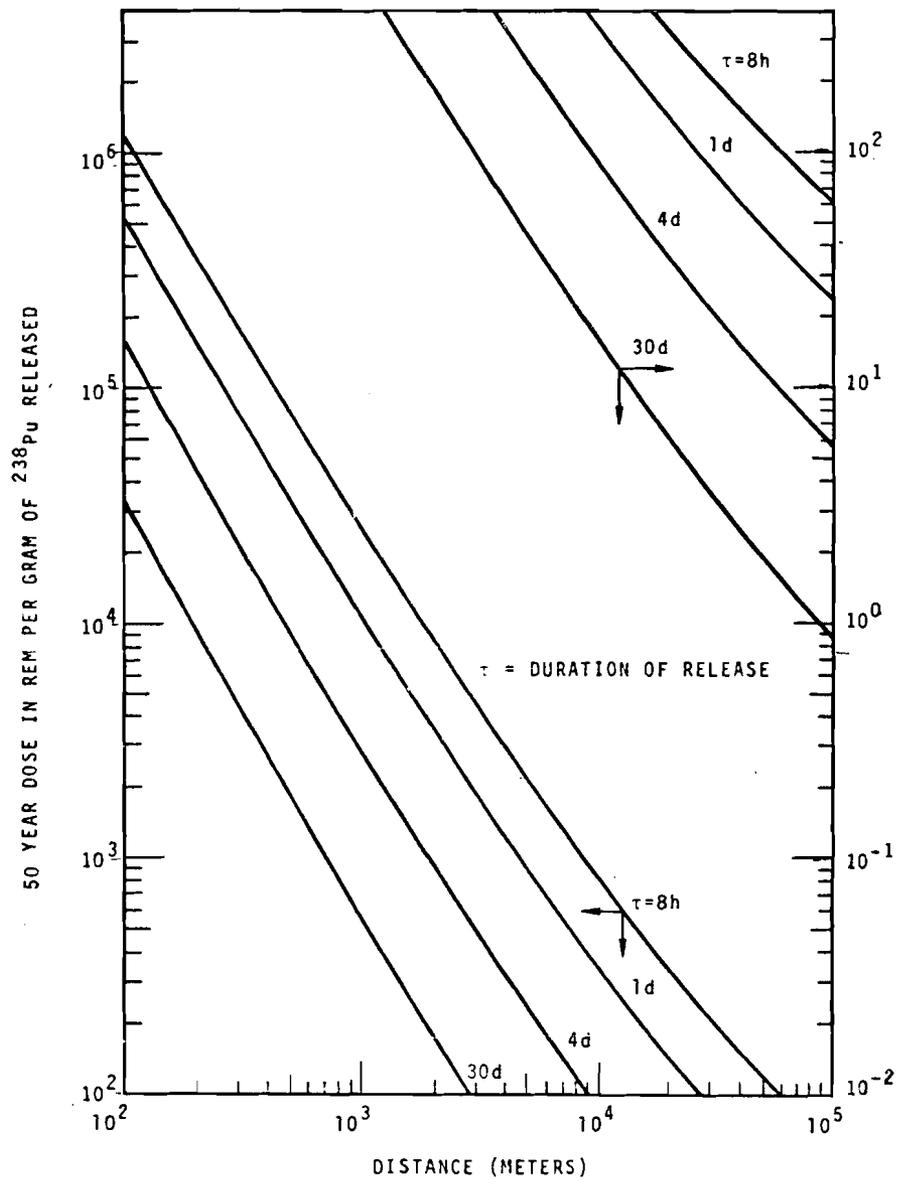


FIGURE B.1. Dose to Bone from Acute Inhalation of Plutonium-238, Ground Level Release

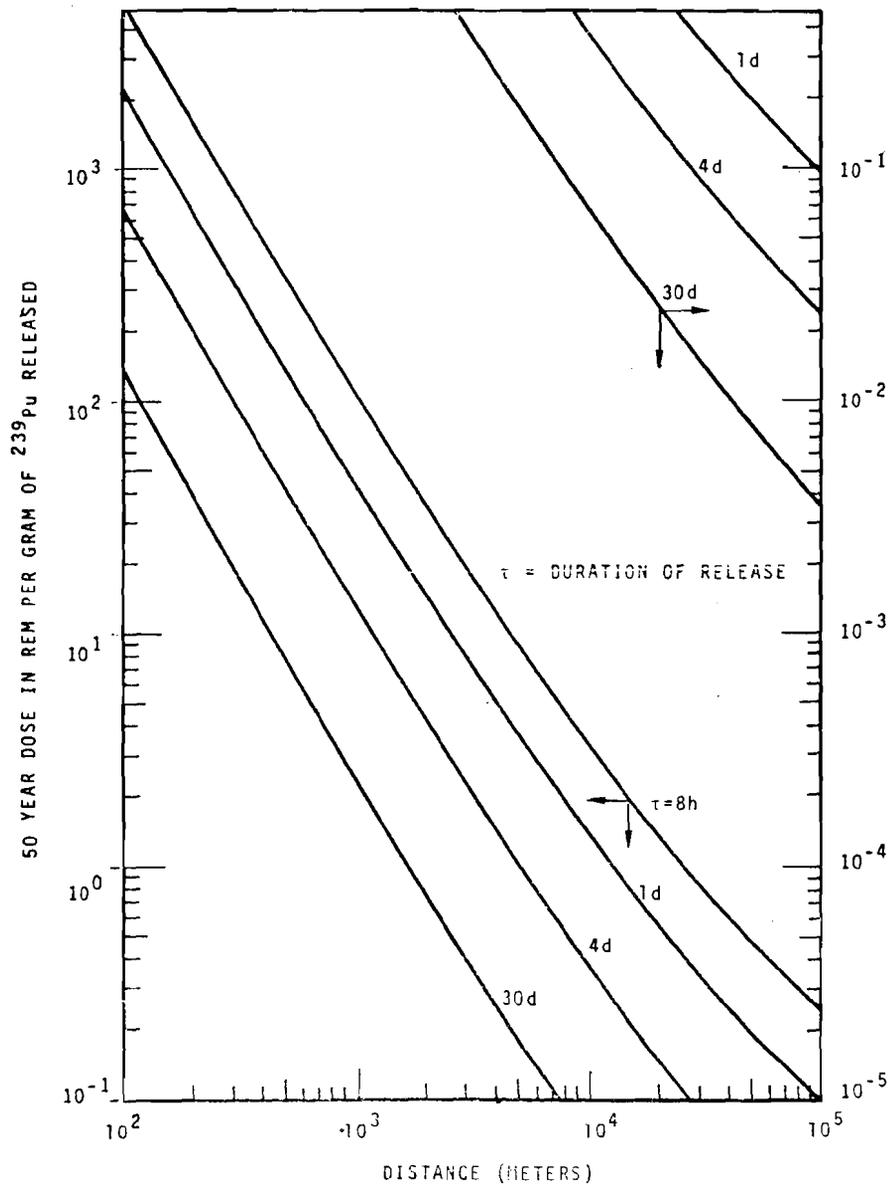


FIGURE B.2. Dose to Bone from Acute Inhalation of Plutonium-239, Ground Level Release

