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Resuspended Particles at
Rocky Flats During July 1973**

by
G. A. Sehmel

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This work was done for the U.S. Energy Research
and Development Administration under Contract
E(45-1)-1830 with Battelle, Pacific Northwest
Laboratories and under Contract E-(29-2)-3533
with Rockwell International, Atomics International
Division

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AIRBORNE ^{238}Pu AND ^{239}Pu ASSOCIATED WITH THE
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ABSTRACT

In July 1973, a field experiment was conducted at Rocky Flats to measure plutonium resuspension. Previously, for this experiment, airborne plutonium concentrations in respirable size ranges were reported. Concentrations were measured at three sites between the eastern security fence and eastern cattle fence. On 13 nonrespirable particle diameter ranges between 10 and 210 μm , ^{239}Pu concentrations ranged from 10^{-8} to 10^{-4} $\mu\text{Ci/g}$. Airborne ^{238}Pu concentrations as a function of particle diameter ranged from 10^{-9} to 10^{-5} $\mu\text{Ci/g}$. Both ^{238}Pu and ^{239}Pu were found on all particle diameter ranges. Airborne plutonium fluxes on nonrespirable particles decreased with both height and distance. The nonrespirable total ^{239}Pu flux ranged from 1×10^{-7} to 6×10^{-4} $\mu\text{Ci/m}^2/\text{day}$.

Airborne $^{238}\text{Pu}/^{239}\text{Pu}$ ratios were always equal to or greater than similar ratios reported measured on the ground surface. Ground surface ratios ranged from 1.5×10^{-2} to 2.2×10^{-2} . Only near the eastern security fence were airborne ratios similar to ground level ratios. At the eastern security fence, airborne $^{238}\text{Pu}/^{239}\text{Pu}$ ratios ranged from 0.2 to 0.4. These airborne ratios were one order of magnitude greater than ground surface ratios. Increased ratios indicate ^{238}Pu is more readily resuspended at greater distances from the eastern security fence than near the eastern security fence.

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INTRODUCTION

In Battelle, Pacific Northwest Laboratory experiments at Rocky Flats in 1973, some evidence⁽¹⁾ was obtained indicating the importance of airborne plutonium transport associated with nonrespirable particles resuspended from contaminated areas. These results include data showing plutonium concentration per gram of soil particles collected in two different air samplers. One sampler was a high-volume filter sampler facing towards the original oil storage area. The second air sampler was a high volume cascade particle impactor preceded by a settling chamber. This settling chamber is referred to as a "cowl". Particles larger than about 60 μm tend to settle out within the cowl and not be collected within the particle cascade impactor. This cowl sampler was faced into the wind by a wind vane.

Airborne particle samples collected at heights near the ground showed a higher plutonium concentration in $\mu\text{Ci/g}$ of soil collected on high volume filter samples than $\mu\text{Ci/g}$ for total airborne particles collected within the particle cascade impactor. Up to twice as much plutonium was associated with airborne soil collected on the high volume air sampler than was associated with particles collected on the stages of the impactor, gram for gram. Since large nonrespirable particles had been excluded from particle cascade impactors by settling in the cowls, these excluded nonrespirable cowl particles may have contained enough plutonium to account for apparent differences in air concentrations between different air sampler types.

In the original experiment, plutonium concentrations on particle cascade impactor stages were measured. Cowl collected soil was subsequently weighed and sized into fractions, but were not analyzed for plutonium at that time. Some soil samples collected in cowls were retained. These retained samples were analyzed for plutonium for the present report.

DESCRIPTION OF ORIGINAL EXPERIMENT

Some understanding of the original experiment is needed to know how the retained nonrespirable cowl samples were collected. A schematic of the July 1973 Rocky Flats resuspension experiment is shown in Figure 1. In this figure are shown three air sampler sites. Site A was located along the eastern security fence near the original oil storage area. Contaminated oil barrel leakage⁽²⁾ in the storage area was the original Pu resuspension source. Subsequently the storage area was covered with asphalt. Resuspension in 1973 occurred from Pu which had been suspended and redeposited from the storage area prior to asphalt covering.

