**PLUTONIUM AIR CONCENTRATIONS
AND PARTICLE SIZE RELATIONSHIPS
IN HANFORD FACILITIES**

B. V. ANDERSEN
I. C. NELSON

DECEMBER 1967

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PLUTONIUM AIR CONCENTRATIONS
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ABSTRACT

Measurement results are presented from a study of a variety of potential employee exposure situations in three major plutonium handling facilities. Plutonium-239 aerosol concentration measurements ranged from near-worldwide fallout values of about one-half fCi/m³ to several thousand fCi/m³. The particle size distributions were found to be bimodal in nature in the majority of the samples examined. The typical aerosol showed an Activity Median Diameter (AMD) of about 0.2 μ with a geometric standard deviation of 1.4 for 60% of the plutonium-239, with 40% exhibiting an AMD of 2.4 μ with a geometric standard deviation of 1.3.

The measured plutonium-239 aerosol parameters were then utilized in the study of relationships of chronic inhalation with current respiratory tract models for deposition and retention of plutonium aerosols. Pulmonary burdens of plutonium-239 for chronic exposure were calculated to be in the picocurie to nanocurie range.

INTRODUCTION

This report presents the results obtained from a comprehensive study of the breathing air as found in three major Hanford facilities utilized almost exclusively in plutonium work programs. The facilities studied were the Isochem (now Atlantic Richfield) Plutonium Purification and Fabrication Facility (PP&FF), the Battelle-Northwest Plutonium Metallurgy Facility (PMF), and the Battelle-Northwest Plutonium Development Laboratories (PDL).

The primary purpose of the study was to provide data over a reasonably representative period of time that would permit evaluation of the probable ranges of internal deposition of plutonium in employees. The employees of interest were those who have worked in plutonium handling facilities for long periods of time, but who have not shown

a measurable internal deposition by the usual excreta analysis techniques.^(1,2) The collected data provides a possible basis for prediction of employee internal plutonium deposition ranges when utilized with deposition and retention models formulated by the International Task Group on lung dynamics.⁽³⁾

SUMMARY AND CONCLUSIONS

Measurement results are presented from a study of a variety of potential employee exposure situations in three major plutonium handling facilities. Plutonium-239 aerosol concentration measurements ranged from about one-half fCi/m³ to several thousand fCi/m³. The typical air concentration measured for radiation zone worker exposure was about 20 fCi/m³, while office areas showed typical exposure concentrations of about 3 to 4 fCi/m³.

Most of the particle size distributions appeared to be bimodal in nature when plotted as histograms. Treatment on a bimodal basis permitted inclusion of large particles, which were found to be present in nearly all of the aerosol samples examined. The typical aerosol for radiation zone locations, showed an Activity Median Diameter (AMD) of about 0.2μ with a geometric standard deviation of 1.4 for 60% of the plutonium-239, with 40% exhibiting an AMD of 2.4μ with a geometric standard deviation of 1.3. The typical aerosol for office areas showed an AMD of 0.15μ with a geometric standard deviation of 1.5 for 80% of the plutonium-239, with 20% having an AMD of 1.5μ with a geometric standard deviation of 1.2.

The concentration and particle size distribution data were then utilized in a study of relationships of chronic inhalation with current respiratory tract models for deposition and retention of plutonium aerosols. Pulmonary burdens of plutonium-239 for chronic exposure were calculated to range from 3.6 pCi to 1.2 nCi with more typical values of 2.4 and 5.9 pCi for radiation zone and office areas respectively.

Lymph burdens were calculated for chronic exposure of twenty years and were double the lung burden calculated in each instance. The lung burden represents an equilibrium condition in contrast to the lymph burden which does not reach equilibrium by the respiratory model used.

The apparatus subsequently described was effective in making the desired measurements and can be recommended in

a program of aerosol characterization. The aerosols characterized in plutonium facilities are highly variable, most often bimodal, and the smaller size fraction tends to be more uniform with time. The large variation between typical and maximum values of aerosol parameters for the radiation worker demands a better measure of the air inspired if meaningful predictions are to be made of individual deposition cases. On the other hand, the data suggest that zone aerosol characterizations are adequate for corridors, offices, etc. in the development of estimates of burdens. The inspired air may be considerably cleaner than that measured, or the lung model describing deposition is overly conservative because values predicted have not been observed in the data from a program of tissue sampling at autopsy. The new International Commission of Radiation Protection (ICRP) lung model is a most useful start in the development of a rational basis for assessment of internal contamination as a result of inhalation of plutonium aerosols. Instrument technology must now meet the challenge with devices capable of adequately determining for the individual worker, the necessary aerosol characterization parameters for correct interpretation of dose.

DISCUSSION

METHODS AND EQUIPMENT

Criteria around which the experimental plan was designed were as follows:

1. All air samples should contain detectable amounts of plutonium-239
2. The standard error in sampling and analysis should be within $\pm 25\%$.
3. The experiment could be conducted on a half-man per year continuing time effort.

The air samples were obtained by use of six constant-flow air samplers that were calibrated to sample at a flow rate of $1 \text{ ft}^3/\text{min}$. These giraffe-type portable samplers have a capacity of $1 \text{ ft}^3/\text{min}$ through a variable resistance of 0 to 20 in. Hg with the air delivery maintained automatically within $\pm 5\%$. The air movers operated satisfactorily with the provision that flow calibration be performed after the pump has been operating for 30 min. Calibration checks taken periodically over a one-year period of continuous operation indicated only minor changes in flow rate characteristics.