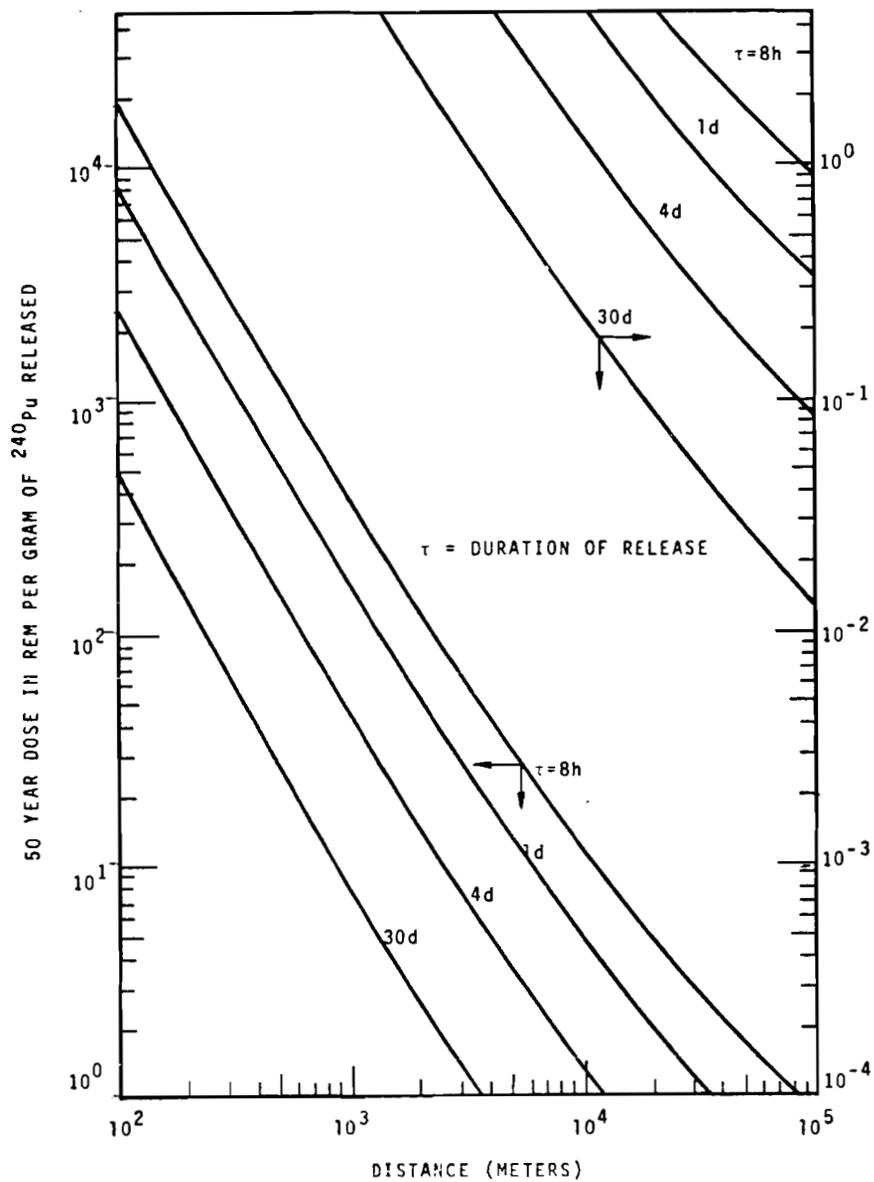


FIGURE B.3. Dose to Bone from Acute Inhalation of Plutonium-240, Ground Level Release

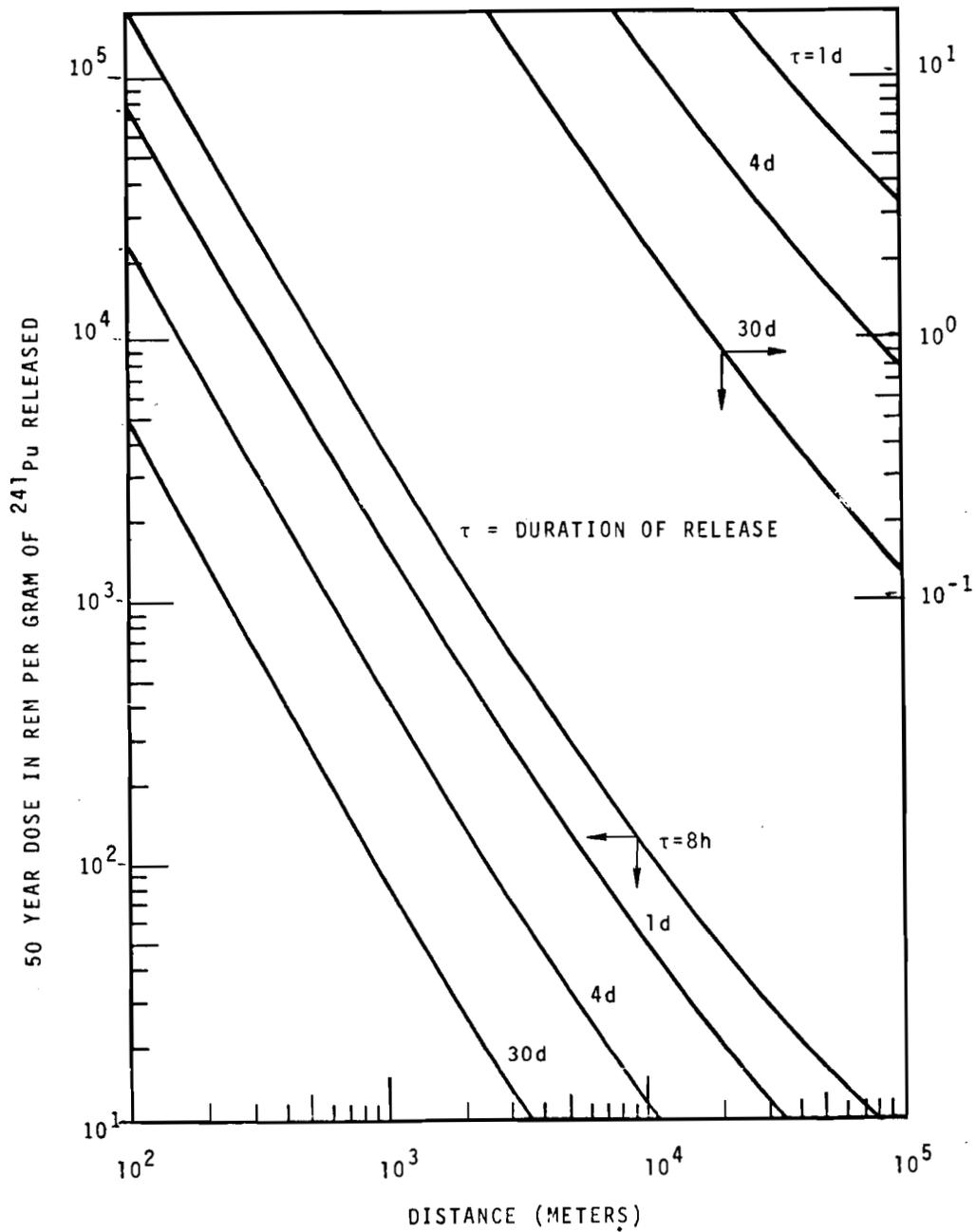


FIGURE B.4. Dose to Bone from Acute Inhalation of Plutonium-241, Ground Level Release