Neg. No. PNL-744429-3

FIGURE 1. Schematic of Rocky Flats Resuspension Tower Locations

Site B was east near the eastern cattle fence. Site AB was between A and Site B. The distance between Sites A and AB was 392 m. From Site AB to B the distance was 277 m. These two distances were added to approximate the

distance between Sites A and B as being 669 m. Both high volume* and cascade particle impactors** were located at each air sampling site.

NONRESPIRABLE AIR SAMPLERS

Nonrespirable airborne particles were collected with cascade particle impactor cowl systems schematically shown in Figure 2. This system⁽³⁾ was assembled from a 0.57 m³/min flowrate Andersen 2000 Incorporated high-volume cascade particle impactor and a rotating cowl attached to the impactor. These impactors separate particles into nominal aerodynamic diameter ranges of 1.1, 2.0, 3.3, and greater than 7 μ m, which are stage 50% cutoff diameters for unit density spheres.

Neg. No. PNL-744581-12

FIGURE 2. Rotating Cowl and Impactor

*General Metal Works, Inc., Model GMWL-2000-hi-vol air sampler with filter holder, 8368 Bridgetown Road, Cleves, Ohio 45002.

**Andersen 2000, Inc., Model 65-100 High-Volume Sampler Head, P.O. Box 20769, AMF, Atlanta, Georgia 30302.

The cowl system will be briefly described. Each cowl system was attached to an air sampling tower through the system support arm. A variable transformer attached to the support arm was used to control air flow rates through the particle cascade impactor. The particle cascade impactor inlet faced downward, but the cowl inlet was horizontal. In order to attach the wind direction sensitive rotating cowl to the impactor, the factory supplied speed ball handle on the impactor inlet was replaced with a threaded spindle extension. The cowl was attached to the spindle extension by a bolt passing through the spindle bearing housing. With the spindle extension attached to the cowl, the wind vane rotated the cowl inlet into the wind. Particles entering the 15 cm diameter cylindrical cowl sampler inlet either settled on the cowl floor or were drawn up into the impactor. Particles settling on the cowl floor will be called "nonrespirable" in this report. These were the particles analyzed for plutonium content.

NONRESPIRABLE PARTICLE SIZING

Airborne nonrespirable particles collected at 0.3 m at Sites A and AB were sized into diameter increments using the sieve ranges of a sonic sifter.* Sieve sizes were 10, 20, 37, 53, 63, 74, 86, 105, 125, 149, 177, and 210 μm . Particles less than 10 μm were retained as an additional size increment.

AIR SAMPLER LOCATIONS

Only portions of Figure 1 are important for the present analysis. These are the locations of particle cascade impactors with cowls. At Sites A and B, these cowl samplers operated for all wind speeds. In contrast, the cowl systems at Site AB were operated as a function of wind speed. One series of these cowl samplers was operated continuously for all wind speeds. Another set operated for wind speeds between 4.1 and 6.3 m/sec and is called the mid-wind speed increment. The third wind speed increment was from 6.3 to 9.8 m/sec and is called the high-wind speed increment.

*Allen Bradley Co., Model L3P Sonic Sifter, Milwaukee, Wisconsin.

Cowl samples were retained after the original experiment for several sampler locations. At Site AB, 0.3 m samples were retained for all three wind speeds. At the 1 m height, samples were retained only for the mid- and high-wind speed increments. At the 2 m height, the mid-wind speed increment cowl sample was retained. At the 10 to 11 m height, cowl samples were retained for both mid- and high-wind speed increments.

Since nonrespirable particles collect within cowls by inertial collection, cowl samples at each height were combined to obtain the total average airborne flux at each height. This means the two samples obtained at 10 and 11 m heights were combined into one sample. Similarly at the 1 m height, two samples were combined. At the 0.3 m height, three samples were combined. Samples at the 0.3 m height were sized into 13 diameter increments.