The particulate material was collected on Gelman GA-4 membrane filters which were mounted in 2 in. disposable filter holders. The filter and filter holder were chosen for ease of handling, storage and the surface collection characteristics of the membrane filter. Field trials indicated that the filter loading pressure drop over a one-week period of continuous operation was well within the operating parameters of the air mover. The weight of dirt and other particulates, averaged over 50 weekly samples, was found to be $12.8 \text{ mg}/10,000 \text{ ft}^3$ of air. The air mover, filter, and filter holder are shown in Figures 1 and 2.

The dirt laden filters were examined by alpha spectrometry to determine the alpha emitters present

and to determine concentrations per unit volume of air. The detectors employed were of the silicon semiconductor type, also known as surface

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FIGURE 1. Constant Flow Air Sampler

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FIGURE 2. Membrane Filter and Holder Showing Dust Loading

barrier detectors. The detectors used have an active area of either 450 or 950 mm² with a depletion depth of 60 μ and are available from several commercial sources.

The detectors were mounted in a vacuum chamber and operated with the Ortec 101-201 low noise amplifier system which is specifically designed for all signal processing between the semiconductor detector and pulse height analyzer. The biased post amplifier which is part of the 101-201 system, is extremely useful for suppression of portions of the spectrum and expansion of the region of interest over a greater number of channels on the analyzer.

The multichannel analyzer used was a Nuclear Data 512 channel unit with the 108 F, M and R units operational. The readouts utilized were a Tektronics 503 oscilloscope and a Monroe Solid State high speed printer. The internal amplifiers in the multichannel analyzer were not used since they are not completely compatible with the Ortec 101 preamplifier.

The alpha spectrometry system is pictured in Figures 3 and 4, and typical calibration and sample results are shown in Figure 5. Counting times used varied from 400 min to 1,000 min depending upon the alpha activity present on the sample. The background count for the system was found to be

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*FIGURE 3. Silicon Semiconductor
Surface Barrier Detectors*

10 ± 2 counts/1,000 min, based on 64 hr background counts taken over weekends.

The counting detection limit based upon a thousand minute count, 30% geometry, and a background subtracted count of 20 counts 1000 min, is about 0.070 disintegration min per sample for plutonium. The result significance was about $\pm 25\%$ for 1.0 standard deviation.

The plutonium particle sizes were then determined by autoradiographic techniques as previously reported in detail,⁽⁴⁾ and will be described briefly in this report.

The membrane air filters were contact exposed to standard nuclear

tract-plates, type NTA, which could be positioned reproducibly in the filter filmholder. Exposure times used ranged from 1,000 to 40,000 min with the exposure packets stored in a refrigerator during the exposure period to reduce track fading. Standard darkroom procedures were employed in film plate development, fixing and washing.

The film plates were examined with an optical microscope at 200X. The number of alpha tracks relating to each particle was tabulated and the entire plate was examined for particles exhibiting more than 10 tracks. When many small particles were present, a

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*FIGURE 4. Electronic Equipment
Used for Alpha Energy Analysis*

^{239}Pu Source - Log Display

Air Sample - Linear Display

FIGURE 5. Counter Calibration and Sample Results

representative area (5-10%) was carefully examined for particles represented by 10 tracks or less. With each group of exposures the number of background tracks was obtained from slides exposed to blank filters.

The particle size was then determined by conversion of the number of tracks per particle to disintegrations per minute, using appropriate correction factors for geometry and self-absorption similar to techniques used by Yagoda.⁽⁵⁾ Calculations were made relating particle diameter to alpha activity, assuming a spherical particle of plutonium oxide with a density of $\sim 11.5 \text{ g/cm}^3$. Since small amounts of other plutonium isotopes besides plutonium-239 are present, a correction factor was included for the isotopic content of the plutonium aerosol in question. The particle sizes were calculated on a unit-density basis so that the particle size distribution data would be directly applicable to the recently proposed lung model curves.⁽³⁾

RESULTS

PLUTONIUM AIR CONCENTRATIONS

The weekly air concentrations for each location are shown as histograms in Appendix B. Most of the sample locations are shown on building plans in Appendix A. Summary tabulations of the plutonium air concentration data are presented in Tables I, II, and III. The concentration is expressed as femto-curies of plutonium-239 per cubic meter of air sampled (fCi/m^3). The average, maximum, and minimum weekly plutonium air concentrations are shown as is the standard deviation of the average weekly plutonium air concentration.