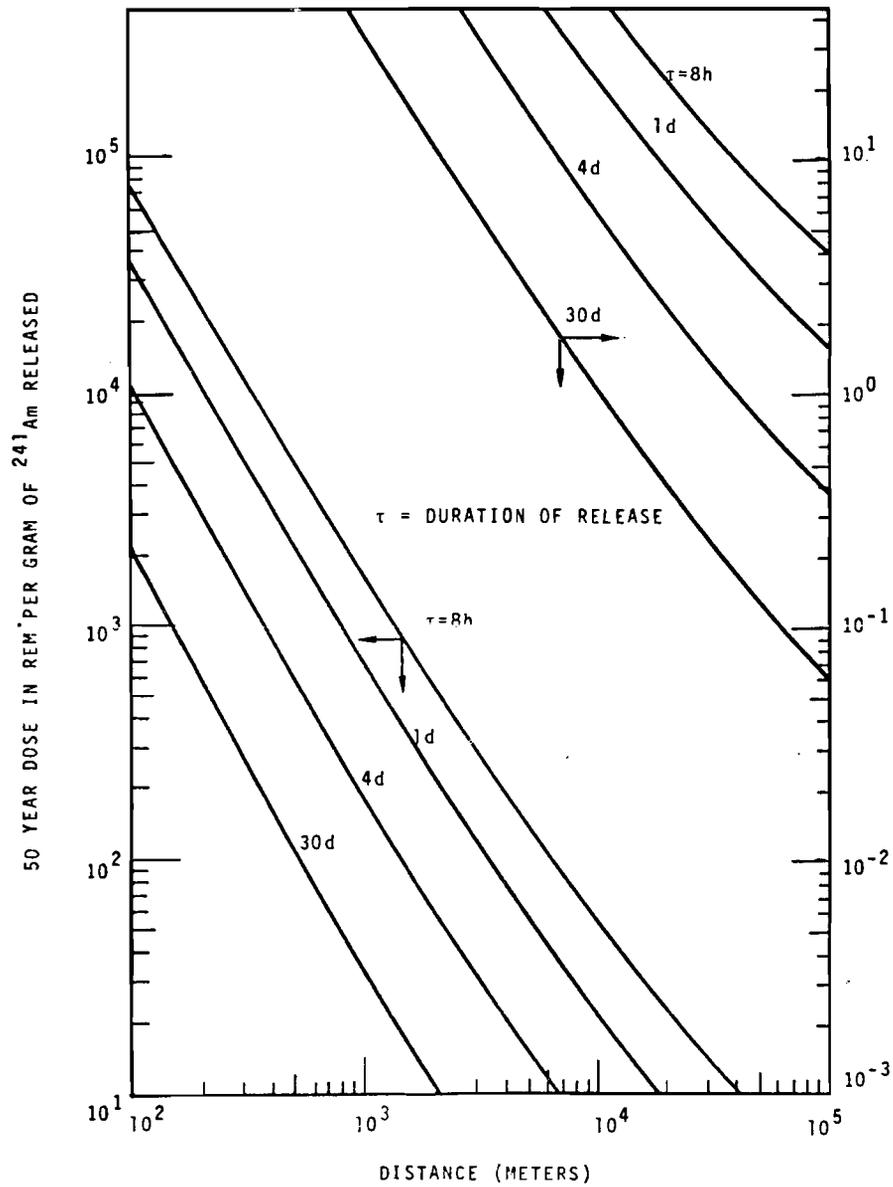


FIGURE B.5. Dose to Bone from Acute Inhalation of Americium-241, Ground Level Release

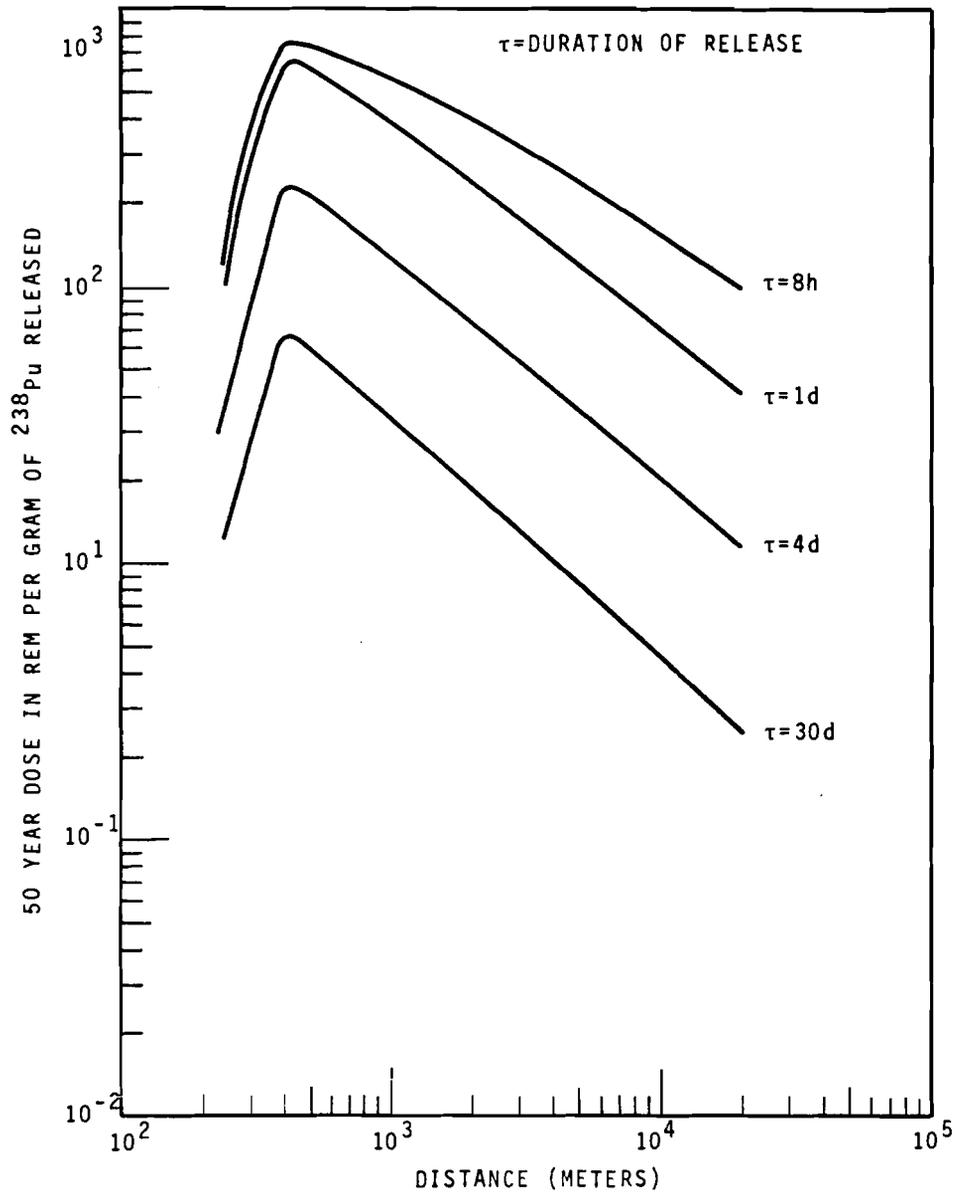


FIGURE B.6. Dose to Bone from Acute Inhalation of Plutonium-238, Release Height 100 Meters

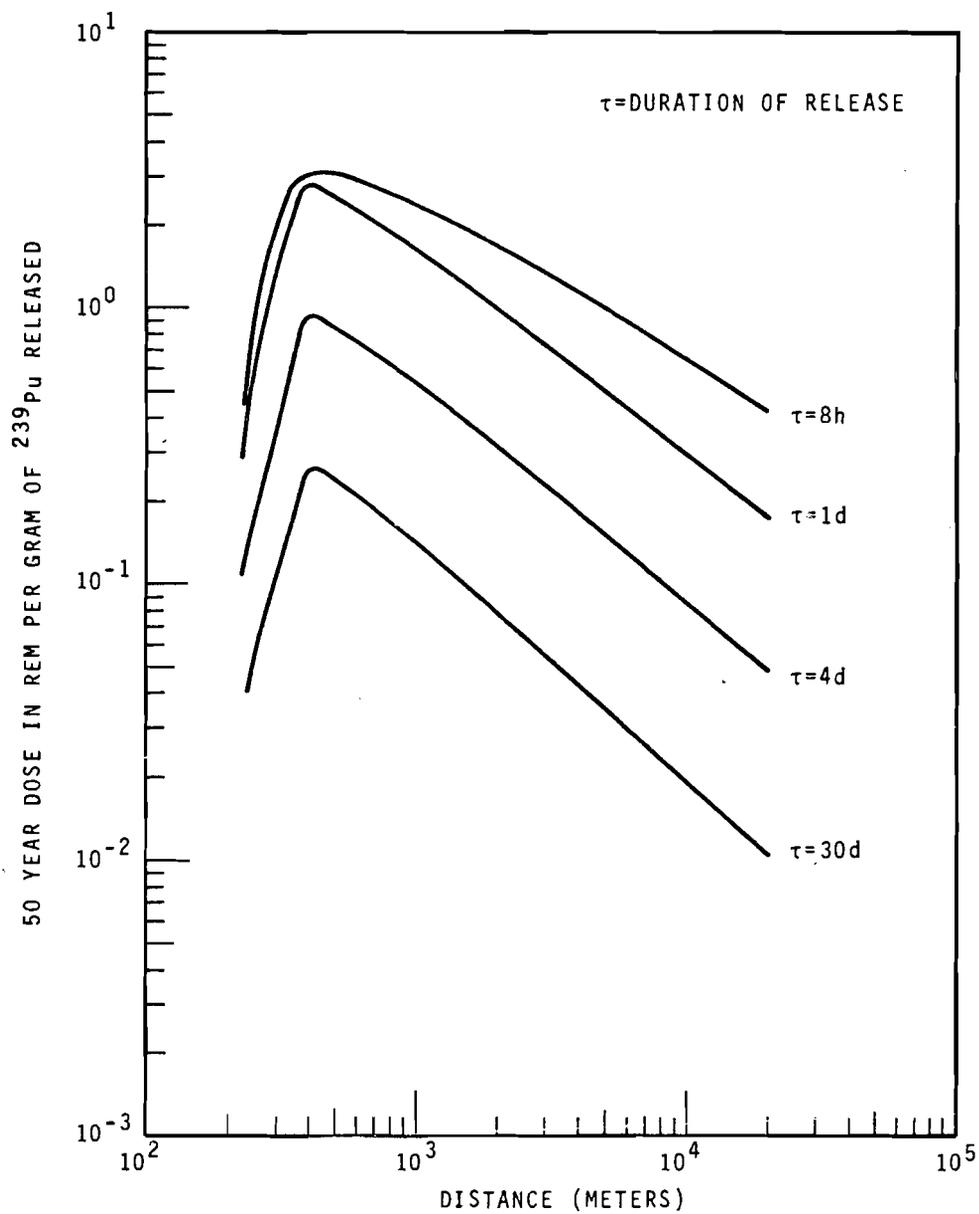


FIGURE B.7. Dose to Bone from Acute Inhalation of Plutonium-239, Release Height 100 Meters