At Site A, the 0.3 m soil sample was sized. The sample collected at the 2 m height had not been sized due to the small soil weight collected with this cowl. Similarly at Site B, only total samples at 2 m and 10 m heights were analyzed. All other samples collected in the other cowls had been discarded after the original experiment.

OBJECTIVES

Current objectives were to determine plutonium transport on nonrespirable particles collected in settling chamber cowls during an experiment at Rocky Flats in July 1973. Results of plutonium analyses were to be used to calculate plutonium distributions on airborne nonrespirable particles as (1) a function of particle size, (2) a function of the mass of airborne soil, (3) a function of height, and (4) a function of distance. Also, ratios of $^{238}\text{Pu}/^{239}\text{Pu}$ were to be calculated and these airborne ratios compared to reported^(4,5) ratios measured for ground surfaces.

EXPERIMENTAL

As previously indicated, samples collected within nondiscarded cowl samples were analyzed for ^{238}Pu and ^{239}Pu . Samples were analyzed for heights

0.3, 1, 2, and 10 m. Samples collected at 0.3 m were sized into 13 size fractions and each size fraction analyzed for ^{238}Pu and ^{239}Pu . Samples collected from 1 to 10 m above ground were not sized. In these cases, total nonrespirable samples were analyzed for plutonium. A total of 31 plutonium analyses were obtained. These Pu results were combined with sampling times and soil weights to determine several calculated parameters.

Plutonium results were obtained from the LFE Laboratories.* Samples were dissolved and plutonium separated for final electrodeposition and radiochemical analyses. Radiochemical results were obtained for both ^{238}Pu and ^{239}Pu .

RESULTS AND DISCUSSION

Radiochemical results indicate airborne fluxes of both ^{238}Pu and ^{239}Pu on nonrespirable particles during July 1973 at Rocky Flats. Several results were calculated from the radiochemical analyses. These include airborne fluxes of both ^{238}Pu and ^{239}Pu , Pu concentrations in $\mu\text{Ci/g}$ of airborne soil, and $^{238}\text{Pu}/^{239}\text{Pu}$ ratios on airborne nonrespirable soil.

Since there was less ^{238}Pu present than ^{239}Pu in the samples, the radiochemical counting statistics for ^{238}Pu results were not as good as for ^{239}Pu results. If error bars in figures showing results are not shown around Pu data points, one sigma counting statistic errors were less than 5%. For larger counting statistic errors, one sigma error bars are shown around Pu results. If counting statistics errors were greater than 100%, error bar lower limits are indicated in the figure by arrows extending from the data point.

In order to calculate airborne nonrespirable fluxes, a simplified assumption was made. The assumption was that particle inertia of nonrespirable particles was sufficient to cause particles to move in a straight trajectory as particles entered cowl inlets. In this case, the airborne flux could then be calculated from the cowl inlet cross sectional area and sampling

*Laboratory for Electronics, Environmental Analysis Laboratories,
2030 Wright Avenue, Richmond, California 94804.

time. Since both plutonium and airborne soil collected in the cowl were measured, airborne fluxes of both plutonium and airborne soil were calculated for nonrespirable particles.

$^{238}\text{Pu}/^{239}\text{Pu}$ ratios were calculated for particles collected within cowls. This ratio was calculated for both the total material collected within cowls as well as a function of particle diameter increments for samples collected 0.3 m above ground level. These airborne ratios will be compared to $^{238}\text{Pu}/^{239}\text{Pu}$ ratios measured on the ground surface throughout the Rocky Flats area. Comparison of these ratios indicate plutonium concentrations on nonrespirable particles may be significantly different than measurements obtained from the top 5 cm deep ground surface. While airborne and ground surface ratios are similar near the eastern security fence, there are significant differences between airborne ratios and ground surface ratios near the eastern cattle fence. These differences are interpreted to indicate ^{238}Pu may resuspend differently than ^{239}Pu . In the following sections results will be discussed in detail.

TOTAL NONRESPIRABLE AIRBORNE PLUTONIUM FLUX

Airborne plutonium fluxes on nonrespirable particles were calculated similar to airborne fluxes of nonrespirable soil particles. The assumption was plutonium on nonrespirable particles approaching the cowl inlet continued in a straight trajectory and, subsequently, the plutonium was collected within each cowl.

