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**PARTICLE SIZE DISTRIBUTION OF
AIRBORNE PLUTONIUM NEAR A
CHEMICAL SEPARATIONS FACILITY**

**DON D. GAY
JAMES R. WATTS**

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MASTER

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PREPARED FOR THE U. S. DEPARTMENT OF ENERGY UNDER CONTRACT DE-AC09-76SR00001

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by

Don D. Gay and James R. Watts

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ABSTRACT

Particulate matter containing Pu-238 and Pu-239,240 emitted from a stack at the Savannah River Plant was collected and fractionated by size with cascade impactors. Two impactor measurements were made at a distance of 200 meters from the H-Area stack and a third was at a distance of 420 meters. Annual collections were made for three years. The Pu-238 and Pu-239,240 concentrations were determined for each of the size fractions. The fractions collected at 200 m from the 62 m stack showed a biphasic curve for both Pu-238 and Pu-239,240. The highest concentrations for both Pu-238 and Pu-239,240 were found in the $<0.49 \mu\text{m}$ fraction, with the secondary peak occurring in the fractions greater than $3.0 \mu\text{m}$. At 420 meters from the stack, the size distribution curves for Pu-238 and Pu-239,240 were not biphasic; only one peak appeared and that was for the fraction less than $0.49 \mu\text{m}$.

The doses to different organs from plutonium inhalation were calculated for the particle size distributions obtained in this study. The maximum potential 50-year dose commitment to the bone, the critical organ, for one year continuous exposure was 13 mrem for Pu-238 and 0.44 mrem for Pu-239,240 at 200 meters from the H-Area stack.

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PARTICLE SIZE DISTRIBUTION OF AIRBORNE PLUTONIUM NEAR A CHEMICAL SEPARATIONS FACILITY

INTRODUCTION

The chemical separations plants at F and H Areas on the Savannah River Plant site release small amounts of Pu-238 and Pu-239,240 from their stacks into the environment. The plutonium is nearly always attached to non-radioactive particles, usually crustal materials. Extensive characterization and measurements of plutonium-bearing particles in the off-gas systems at SRP's chemical separation facilities have been done by Sanders.¹⁻⁵ The geometric mean (with the standard deviation) of particles collected from the sand filter effluent and at the 50 foot level of the F-Area stack was $5.43 \pm 2.69 \mu\text{m}$.⁵ Particle size distributions of plutonium-bearing particles in an off-gas system at Los Alamos National Laboratory using Anderson eight stage impactors^{6,7} showed activity median aerodynamic diameters (AMAD) of $4.0 \mu\text{m}$ and $2.7 \mu\text{m}$ from fabrication facilities, and $0.3 \mu\text{m}$ for a recovery facility.

This study was undertaken to delineate the particle size distributions of plutonium-bearing particles in areas surrounding a chemical separations facility. In addition, 50-year dose commitments to different human organs (lung, bone, and liver) were calculated from the particle size distributions obtained.

METHODS

A Sierra Instruments Series 230, Model 235, High Volume Cascade Impactor equipped with Whatman filter papers was used to collect the Pu-bearing particles of different sizes released from the H-Area chemical separations facility. The impactor has five fractionation stages plus a backup filter. The backup filters were either asbestos or a glass fiber matrix. The equivalent aerodynamic diameters (μm) for the five stages for 50 percent collection efficiency and at an intake rate of $1.13 \text{ m}^3/\text{min}$ were:

stage 1	> 7.2
2	3.0 - 7.2
3	1.5 - 3.0
4	0.95 - 1.5
5	0.49 - 0.95
backup	< 0.49

The average flow rate was 1.10 m³/min. The impactor was placed northwest of the H-Area stack at distances of either 200 meters or 420 meters from the 62 meter stack and 1.1 meters above ground.

The Whatman filter papers were changed weekly from September 1975 through September 1978 but were combined by stages into three sampling periods, each of one year duration. The sampling periods were:

September 18, 1975 to September 15, 1976
September 16, 1976 to September 15, 1977
September 16, 1977 to September 15, 1978

All plutonium analyses were done at the Savannah River Laboratory by standard procedures.⁸

RESULTS

Releases from the chemical separations facility at H-Area are predominantly Pu-238 with lesser quantities of Pu-239,240. The plutonium concentrations over the study period for the various particle sizes are shown in Table 1. For either Pu-238 or Pu-239,240 and for all sampling periods, the largest amount of activity is associated with the smallest size fraction collected, i.e., the < 0.49 μ m fraction. The data in Table 1 show that an unusually large amount of Pu-238 was present during the 1976-1977 period.

The particle size fractionation pattern for Pu-238 and Pu-239,240 (Figures 1 and 2) shows graphically a biphasic plot for Pu-238 and Pu-239,240 for the material collected 200 m from the H-Area stack. The larger peak in each case is for the 0.49 μ m fraction. The secondary peak corresponding to the fraction > 3.0 μ m disappeared at 420 m.

The amount of Pu-238 released from the stack during the 1975-1976 sampling period (Table 2) was 14.84 mCi.^{9,10} During this period, 19.5 pCi of Pu-238 was collected with an average median particle diameter range of 0.45 - 0.50 μ m. In the 1976-1977 period, the Pu-238 released was 2.27 mCi, a factor of 6 less than the previous period.^{10,11} However the Pu-238 collected was 667 pCi, a factor of 34 greater than the previous period. The median particle diameter range increased to 1.00 - 1.20 μ m. For the collection 420 m from the stack in 1977-1978, the median aerodynamic particle diameter range of Pu-238 collected was 0.35 - 0.45 μ m; 279 pCi was collected from 7.68 mCi released.^{11,12} The Pu-239,240 activity for the 420 m sampling distance in 1977-1978 shows a median aerodynamic particle diameter range of 0.4 - 0.6 μ m. There was no Pu-239,240 released from H-Area, but 20.8 pCi was collected, presumably from F-Area, resuspension, and fallout.