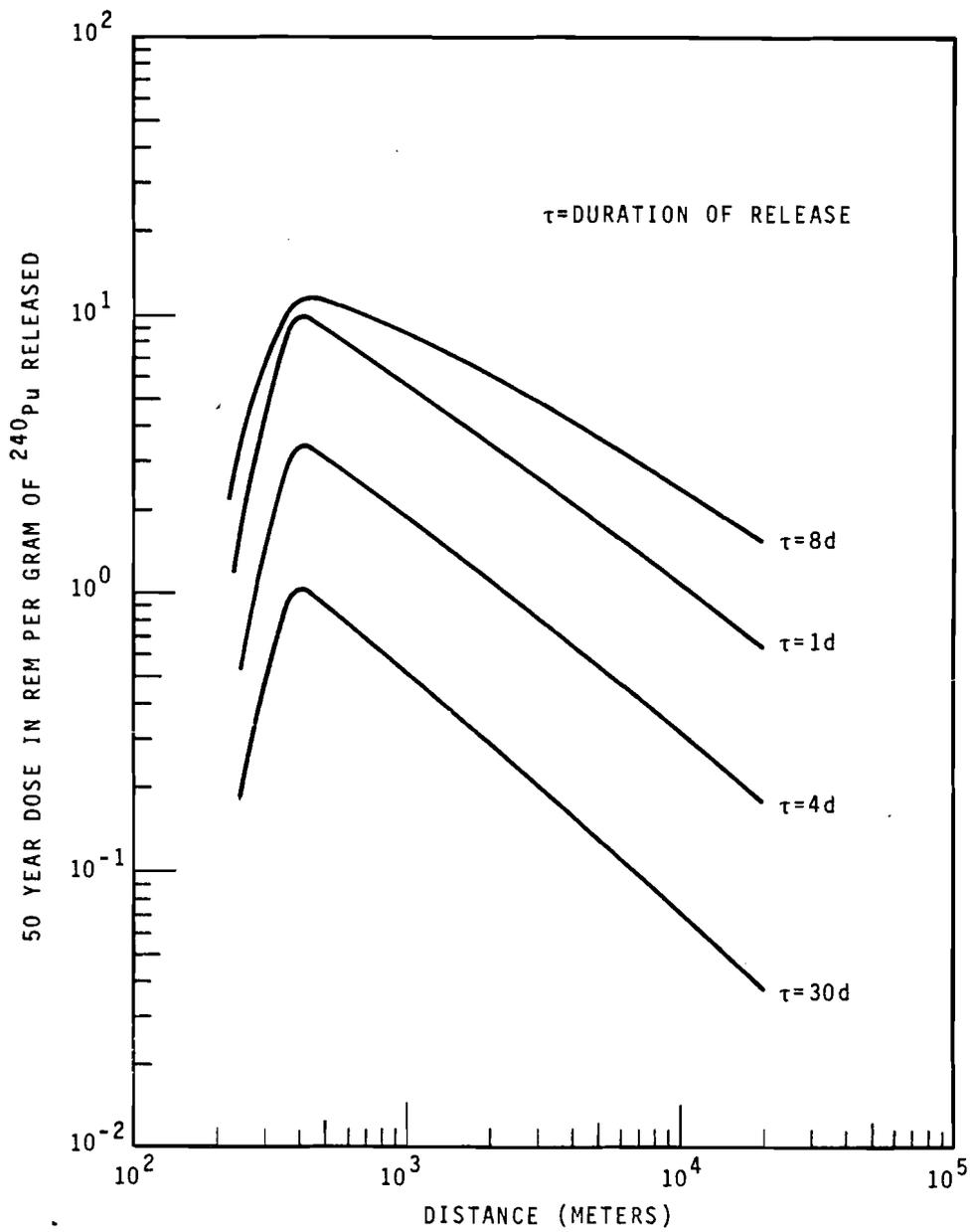


FIGURE B.8. Dose to Bone from Acute Inhalation of Plutonium-240, Release Height 100 Meters

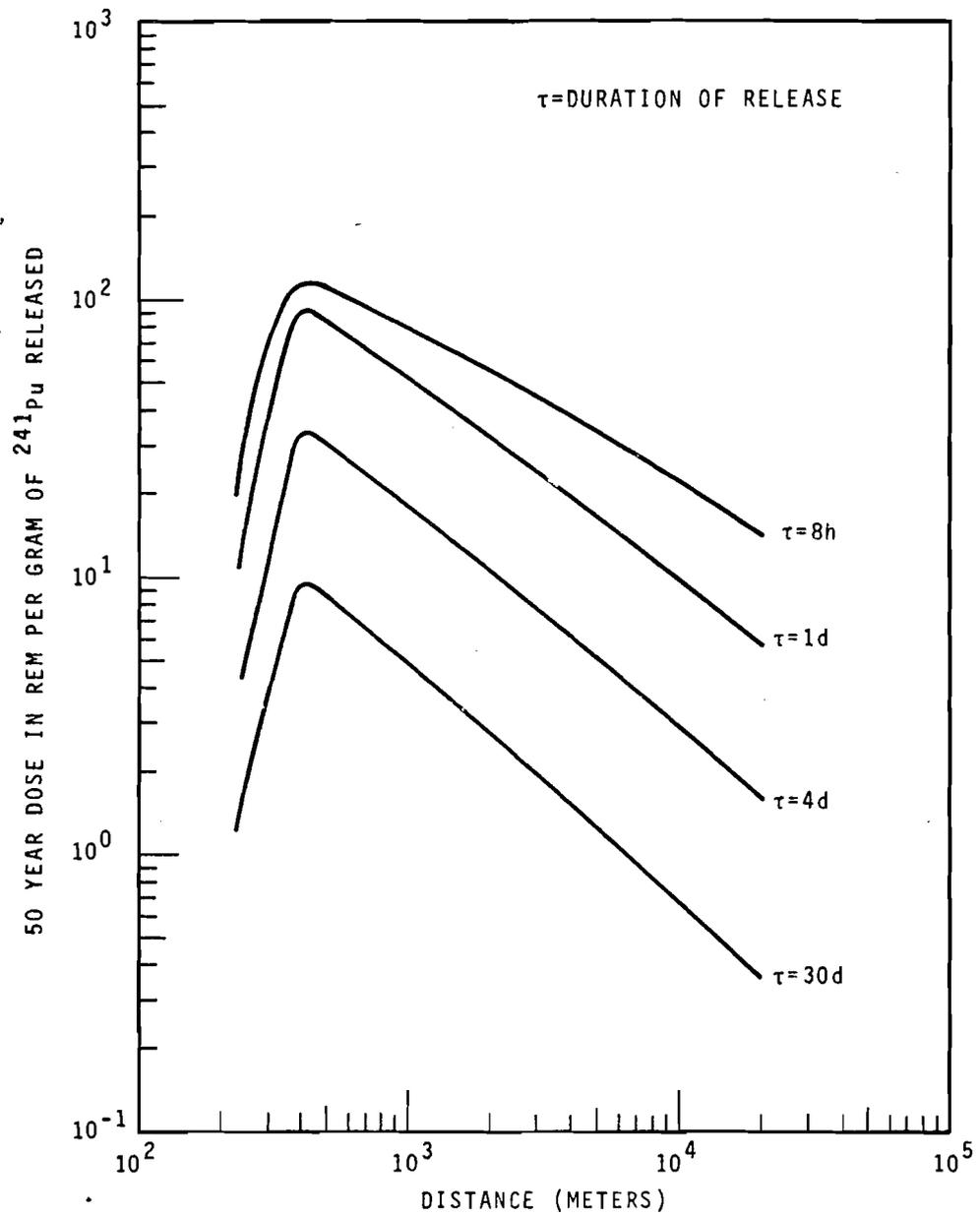


FIGURE B.9. Dose to Bone from Acute Inhalation of Plutonium-241, Release Height 100 Meters