In Figure 3, the airborne ^{239}Pu nonrespirable horizontal flux in $\mu\text{Ci}/\text{m}^2/\text{day}$ is shown as a function of sampling distance and sampling height. As might be expected, the maximum airborne ^{239}Pu flux on nonrespirable particles was at Site A near the original oil storage area. The maximum airborne ^{239}Pu flux was $6 \times 10^{-4} \mu\text{Ci}/\text{m}^2/\text{day}$. The airborne flux decreased with both distance and sampling height. At Site A, the ^{239}Pu flux decreased over 1 order of magnitude as sampling height was increased from 0.3 to 2 m above ground level. Similarly, at Site AB, the airborne ^{239}Pu nonrespirable flux again decreased about 1 order of magnitude as sampling height was increased

from 0.3 to 1 to 2 m above ground level. Airborne ^{239}Pu fluxes on nonrespirable particles decreased almost 2 orders of magnitude between sampling Sites A and AB. However, between sampling Sites AB and B, airborne ^{239}Pu nonrespirable fluxes did not show a significant variation with distance.

By comparing data for the three different sites, the conclusion is the airborne nonrespirable ^{239}Pu flux does not decay as a simple exponential function of distance from Site A. In addition, data for sampling heights above 1 m at Sites AB and B indicate the airborne ^{239}Pu flux does not significantly decrease for heights greater than 1 m up to 10 m. It is unknown how high above 10 m one would have to sample in order to show an additional significant plutonium decrease at higher elevations.

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FIGURE 3. Decrease with Distance of Total ^{239}Pu Flux on Nonrespirable Particles

Similar results are shown in Figure 4 for total ^{238}Pu flux on nonrespirable particles as a function of a sampling site and sampling height. The maximum airborne ^{238}Pu nonrespirable flux was $1.2 \times 10^{-5} \mu\text{Ci}/\text{m}^2/\text{day}$ and was at the 0.3 m sampling height at Site A. Again, at Site A as well as Site AB, airborne ^{238}Pu fluxes decreased rapidly as sampling height increased from 0.3 up to 10 m. However, from Site AB to B, an unexplained observation is made. The 2 and 10 m airborne ^{238}Pu fluxes at Site B were greater than at Site AB. An explanation for this increase is not apparent, but is believed to be real since the increase is supported by comparing analyses uncertainties in plutonium measurements. Error bars for Site B show a ^{238}Pu flux range significantly above error bars around Site AB.

Neg. No. PNL-767208-6

FIGURE 4. Decrease with Distance of Total ^{238}Pu Flux on Nonrespirable Particles

NONRESPIRABLE AIRBORNE Pu FLUX AS A FUNCTION OF SOIL PARTICLE DIAMETER

Airborne ^{239}Pu horizontal fluxes on nonrespirable particles at Sites A and AB are shown in Figure 5. In this case, particle diameter ranges include additional ranges from 10 to 20 μm and below 10 μm . At Site A there appears to be an inverse relationship between plutonium flux and an increase in particle diameter for particle diameters between 20 and 210 μm . However, this apparent relationship between ^{239}Pu flux and particle diameter appears to be much different for particle diameters smaller than 20 μm and greater than 210 μm .

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FIGURE 5. Decrease in Airborne ^{239}Pu Flux on Nonrespirable Particles from Site A to Site AB

At Site AB the relationship between airborne ^{239}Pu horizontal flux and particle diameter appears to be more consistent. ^{239}Pu fluxes appear to be nearly inversely proportional to an increase in particle diameter for all particle diameters above about $10\ \mu\text{m}$. It is only for particles less than $10\ \mu\text{m}$ that the relationship is not inversely proportional to particle diameter. At the present time, one can only speculate as to whether the relationship should be linear or curved for all data points.