The plutonium air concentrations did not show any unexpected trends and the values found could generally be related to the sampling location. The highest concentrations were found in those rooms where the largest amounts of material were handled, and active processing was being undertaken even

TABLE I. Plutonium Air Concentrations
Isochem Plutonium Purification and
Fabrication Facility

No.	Location Description	Sampling Period Weeks	Concentration fCi/m ³			
			Average	Standard Deviation	Maximum	Minimum
1	Corridor 1	14.0	2.9	2.2	9.8	1.1
2	Room 100	14.7	3.5	1.7	8.1	1.5
3	Duct Level Maint. Shop	14.7	4.2	2.1	9.8	1.3
4	Corridor 2	13.1	4.5	6.9	25.6	0.9
5	Final Inspection	11.3	6.0	3.0	11.1	1.9
6	Corridor 236	13.1	9.3	8.2	30.9	1.4
7	Corridor, Analytical Lab.	14.0	11.8	6.0	30.2	5.3
8	Room 167	13.9	12.5	8.6	38.0	7.0
9	Room 239-A	14.7	30.7	6.4	244.0	1.1
10	Fab. Line C-Line	13.3	55.6	340.0	348.0	1.8
11	Room 116 change room	11.2	394.0	1180.0	3970.0	11.7
12	Room 228 near 9-B	11.2	1340.0	3500.0	9260.0	18.1

TABLE II. Plutonium Air Concentrations
BNW Plutonium Metallurgy Facility

No.	Location Description	Sampling Period Weeks	Concentration fCi/m ³			
			Average	Standard Deviation	Maximum	Minimum
1	Foyer	14.2	2.0	1.2	5.3	0.9
2	Room No. 17	14.3	3.2	2.8	8.9	0.6
3	Hall No. 6	14.0	4.6	2.0	8.8	2.0
4	Room No. 35	14.0	13.2	20.0	73.9	1.5
5	Room 5	14.0	15.2	31.0	125.0	2.1
6	Change Room	14.3	49.5	160.0	626.0	1.1

though in closed hoods and glove boxes. The hallways or corridors between rooms showed only minor amounts of plutonium in the air samples. In all three buildings examined, the change room was found to have levels of plutonium air contamination well above the halls on either side. This is undoubtedly caused by resuspension of particles from contaminated clothing during clothing removal and handling.

The minimum weekly plutonium air concentration measured was 0.5 fCi/m³. It is of interest to compare this value to those reported for worldwide fallout from weapons testing. Perkins⁽⁶⁾ reported fallout concentrations of plutonium-239 ionizing from 0.035 to 0.48 fCi/m³ for a location near Richland, Washington, during 1962-1964. Results reported by the U.S. Public Health Service (US-PHS)⁽⁷⁾ from 11

TABLE III. Plutonium Air Concentrations
BNW Plutonium Development Laboratories

No.	Location Description	Sampling Period Weeks	Concentration fCi/m ³			
			Average	Standard Deviation	Maximum	Minimum
1	Main Hallway	10.5	2.3	2.3	8.1	0.8
2	Main Hallway	9.6	3.5	2.5	8.1	1.1
3	Room 146	8.3	4.1	1.7	6.2	1.2
4	Room 225	18.0	4.2	3.2	11.0	0.5
5	Corridor 2nd Floor	17.7	4.6	3.8	13.9	0.7
6	Change Room	12.0	4.8	2.6	9.9	1.5
7	Room 138	18.6	5.8	4.8	16.1	1.8
8	Back Dock	20.9	7.3	7.4	24.8	1.2
9	Room 131	10.4	10.0	7.2	23.8	1.0

of their Radiation Surveillance Network stations for November, 1965, through March, 1966, range from 0.03 to 0.18 fCi/m³. Drobinski⁽⁸⁾ of the US-PHS in another article listed plutonium-239 concentrations in air ranging from 0.07 to 0.44 fCi/m³. Thus, the maximum fallout values for plutonium-239 are similar to the minimum values found in buildings where plutonium-239 is handled extensively in large amounts.

PLUTONIUM PARTICLE SIZE

The particle size data was plotted on log-normal probability graphs and also as simple histograms. It was immediately apparent that the particle size distribution was in many cases bimodal or even multimodal in nature.

This is not unexpected when we consider that the samples were taken over a weekly period and represent the possible contribution from a number of sources. There seemed to be a small size fraction ranging up to about 0.5 μ and a number of larger particles ranging

in size up to about 10 μ . The number of larger particles found in any one sample was few, but the contribution to the total mass or activity was significant.

There is considerable advantage to interpreting the particle size distribution in terms of a log-normal distribution so that the final data may be utilized with the latest lung model. A semiempirical method for fitting bimodal distributions has been presented by Irani and Callis.^(9,10)

The final equation they developed is

$$P = 50 - 100 f \operatorname{erf} [\ln M / \bar{M}_1 / \ln \sigma_1] \\ - 100 (1 - f) \operatorname{erf} [\ln M / \bar{M}_2 / \ln \sigma_2], \quad (1)$$

where

P = percent by weight greater than size M,

M = diameter in microns,

$\bar{M}_1$ = activity median diameter (fraction one),

$\bar{M}_2$ = activity median diameter (fraction two),

σ_1 = geometric standard deviation of fraction one,
 σ_2 = geometric standard deviation of fraction two,
 f = fraction one,
 erf = error function from statistical tables.