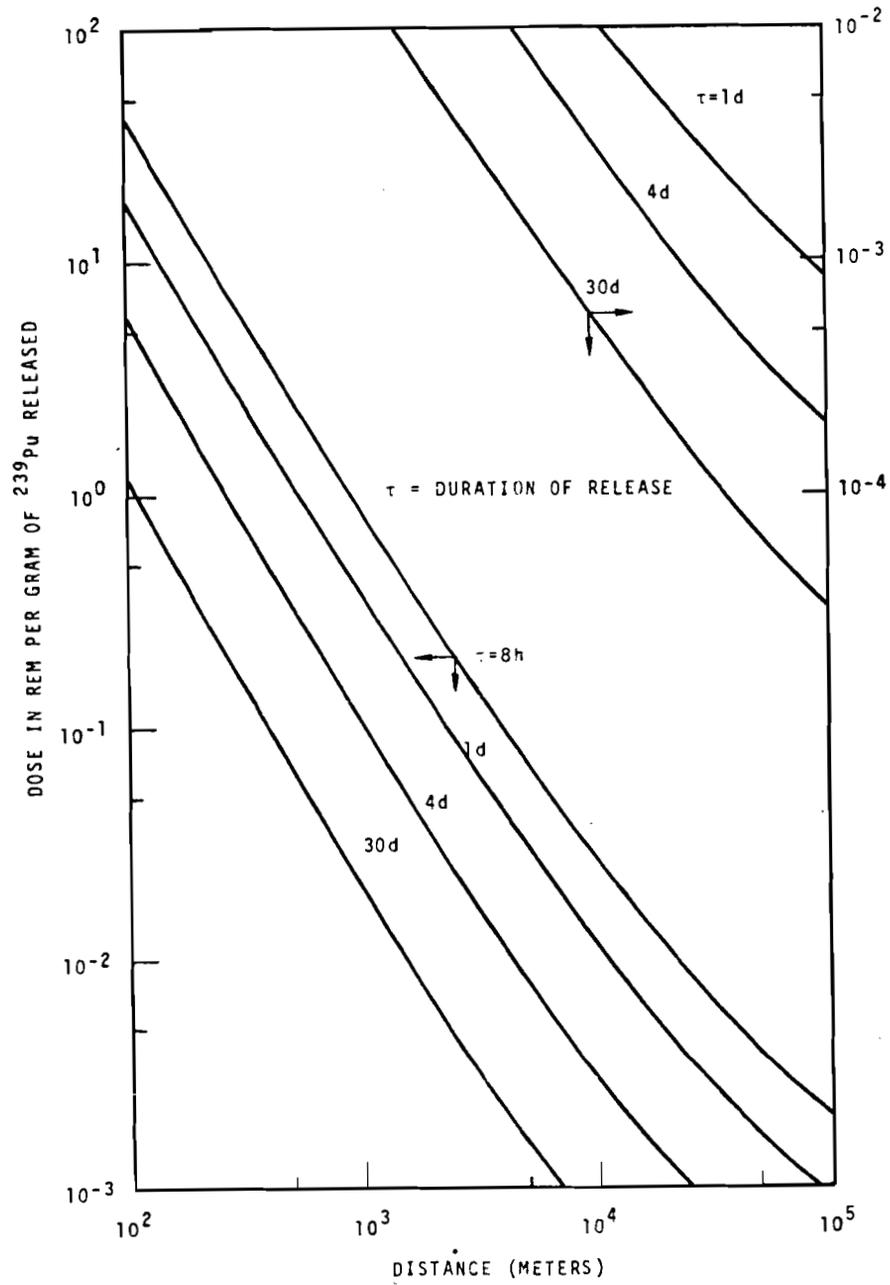


FIGURE B.12. Dose to Lung from Acute Inhalation of Plutonium-239, Ground Level Release

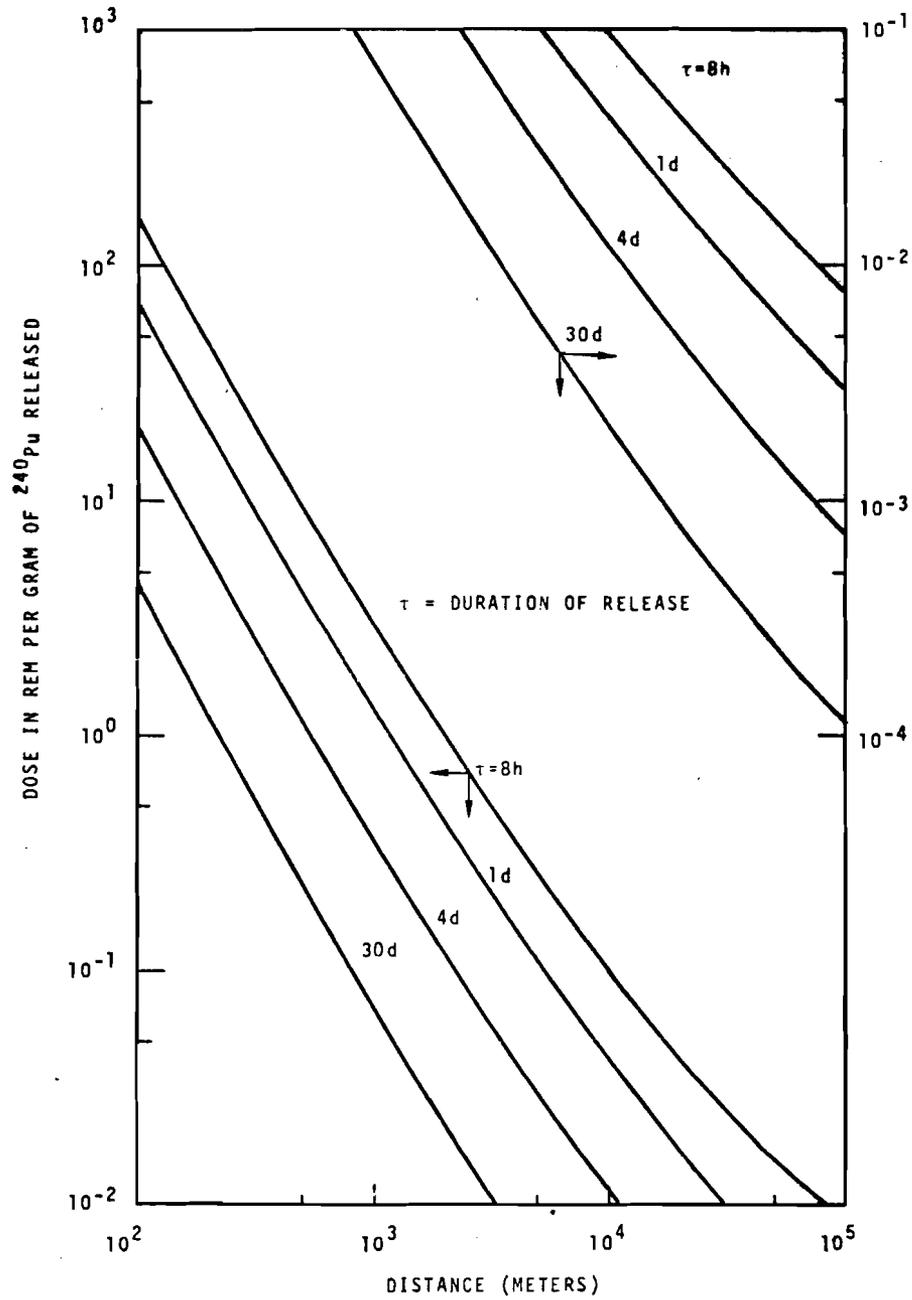


FIGURE B.13. Dose to Lung from Acute Inhalation of Plutonium-240, Ground Level Release