The 50-year dose commitment was calculated on the basis of the inhalation of plutonium according to the measured size distributions. The effective particle size for each stage of the cascade impactor was calculated as the geometric mean of the limits. Assumed lower and upper limits were 0.01 μm and 10 μm , respectively. Respiratory tract deposition data were obtained from the Task Group Lung Model.¹³ Clearance factors and rates were those recommended in ICRP Publication 19 (Reference 14).

The 50-year dose commitments to the lung, bone, and liver for the three time periods are shown in Table 3. The maximum 50-year dose commitment for one year continuous exposure is 13 mrem to the bone. The differences in dose commitments are primarily due to variation in the fraction of particles in the less than 0.49 μm range. The deposition in the pulmonary section of the respiratory tract is over twice as large for this size as for any other impactor size range.

CONCLUSION

The particle size fraction collected which contains the largest percentage of activity for either Pu-238 or Pu-239,240 is the fraction < 0.49 μm . This size fraction contains the most activity whether at 200 m or 420 m from the H-Area stack. Based on this observation, it seems reasonable to assume that this size fraction also contains the most activity within the stack.

In the event the size distribution is not known when calculating a dose to man from SRP chemical separation operations, a realistic estimate of the dose can be obtained by using a particle size of 0.5 μm in the calculation.

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TABLE 1.

Measured Air Concentration of Plutonium Near a Chemical Separations Facility

			Particle Size Range, (μm)						
Radio-nuclide	Sampling Distance, m	Sampling Period, yr	>7.2	7.2 - 3.0	3.0 - 1.5	1.5 - 0.95	0.95 - 0.49	<0.49	
			Pu Concentration, 10 ⁻¹⁸ Ci/m ³						Total
²³⁸ Pu	200	75-76	2.96 _{+0.06} [*]	3.40 _{+0.06} [*]	1.61 _{+0.04}	3.79 _{+0.06}	3.51 _{+0.14}	15.3 _{+1.0}	30.6
	200	76-77	178 ₊₁	256 ₊₁	93.4 _{+0.7}	83.5 _{+0.6}	121 ₊₁	414 ₊₄	1145.9
	420	77-78	6.20 _{+0.17}	9.37 _{+0.21}	9.64 _{+0.2}	30.1 _{+0.4}	139 ₊₁	293 ₊₅	487.3
^{239,240} Pu	200	75-76	4.14 _{+0.07}	1.36 _{+0.03}	0.66 _{+0.03}	1.17 _{+0.03}	2.72 _{+0.11}	5.07 _{+0.5}	15.1
	200	76-77	2.84 _{+0.14}	3.27 _{+0.14}	1.54 _{+0.08}	2.63 _{+0.10}	6.17 _{+0.17}	16.0 _{+0.7}	32.5
	420	77-78	0.89 _{+0.07}	1.14 _{+0.07}	1.48 _{+0.08}	4.45 _{+0.16}	13.2 _{+0.2}	14.7 _{+1.1}	35.9

* Standard error in counting.

TABLE 2.

Plutonium Collected Annually by Cascade Impactors Near a Chemical Separations Facility

Radio-nuclide	Sampling Period	Distance of Sampler from Stack, m	Amount Pu Released, 10^{-3} Ci	Pu Collected by Cascade Impactor, 10^{-12} Ci	Air Concentration, 10^{-18} Ci/m ³	Median Aerodynamic Particle Diameter Range, μ m
²³⁸ Pu	75-76	200	14.84	19.5 $\pm$ 0.78*	30.6 $\pm$ 1.2*	0.45-0.50
	76-77	200	2.27	667 $\pm$ 6.67	1146 $\pm$ 11	1.00-1.20
	77-78	420	7.68	279 $\pm$ 5.58	487 $\pm$ 10	0.35-0.45
^{239,240} Pu	75-76	200	0.12	9.7 $\pm$ 0.49	15.1 $\pm$ 0.8	1.00-1.20
	76-77	200	0.02	18.9 $\pm$ 0.76	32.5 $\pm$ 1.3	0.40-0.50
	77-78	420	0.00	20.8 $\pm$ 1.04	35.9 $\pm$ 1.8	0.40-0.60

* Standard error in counting.

TABLE 3.

**The 50-Year Dose Commitment to an Individual From One-Year Exposure
near a Chemical Separations Facility**

<u>Radionuclide</u>	<u>Sampling Period, yr</u>	<u>Sampling Distance, m</u>	<u>50-Year Dose Commitment For a One Year Occupational Exposure, mrem</u>			<u>50-Year Dose Commitment From One Year Continuous Exposure, mrem</u>		
			<u>Lung</u>	<u>Bone</u>	<u>Liver</u>	<u>Lung</u>	<u>Bone</u>	<u>Liver</u>
^{238}Pu	75-76	200	0.04	0.12	0.05	0.14	0.43	0.19
	76-77	200	1.2	3.8	1.7	4.0	13	5.8
	77-78	420	0.76	2.2	1.0	2.6	7.6	3.4
$^{239,240}\text{Pu}$	75-76	200	0.01	0.04	0.02	0.05	0.16	0.07
	76-77	200	0.04	0.12	0.05	0.13	0.43	0.18
	77-78	420	0.04	0.13	0.06	0.15	0.44	0.19

FIGURE 1. ^{238}Pu Distribution for the Particle Size Fractions Collected by a Cascade Impactor Adjacent to a Chemical Separations Facility

FIGURE 2. $^{239,240}\text{Pu}$ Distribution for the Particle Size Fractions Collected by a Cascade Impactor Adjacent to a Chemical Separations Facility