Evaluation of the data was conducted by plotting the particle size data on log-normal probability graphs and also as a histogram as shown in Figures 6 and 7. The median sizes were then chosen by examination of the histogram and

lines 1 and 2 drawn as asymptotes on the log-normal probability plot. The next step is to obtain first approximations of σ_1 and σ_2 from these lines and compute the value of f at various values of P . If the value of f turns out to be independent of P and M , the estimated values of σ_1 and σ_2 are accurate. Otherwise, successive approximations are necessary. Table IV demonstrates the constancy of f for the data in Figures 6 and 7. This method was used to treat the data and

FIGURE 6. Heterogeneous Distribution
Case 2-10

FIGURE 7. Histogram of Case 2-10

TABLE IV. Heterogeneous Distribution
Case 2-10

M, μ	P (measured)	f (calculated)*
2.20	20	0.50
1.95	30	0.55
1.65	40	0.56
1.32	50	0.50
0.82	60	0.53
0.43	70	0.54
Average		0.51 ± 0.05

* Assuming $\bar{M}_1 = 0.4 \mu$, $\bar{M}_2 = 2.1 \mu$,
 $\sigma_1 = 1.6$, $\sigma_2 = 1.2$

resulted in the summary tabulations as shown in Tables V, VI and VII. A complete listing of the results for individual samples is shown in Appendix C.

PREDICTION OF RESPIRATORY TRACT BURDENS

The lung model used by the ICRP to develop values for Maximum Permissible Concentration Air (MPC)_a has been revised by the Task Group on Lung Dynamics of the ICRP.⁽³⁾ This revision provides a more realistic and quantitative model for the deposition and clearance of dusts and particulate matter from the respiratory tract.

The dust deposition model utilizes and depends on dust sampling data such as obtained here. The model describes the dust deposited in terms of three compartments: the nasopharynx (N-P) beginning with the anterior nares and

TABLE V. Plutonium Particle Size Distributions Isochem Plutonium Purification and Fabrication Facility

No.	Location Description	Fraction One			Fraction Two		
		AMD, μ *	σ_g **	f_1 ***	AMD, μ	σ_g	f_2
1	Corridor 1	0.14	1.22	1.00	-	-	-
2	Room 100	0.14	1.23	0.88	0.87	1.20	0.12
3	Maint. Shop (Duct Level)	0.15	1.27	0.87	1.15	1.15	0.13
4	Corridor 2	0.12	1.30	0.84	0.92	1.22	0.16
5	Final Inspection	0.23	1.78	0.83	2.22	1.18	0.17
6	Corridor 236	0.19	1.30	0.53	2.36	1.24	0.47
7	Corridor Analytical Lab.	0.19	1.36	0.57	2.11	1.25	0.43
8	Room 167	0.18	1.25	0.73	1.46	1.28	0.27
9	Room 239-A	0.19	1.25	0.47	3.58	1.42	0.53
10	Fab. Line (C)	0.27	1.40	0.46	3.28	1.22	0.54
11	Room 116, Change Room	0.32	1.45	0.43	2.20	1.30	0.57
12	Room 228 (Near 9B)	0.36	1.54	0.29	2.20	1.81	0.71

* Activity or Mass Median Diameter

** Geometric Standard Deviation

*** Fraction of Activity

TABLE VI. Plutonium Particle Size Distributions BNW Plutonium Metallurgy Facilities

No.	Location Description	Fraction One			Fraction Two		
		AMD, μ *	σ_g **	f_1 ***	AMD, μ	σ_g	f_2
1	Foyer	0.16	1.91	0.90	1.31	1.22	0.10
2	Room 17	0.17	1.33	0.62	2.35	1.12	0.38
3	Hall 6	0.43	1.16	0.65	1.49	1.26	0.35
4	Room 35	0.23	1.38	0.49	2.56	1.34	0.51
5	Room 5	0.21	1.26	0.49	1.97	1.45	0.51
6	Change Room	0.21	1.37	0.45	4.03	1.23	0.55

* Activity Median Diameter

** Geometric Standard Deviation

*** Fraction of Activity

TABLE VII. Plutonium Particle Size Distributions BNW Plutonium Development Laboratories

No.	Location Description	Fraction One			Fraction Two		
		AMD, μ *	σ_g **	f_1 ***	AMD, μ	σ_g	f_2
1	Main Hallway	0.14	1.65	0.55	2.1	1.32	0.45
2	Main Hallway	0.13	1.53	0.86	1.96	1.24	0.14
3	Room 146	0.19	1.38	0.72	1.47	1.26	0.28
4	Room 225	0.10	1.32	0.97	0.29	1.39	0.03
5	Corridor 2nd Floor	0.14	1.25	0.93	0.82	1.13	0.07
6	Change Room	0.15	1.77	0.89	3.32	1.18	0.11
7	Room 138	0.30	1.92	0.58	2.10	1.34	0.42
8	Back Dock	0.10	1.89	1.00	-	-	-
9	Room 131	0.21	1.21	0.52	2.38	1.15	0.48

* Activity or Mass Median Diameter

** Geometric Standard Deviation

*** Fraction of Activity

extending back and down to the level of the larynx; the trachea and bronchial tree (T-B) including the terminal bronchioles; and the pulmonary compartment (P). The deposition and clearance processes as schematically shown in the Task Group report are reproduced in Figure 8. The letters D_2 through D_5 are the fraction of the inhaled aerosol D_1 , which are exhaled or deposited in the various compartments. The Task Group concluded that these deposition fractions are dependent on the activity (or mass) median aerodynamic diameter (AMAD or MMAD) of

the aerosol. The deposition fractions of the nasopharynx and pulmonary compartments are strongly dependent on particle size, but are relatively independent of particle size in the tracheobronchia compartment. The Task Group has taken into account the breathing rate of the moderately active worker and the deposition fractions for 1450 ml tidal volume or ~ 20 liter/min. From these deposition fractions and a knowledge of the particle size of the aerosol inhaled, the depositions in the various respiratory compartments may be estimated.