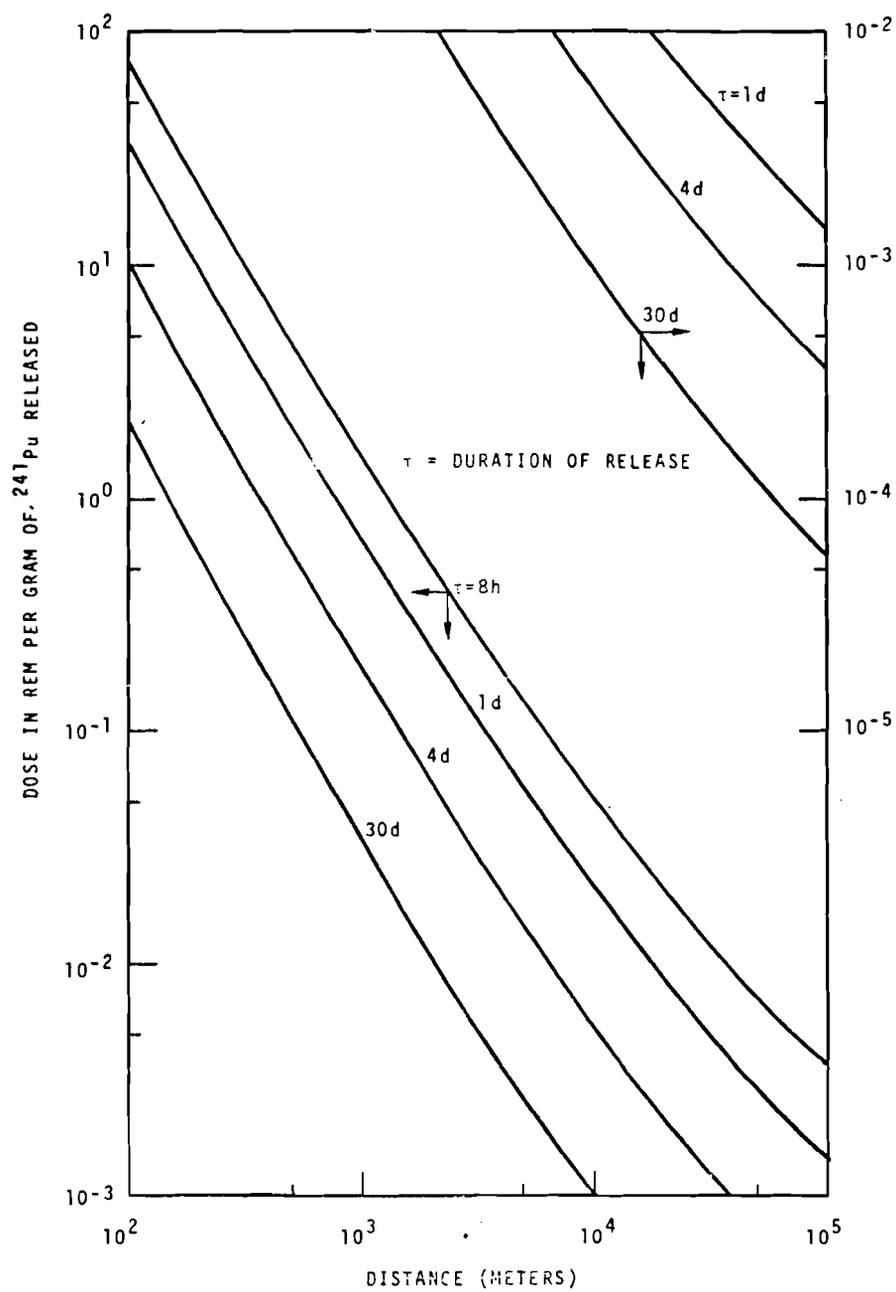


FIGURE B.14. Dose to Lung from Acute Inhalation of Plutonium-241, Ground Level Release

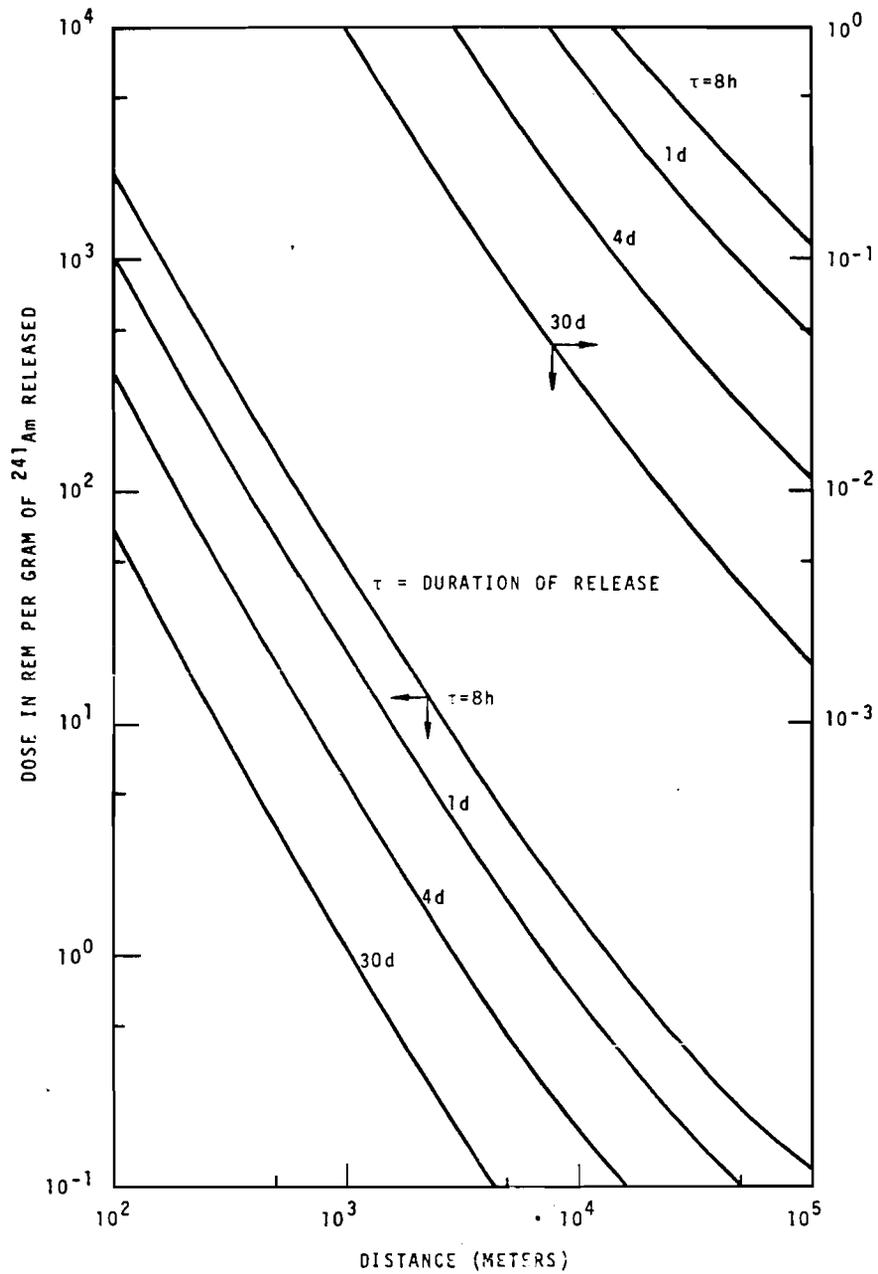


FIGURE B.15. Dose to Lung from Acute Inhalation of Americium-241, Ground Level Release