FIGURE 8. Schematic Portrayal of All Dust Deposition Sites and Clearance Processes

The letters "a" through "j" in Figure 8 indicate the clearance pathways into the blood, G.I. tract and lymph. Each pathway clears a specific fraction of the deposited material, and the fraction cleared and the rate of clearance vary depending on the solubility of the deposited material. Aerosols have been divided into three general classes depending on the rate of clearance. Class D aerosols have a clearance half-time of <1 day to 10 days. Class W aerosols have half-times of 10 to 100 days and Class Y aerosols have half-times of >100 days. A tabulation of recommended clearance half-times for a number of compounds is appended to the Task Group report.

In the following discussion the aerosol is assumed to consist of plutonium-239 oxide, which is a Class Y material with relatively long retention. The recommended value for effective clearance half-time from the pulmonary region to blood is 500 days.

Let Q_{pe} be that quantity of material at any time t in compartment P which is subject to elimination via the blood, or pathway e, with an effective elimination constant λ_e . Similarly, the other quantities in compartment P can be described as

Q_{pf} = the amount subject to elimination from P via the G.I. tract, pathway f, with an elimination constant of λ_f ,

Q_{pg} = the amount subject to elimination from P via the G.I. tract, pathway g, with an elimination constant of λ_g ,

Q_{ph} = the amount subject to elimination from P via the lymph system, path-

way h, with an effective elimination constant of λ_h .

Note that $Q_{pe} + Q_{pf} + Q_{pg} + Q_{ph} = Q_p$, the total quantity of material in the pulmonary compartment at any time.

Letting D_1 be that quantity of aerosol inhaled per day, the change in subcompartment e of P with time may be written

$$dQ_{pe}/dt = -\lambda_e Q_{pe} + f_e D_5 D_1, \quad (2)$$

from which

$Q_{pe} = f_e D_5 D_1 / \lambda_e [1 - \exp(-\lambda_e t)]$. (3)
Similarly, the other subcompartment values are developed such that the quantity in the pulmonary compartment at any time t is given by

$$\begin{aligned} Q_p = & \left[f_e / \lambda_e [1 - \exp(-\lambda_e t)] \right. \\ & + f_f / \lambda_f [1 - \exp(-\lambda_f t)] \\ & + f_g / \lambda_g [1 - \exp(-\lambda_g t)] \\ & \left. + f_h / \lambda_h [1 - \exp(-\lambda_h t)] \right] \\ & D_1 D_5. \end{aligned} \quad (4)$$

If we assume that the period of inhalation is long compared to the effective elimination constant of 500 days, the equilibrium quantity in the pulmonary compartment becomes:

$$\begin{aligned} Q_p(\infty) = & D_1 D_5 (f_e / \lambda_e + f_f / \lambda_f \\ & + f_g / \lambda_g + f_h / \lambda_h). \end{aligned} \quad (5)$$

The values of the elimination parameters for plutonium oxide are⁽¹⁾

$$f_e = 0.05 \quad f_f = 0.40 \quad f_g = 0.40 \quad f_h$$

$$= 0.15,$$

$$\lambda_e = 1.4 \times 10^{-3} d^{-1} \quad \lambda_f = 6.9$$

$$\times 10^{-1} d^{-1} \quad \lambda_g$$

$$= 1.4 \times 10^{-3} d^{-1} \quad \lambda_h = 1.4 \times 10^{-3} d^{-1},$$

$$\lambda_i = 1.4 \times 10^{-3} d^{-1},$$

and

$$Q_p(\infty) = 4.3 \times 10^2 D_1 D_5. \quad (6)$$

The equilibrium burden may be calculated by knowing the daily quantity inhaled and the activity or mass median aerodynamic diameter of the aerosol such that D_5 may be found.

A similar calculation may be made to arrive at the quantity in the lymph nodes at any time t . It should be noted that of the material entering the lymph system, one fraction f_i , passes on to blood with an effective elimination constant of λ_i while the remainder is subject only to radioactive decay. Now:

$$dQ_{Li}/dt = \lambda_h^b f_h f_i D_1 D_5 / \lambda_h$$

$$[1 - \exp(-\lambda_h t)] - \lambda_i Q_{Li}, \quad (7)$$

where Q_{Li} is the fraction of the quantity in the lymph which is eliminated via the blood pathway, i ; λ_h^b is the biological elimination constant for pathway h . Also

$$dQ_{LS}/dt = \lambda_h^b f_h (1 - f_i) D_5 D_1 / \lambda_h$$

$$[1 - \exp(-\lambda_h t)] - \lambda_r Q_{LS}, \quad (8)$$

where Q_{LS} is the fraction of the quantity in the lymph which is subject only to radioactive decay with a constant of λ_r . Then

$$Q_{Li} = \lambda_h^b f_h f_i D_5 D_1 / \lambda_h \left[1/\lambda_i \right.$$

$$- 1/(\lambda_i - \lambda_h) \exp(-\lambda_h t)$$

$$- [1/\lambda_i - 1/(\lambda_i - \lambda_h)]$$

$$\exp(-\lambda_i t) \left. \right], \quad (9)$$

and

$$Q_{LS} = \lambda_h^b f_h (1 - f_i) D_5 D_1 / \lambda_h \left[1/\lambda_r \right.$$

$$- 1/(\lambda_r - \lambda_h) \exp(-\lambda_h t)$$

$$- [1/\lambda_r - 1/(\lambda_r - \lambda_h)] \exp$$

$$(-\lambda_r t) \left. \right]. \quad (10)$$

In the case of plutonium-239 oxide λ_r is essentially zero. Also $\lambda_i \sim \lambda_h \sim \lambda_h^b$ and the above expressions reduce approximately to

$$Q_{Li} = f_i f_h D_5 D_1 \left[1/\lambda_i [1 - \exp$$

$$(-\lambda_i t)] - t \exp(-\lambda_i t) \right], \quad (11)$$

$$Q_{LS} = f_h (1 - f_i) D_5 D_1 \left[t - 1/\lambda_h \right.$$

$$[1 - \exp(-\lambda_h t)] \left. \right]. \quad (12)$$

Since equilibrium is not reached in the lymph nodes during an individual lifetime, a period of 20 yr has been chosen to reflect the plutonium exposure to date. Since $Q_L = Q_{LS} + Q_{Li}$, substitution of constants into the sum of equations 11 and 12 yields $Q_L = 9.0 \times 10^2 D_5 D_1$ at 20 yr for plutonium-239 oxide.

A summary of all of the air concentration and particle size data collected is shown as Table VIII. To illustrate the use of both particle size data and aerosol deposition and clearance data, the pulmonary and lymph burdens are calculated for the typical and maximum characterizations for the radiation zone worker, those who frequent areas of high plutonium concentrations. The pulmonary and lymph burdens are also calculated for the typical and minimum characterizations for the nonradiation worker, who are typified by the office worker in a plutonium handling building. In the case of a distinct bimodal distribution, the burdens are calculated from

$Q_P = 430 [f_1 D_{51} + f_2 D_{52}] D_1$ for the two parts of the distribution where f_1 and f_2 are the fractions of D_{51} and D_{52} respectively. In the typical case for the radiation worker

$$\begin{aligned} Q_P &= 430 \text{ dy } [0.60 \times 0.39 + 0.40 \\ &\quad \times 0.14] 0.19 \text{ pCi/day} \\ &= 24 \text{ pCi.} \end{aligned}$$

Similarly, the values are calculated for the other distribution parameters and the results are presented in Table IX.

The lymph and pulmonary retention values shown in Table IX indicate that the lymph nodes may well be the organ receiving the largest radiation dosage. Not only is the total retention of the lymph twice that of the pulmonary region, but the weight of the lymph nodes of concern is considerably less. Pochin⁽¹¹⁾ measured the weight of the tracheobronchial lymph glands in ten individuals and found a mean weight for a population of ages 18 to 65 of about 15 g. The pulmonary region of the lungs may be about

TABLE VIII. Summary Table of Plutonium Air Concentration and Particle Size Distribution Data

Facility	fCi/m ³	Fraction One			Fraction Two		
		AMD, μ	σ_g	f_1	AMD, μ	σ_g	f_2
Work Areas							
PP&FF	30.7	0.24	1.36	0.50	2.46	1.36	0.50
PMF	20.6	0.27	1.29	0.52	2.51	1.32	0.48
P D L	6.5	0.18	1.61	0.78	2.16	1.20	0.22
Typical	19.0	0.28	1.40	0.60	2.40	1.30	0.40
Maximum	1340.0	0.36	1.54	0.29	2.20	1.81	0.71
Office Areas							
PP&FF	4.2	0.16	1.35	0.88	1.29	1.19	0.12
PMF	2.6	0.16	1.62	0.76	1.83	1.17	0.24
P D L	3.7	0.14	1.47	0.78	1.45	1.30	0.22
Typical	3.5	0.15	1.50	0.80	1.50	1.20	0.20
Minimum	2.0	0.16	1.91	0.90	1.31	1.22	0.10

TABLE IX. Estimates of Pulmonary and Lymph Retention of Plutonium-239 for Chronic Exposure to Plutonium Oxide Aerosols

	D_1 pCi/d	AMD, μ	D_5 Effective	Q_P pCi	Q_{L20} pCi
Radiation Zone Worker					
Typical	0.19	0.28/2.4	0.29	24	49
Maximum	13.4	0.36/2.2	0.20	1200	2400
Office Worker					
Typical	0.035	0.15/1.5	0.39	5.9	12
Minimum	0.02	0.16/1.3	0.42	3.6	7.5

half of the total weight of the lung or about 500 g since the weight of the lungs in the standard man⁽¹²⁾ has been fixed at 1000 g. The available results from actual analysis of human lungs and lymph nodes⁽¹³⁻¹⁵⁾ indicate that the plutonium concentration in the lymph nodes is on the order of five to ten times that in the lung. It is predicted by the respiratory model that this difference in concentration will get larger with residence time as the plutonium leaves the lung.