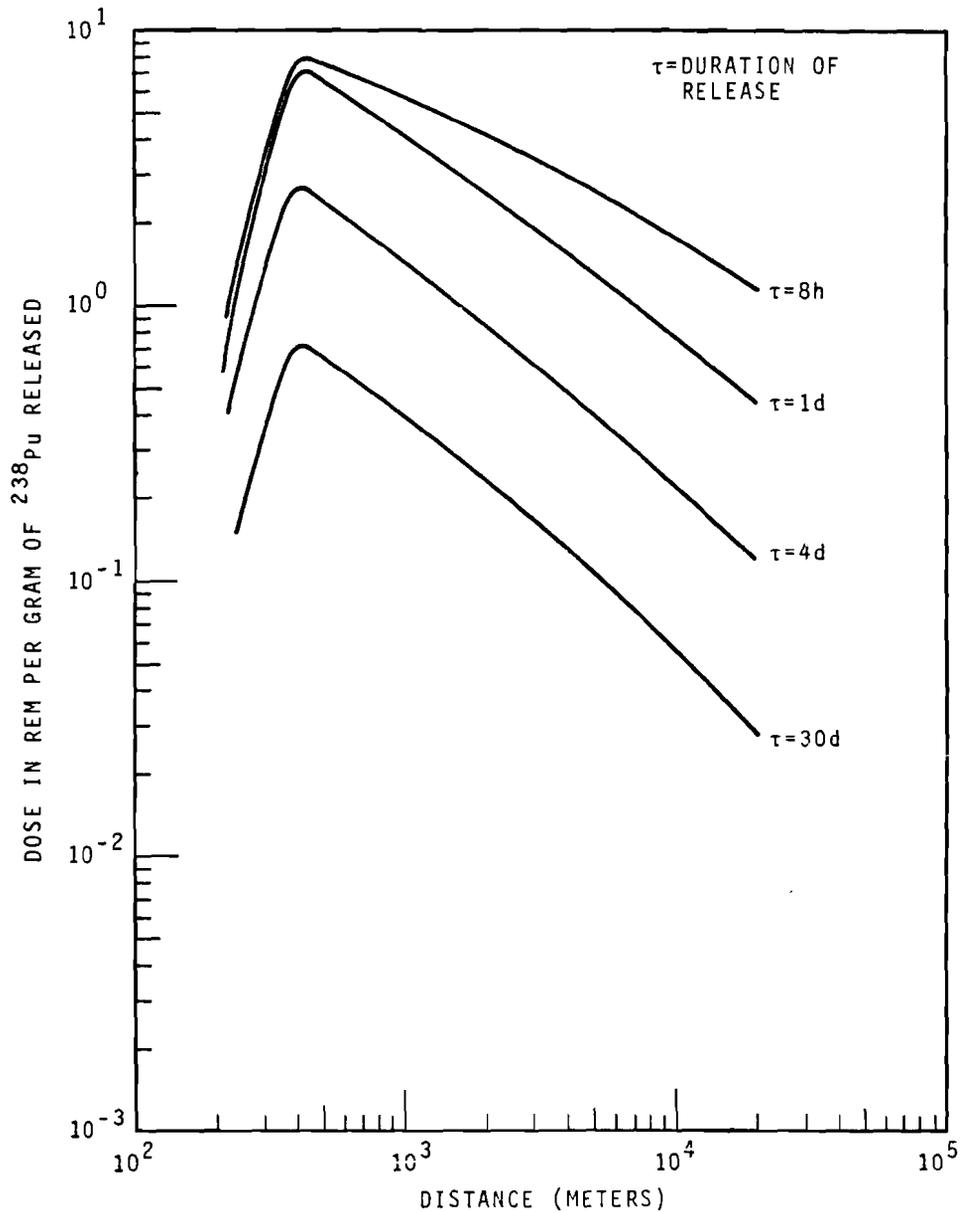


FIGURE B.16. Dose to Lung from Acute Inhalation of Plutonium-238, Release Height 100 Meters

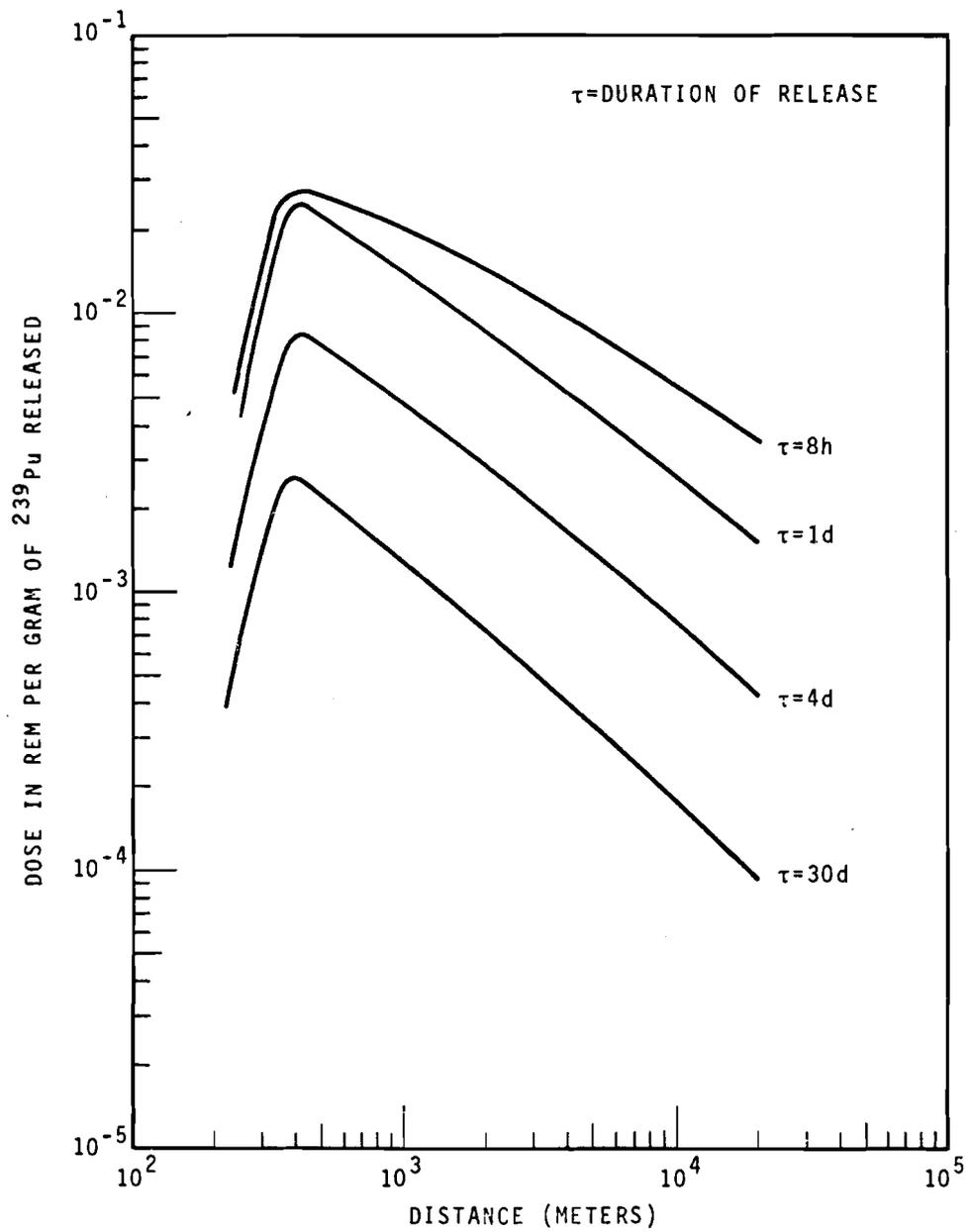


FIGURE B.17. Dose to Lung from Acute Inhalation of Plutonium-239, Release Height 100 Meters