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APPENDIX A

Appendix A contains building plans which show most of the air sampling locations used in this study. Sample location numbers are the same as those shown in Tables I through VII in the main body of the report.

First Floor Plan

FIGURE A-1: Plutonium Purification and Fabrication Facilities

First Floor Plan

FIGURE A-2. Plutonium Purification and Fabrication Facilities

Floor Plan

FIGURE A-3. Plutonium Metallurgy Building

FIGURE A-4. Plutonium Development Laboratories

Second Floor Plan

FIGURE A-5. Plutonium Development Laboratories

APPENDIX B

Appendix B contains graphs showing the changes in plutonium air concentration at each sample location during the 10 to 20 weeks of sampling. The sample locations are shown in the same order as in Tables I through VII in the main report.

FIGURE B-1. Plutonium Air Concentration

FIGURE B-2. Plutonium Air Concentration

FIGURE B-3. Plutonium Air Concentration

FIGURE B-4. Plutonium Air Concentration

FIGURE B-5. Plutonium Air Concentration

FIGURE B-6. Plutonium Air Concentration

FIGURE B-7. Plutonium Air Concentration

FIGURE B-8. Plutonium Air Concentration

FIGURE B-9. Plutonium Air Concentration

FIGURE B-10. Plutonium Air Concentration

FIGURE B-11. Plutonium Air Concentration

FIGURE B-14. Plutonium Air Concentration

FIGURE B-15. Plutonium Air Concentration

FIGURE B-12. Plutonium Air Concentration

FIGURE B-13. Plutonium Air Concentration

FIGURE B-16. Plutonium Air Concentration

FIGURE B-17. Plutonium Air Concentration

FIGURE B-18. Plutonium Air Concentration

FIGURE B-19. Plutonium Air Concentration

FIGURE B-20. Plutonium Air Concentration

FIGURE B-21. Plutonium Air Concentration

FIGURE B-22. Plutonium Air Concentration

FIGURE B-23. Plutonium Air Concentration

FIGURE B-24. Plutonium Air Concentration

FIGURE B-25. Plutonium Air Concentration

FIGURE B-26. Plutonium Air Concentration

FIGURE B-27. Plutonium Air Concentration

APPENDIX C

Appendix C is a tabulation of the particle size parameters. It includes activity median diameters (AMD, μ), geometric standard deviations (σ_g) and fraction of activity (f_1 and f_2) for each of the air samples examined for particle size.

PARTICLE SIZE DISTRIBUTION DATA

Sample Location No.		Fraction One			Fraction Two		
		AMD, μ	σ_g	f_1	AMD, μ	σ_g	f_2
1. Plutonium Purification and Fabrication Facility							
1	2	0.13	1.25	1.0	-	-	-
1	4	0.12	1.20	1.0	-	-	-
1	1	0.15	1.16	1.0	-	-	-
1	12	0.15	1.25	1.0	-	-	-
2	5	0.14	1.15	0.84	0.80	1.20	0.16
2	7	0.13	1.15	0.83	1.10	1.18	0.17
2	11	0.16	1.46	1.00	-	-	-
2	12	0.13	1.12	0.83	0.70	1.21	0.17
3	3	0.15	1.17	0.89	1.20	1.17	0.11
3	4	0.14	1.40	0.83	1.00	1.15	0.17
3	8	0.12	1.40	0.80	1.30	1.15	0.20
3	11	0.18	1.15	0.96	0.90	1.11	0.04
4	3	0.14	1.20	0.85	1.10	1.22	0.15
4	4	0.11	1.12	0.83	1.00	1.11	0.17
4	5	0.11	1.64	1.00	-	-	-
4	8	0.12	1.12	0.68	0.80	1.28	0.32
5	4	0.21	1.24	0.50	2.50	1.20	0.50
5	5	0.34	2.13	1.0	-	-	-
5	6	0.20	2.10	1.0	-	-	-
5	8	0.14	1.27	0.81	1.50	1.12	0.19
6	2	0.19	1.30	0.73	1.70	1.12	0.27
6	5	0.14	1.20	0.64	1.20	1.34	0.36
6	7	0.24	1.40	0.24	3.30	1.30	0.76
6	8	0.22	1.40	0.52	2.10	1.14	0.48
7	1	0.25	1.22	0.44	3.00	1.20	0.56
7	6	0.22	1.45	0.49	2.3	1.39	0.51
7	8	0.16	1.28	0.76	0.95	1.20	0.24
7	10	0.17	1.50	0.58	1.30	1.19	0.42
8	5	0.20	1.20	0.60	1.60	1.13	0.40
8	8	0.15	1.15	0.80	1.00	1.25	0.20
8	9	0.18	1.21	0.80	0.55	1.83	0.20
8	10	0.20	1.44	0.70	2.20	1.12	0.30
9	2	0.40	1.40	0.22	8.10	1.50	0.78
9	11	0.18	1.17	0.85	1.70	1.18	0.15
9	12	0.14	1.30	0.82	1.10	1.11	0.18
9	13	-	-	-	0.78	1.45	1.00
10	3	0.18	1.40	0.53	1.20	1.18	0.47
10	8	0.39	1.50	0.25	6.00	1.18	0.75
10	9	0.20	1.38	0.37	1.30	1.33	0.63
10	10	0.36	1.36	0.67	2.50	1.16	0.33

Sample Location No.		Fraction One			Fraction Two		
		AMD, μ	σ_g	f_1	AMD, μ	σ_g	f_2
1. <u>Plutonium Purification and Fabrication Facility (contd)</u>							
11	3	0.35	1.60	0.24	2.80	1.33	0.76
11	5	0.30	1.40	0.50	1.70	1.40	0.50
11	8	0.23	1.26	0.48	1.90	1.21	0.52
11	10	0.40	1.60	0.50	2.10	1.25	0.50
12	3	0.40	1.67	0.68	1.9	1.15	0.32
12	5	-	-	-	2.5	1.84	1.00
12	7	-	-	-	2.00	2.20	1.00
12	8	0.30	1.35	0.49	2.2	1.38	0.51
2. <u>Plutonium Metallurgy Facility</u>							
1	-2	0.30	2.80	1.0	-	-	-
1	-3	0.11	1.20	0.77	1.1	1.26	0.23
1	-4	0.11	1.96	1.0	-	-	-
1	-6	0.05	1.92	1.0	-	-	-
1	-10	0.26	1.35	0.74	1.5	1.22	0.26
2	-2	0.18	1.40	0.75	1.5	1.18	0.25
2	-3	0.20	1.43	0.53	2.4	1.08	0.47
2	-4	0.14	1.14	0.59	2.8	1.12	0.41
3	-1	0.20	1.20	0.37	1.6	1.31	0.63
3	-3	0.18	1.15	0.87	1.1	1.17	0.13
3	-4	0.18	1.20	0.83	1.1	1.20	0.17
3	-11	0.13	1.10	0.53	1.6	1.25	0.47
4	-5	0.20	1.43	1.0	-	-	-
4	-7	0.30	1.37	0.41	2.7	1.15	0.59
4	-11	0.27	1.25	0.19	2.6	1.35	0.81
4	-12	0.20	1.30	0.34	2.4	1.50	0.66
5	-4	0.20	1.21	0.30	1.7	1.68	0.70
5	-5	0.19	1.16	0.65	1.7	1.12	0.35
5	-10	0.30	1.65	0.36	2.5	1.65	0.65
5	-14	0.18	1.17	0.66	1.8	1.10	0.34
6	-1	0.30	1.27	0.41	3.1	1.19	0.59
6	-2	0.15	1.15	0.51	0.94	1.34	0.49
6	-5	0.53	1.42	0.06	6.8	1.21	0.94
6	-8	0.18	1.55	0.83	0.84	1.14	0.17
3. <u>Plutonium Development Laboratories</u>							
1	-2	0.19	1.18	0.36	1.8	1.27	0.64
1	-3	0.09	2.2	1.0	-	-	-
1	-5	0.22	1.22	0.21	2.5	1.18	0.79
1	-6	0.17	1.17	0.63	1.7	1.12	0.37
2	-1	0.15	1.4	1.0	-	-	-
2	-2	0.07	2.1	1.0	-	-	-
2	-3	0.24	1.3	0.50	2.1	1.24	0.50
2	-5	0.11	1.2	0.95	0.6	1.22	0.05
3	-2	0.19	1.29	0.69	1.1	1.33	0.31
3	-3	0.24	1.83	0.64	2.0	1.20	0.36
3	-4	0.17	1.16	0.83	0.8	1.35	0.17
3	-6	0.18	1.30	0.73	1.6	1.19	0.27
4	-2	0.12	1.13	0.93	0.29	1.41	0.07
4	-3	0.08	1.60	1.0	-	-	-
4	-4	0.08	1.44	1.0	-	-	-
4	-5	0.12	1.10	0.94	0.29	1.38	0.06
5	-2	0.15	1.20	0.87	1.10	1.12	0.13
5	-4	0.12	1.10	0.92	0.65	1.10	0.08
5	-5	0.12	1.11	0.92	0.53	1.19	0.08
5	-7	0.16	1.55	1.00	-	-	-

Sample Location No.		Fraction One			Fraction Two		
		AMD, μ	σ_g	f_1	AMD, μ	σ_g	f_2
3. <u>Plutonium Development Laboratories</u> (contd)							
6	-1	0.17	1.82	1.0	-	-	-
6	-2	0.12	1.11	0.83	0.58	1.24	0.17
6	-3	0.16	2.13	1.0	-	-	-
6	-6	0.13	1.95	0.72	0.94	1.15	0.28
7	-2	0.35	1.55	0.55	2.1	1.10	0.45
7	-3	0.25	1.6	0.60	2.0	1.25	0.40
7	-4	0.30	2.0	0.64	1.4	1.36	0.36
7	-5	0.30	2.56	0.52	2.7	1.62	0.48
8	-2	0.15	1.80	1.0	-	-	-
8	-3	0.10	2.00	1.0	-	-	-
8	-4	0.05	1.88	1.0	-	-	-
9	-1	0.23	1.25	0.34	3.0	1.07	0.66
9	-2	0.23	1.17	0.65	2.2	1.08	0.35
9	-3	0.14	1.14	0.54	1.5	1.31	0.46
9	-4	0.24	1.33	0.53	2.3	1.16	0.47

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