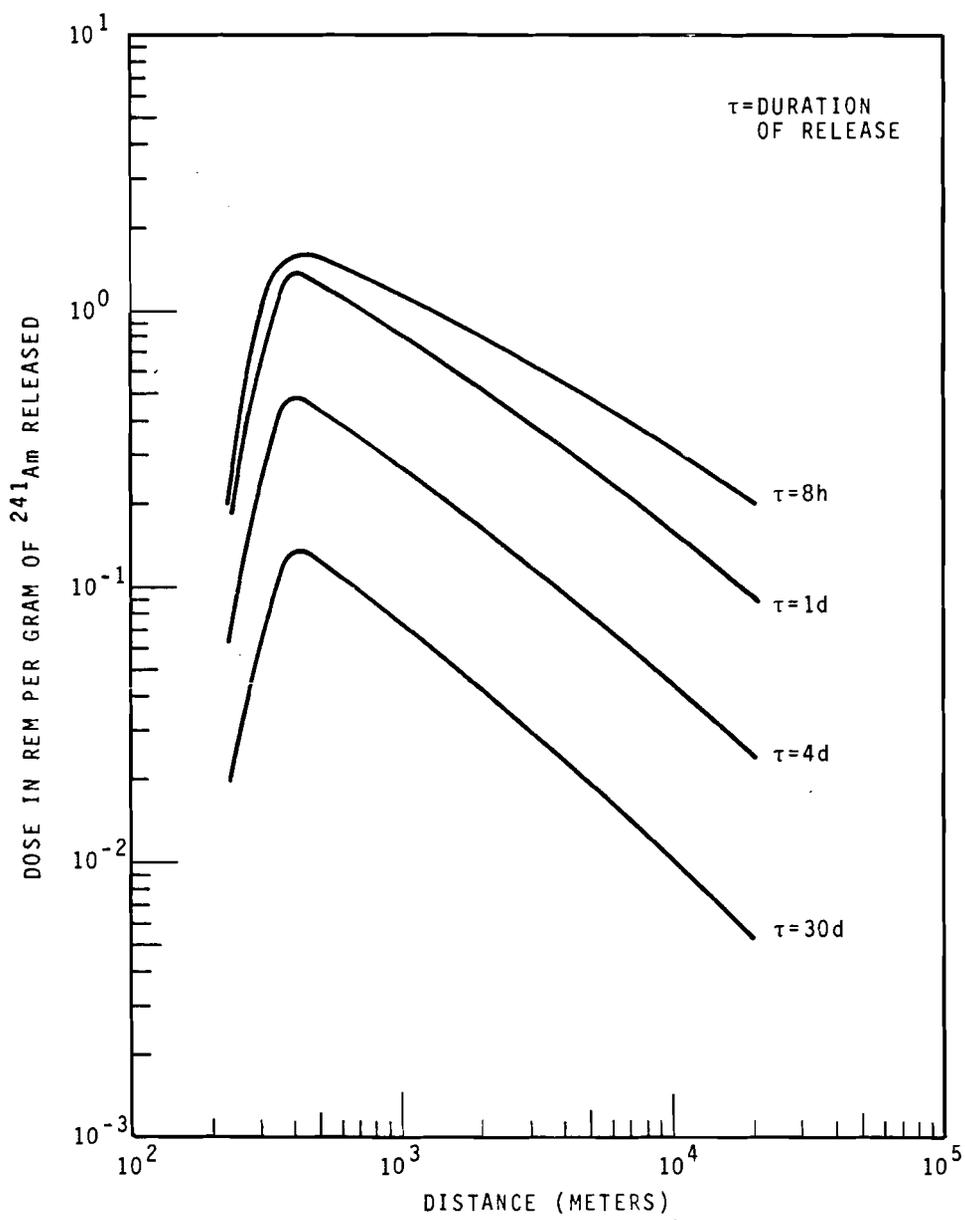


FIGURE B.20. Dose to Lung from Acute Inhalation of Americium-241, Release Height 100 Meters

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

APPENDIX C

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

- 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. ⁽¹⁾

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. ⁽⁶⁾ 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 $6.17 \times 10^{-2} \text{ Ci/g (Pu}^{239}\text{)}$, quantities of particles less than $10 \mu\text{m (AED)}$ generated ranged from 10^{-6} to $10^{-5} \text{ g/cm}^2/\text{sec}$ in dry air and 6×10^{-12} to $8 \times 10^{-6} \text{ g/cm}^2/\text{sec}$ 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. ⁽⁷⁾

Mishima found very little plutonium airborne during the oxidation in dry air of ignited, small specimens of unalloyed metal. From 3×10^{-6} to 5×10^{-5} percent 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 $15 \mu\text{m Aerodynamic Equivalent Diameter (AED)}$. ⁽⁸⁾ 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 $4.2 \mu\text{m}$. ⁽⁹⁾ 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.⁽¹⁰⁾ 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

1. 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.⁽¹⁾ 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.⁽¹¹⁾ 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.)⁽¹²⁾ Under static conditions (materials made airborne by naturally occurring phenomena), only 0.05% was found airborne.⁽¹³⁾ 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-hour 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 hours) 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-hour 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-hour period. During the course of a gasoline fire (20 to 45 minutes) 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-hour 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-hour 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 hours at 2.5 mph (85% less than $10\mu\text{m}$ AED and 5.4% in 24 hours 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 minutes) 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 hours at 2.5 mph (85% less than $10\mu\text{m}$ AED) and 0.7% from the residue in 24 hours 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 hour 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 hours.⁽¹²⁾ At the higher velocity entrainment appears to be essentially complete in 2 hours.⁽¹²⁾ 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".

2. 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.⁽¹⁾ The particle size of the oxide produced by calcination depends upon the conditions which the precursor and oxide are prepared.⁽¹⁴⁾ Although the size of the basic oxide particles produced appear to increase with temperature,^(15,13) the size of the aggregates showed no correlation to calcination temperature and indicated a Mass Median Diameter of 8 to $12\mu\text{m}$.⁽¹³⁾ Oxide particles were 26 to 68% of the size of their precursor.⁽¹³⁾ 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 updraft of air.⁽¹¹⁾ 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-hour 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.

3. Plutonium Fluoride Powder - Plutonium fluoride is stable in moist air up to about 300 °C with complete conversion to the dioxide around 600 °C. ⁽¹⁾

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 upsweep of air up to 100 cm/sec. ⁽¹¹⁾ The source material was a green, finely divided free-flowing powder composed of spheres and agglomerates with a MMD of 38 μ 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.

4. 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 hours. Less than $10^{-6}\%$ was airborne from the solid residues during a 24-hour 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 airsweep 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 hour 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.⁽¹³⁾ 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.⁽¹²⁾ 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 ranges from 3.4×10^{-4} to 0.1% at a nominal velocity of 50 cm/sec and 0.05 to 8% at 100 cm/sec.⁽¹²⁾

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.⁽¹²⁾ 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 μm AED) from sandy soil to 0.06% from an asphalt/gravel mixture of which 50% was associated with particles less than 10 μm . 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 μm AED) and 2.6% from a stainless steel surface (30% less than 10 μm AED).

Involving the nitrate solution in a fire increased the fractional airborne release. As much as 0.04% (18% less than 10 μm) from sandy soil, 0.1% from vegetation covered soil (80% less than 10 μm AED), 0.24% (68% less than 10 μm AED) from an asphalt/gravel mixture and 1.1% (40% less than 10 μm 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 μm AED) and from stainless steel to 11.4% (35% less than 10 μm 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%.

- Aerosol Behavior

1. 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⁽¹⁸⁾ (1170 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/cm³ in a few minutes. Particle populations of less than 10^5 particles/cm³ are found even in dusty operations.⁽¹⁹⁾

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/cm³ will persist, the mass of plutonium airborne as oxide would be 2 g/m^3 . Generating a monodispersal aerosol is a difficult task. Some authors have offered experimental confirmation of the tendency of environmental aerosols for a "self-preserving size distribution".^(21,22) 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⁽²³⁾ 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 μm plutonium dioxide with a density of 2 (due to agglomeration) with a population of 10^6 particles/ cm^3 , they estimated the mass concentration would be 33 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 feet ($90,613 \text{ m}^3$) the airborne concentration is in the order of 45 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.,⁽²⁴⁾ vaporized PuO_2 and UO_2 in a 0.8 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 minutes 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 $0.375 \text{ g}/\text{m}^3$ but measured mass concentrations range from 0.0105 to $0.071 \text{ g}/\text{m}^3$ (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 μ m AED are not respirable and do not constitute an inhalation hazard. Thus a maximum mass concentration in the neighborhood of 100 mg Pu/m³ seems reasonable.

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

Fish, et al.,⁽²⁵⁾ 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.⁽²⁶⁾ 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 μCi of airborne material (0.003 g) was

released through a damaged exhaust system.⁽²⁷⁾ 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.⁽²⁸⁾ 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 non-nuclear detonations of weapons, a value of $1000 \mu\text{g Pu/m}^2$ was established as safe for life-time occupancy (weather being the sole resuspension force) although a value of $3500 \mu\text{g/m}^2$ was determined as safe.⁽²⁹⁾ Resuspension factors in the range of 7×10^{-7} to $7 \times 10^{-5} \text{ m}^{-1}$ were found.⁽³⁰⁾ Resuspension factors in the order of 10^{-13} m^{-1} were reported for the thoroughly weathered material.⁽³¹⁾ 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.⁽³²⁾

The resuspension of plutonium particles has also been measured. Jones and Pond⁽³³⁾ measured the resuspension from paper covered, plastic covered and waxed linoleum surfaces. Particles ranged from 0.4 to 60 μm . 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⁽³⁴⁾ measured resuspension of aged material from concrete surface in an old production facility. Factors for plutonium oxide ranged from 10^{-6} to 10^{-7} m^{-1} for no movement before the test, 10^{-3} m^{-1} for no movement after the test, 10^{-3} m^{-1} with air circulation over the surface, to 10^{-2} 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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