Similar data for airborne ^{238}Pu fluxes in $\mu\text{Ci}/\text{m}^2/\text{day}$ are shown in Figure 6 for Sites A and AB at 0.3 m. Similar to Figure 5 for ^{239}Pu airborne fluxes, ^{238}Pu data at Site A indicate an inverse relationship may be approximate for particle diameters between 20 and $210\ \mu\text{m}$. The linear relationship does not appear to hold for particle diameters greater than $210\ \mu\text{m}$.

The ^{238}Pu flux data at Site AB appear to be significantly different than airborne plutonium fluxes for other data in Figures 5 and 6. In this case, the airborne ^{238}Pu flux at Site AB appears to be nearly independent of particle size for all particle diameters except for one anomaly. This anomaly for particles between 105 to $125\ \mu\text{m}$ is significantly less than the remaining data. It is unknown why this anomaly occurs.

In Figures 5 and 6, airborne ^{239}Pu and ^{238}Pu fluxes were shown to decrease from Site A to Site AB. This decrease is shown in Figure 7 as Site AB/Site A ratios of airborne fluxes. There is a difference between these ratios for ^{239}Pu and ^{238}Pu . For ^{238}Pu , the Site AB/Site A ratio ranges from about 10^{-2} to 3×10^{-1} . In contrast, the airborne ^{239}Pu Site AB/Site A ratio is less and ranges from about 4×10^{-4} to 4×10^{-2} .

If both ^{239}Pu and ^{238}Pu were uniformly distributed on the ground surface soil and were resuspended similarly, one would expect Site AB/Site A ratios for ^{239}Pu and ^{238}Pu would be very similar and overlapping. Obviously, this is not the case. Thus, one must conclude that ^{238}Pu is resuspended more readily than ^{239}Pu . Increased ^{238}Pu resuspendability may be conjectured to be caused by local soil differences in chemical action or by biodegradation of ^{238}Pu preferentially to ^{239}Pu at distances away from Site A. Possibly at Site A, ground surface Pu concentrations were sufficiently high to have minimized any biodegradation. Additional data are needed to clarify ^{238}Pu enrichment processes.

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FIGURE 6. Decrease in Airborne ^{238}Pu Flux on Nonrespirable Particles from Site A to Site AB

Neg. No. PNL-767208-10

FIGURE 7. Nonrespirable Airborne ^{238}Pu and ^{239}Pu Flux Ratios of Site AB/Site A as a Function of Particle Diameter

Pu CONCENTRATION ON NONRESPIRABLE AIRBORNE SOIL

Airborne plutonium concentrations were normalized to the soil collected with the airborne plutonium. Calculated results for ^{239}Pu are shown in Figure 8. Plutonium concentrations in $\mu\text{Ci/g}$ are shown as a function of particle diameter as determined with both particle cascade impactors for respirable diameters as well as the sieve sizes for nonrespirable particles. Data for respirable particles in the aerodynamic size range were previously reported.⁽¹⁾ ^{239}Pu was attached to all particle diameter ranges at both Site A and Site AB. However, concentrations at Site A were significantly greater than at Site AB. This increase is expected since Site A was nearer the original oil storage area.

At each site, Pu concentrations in $\mu\text{Ci/g}$ indicate general continuous relationships as a function of particle diameter. For nonrespirable particle diameter ranges determined from sieve sizes, the data could be approximated by a straight line inversely proportional to particle diameter. This relationship, however, is not really clear cut. The relationship is complicated by collection of both contaminated onsite and uncontaminated offsite nonrespirable particles within the cowl.

For respirable particles, the ^{239}Pu $\mu\text{Ci/g}$ was nearly independent of particle diameter. This independency might suggest plutonium attachments are volume phenomena for these respirable particles. In contrast, Pu attachment is expected to be controlled by available particle surface area for nonrespirable particles. However, additional data will be required to definitely conclude how plutonium particles are attached to airborne particles in both the respirable and nonrespirable size ranges.

^{238}Pu concentrations in $\mu\text{Ci/g}$ on airborne soil are shown in Figure 9. In this case, only nonrespirable particle diameter ranges are shown. There was insufficient ^{238}Pu collected in the particle cascade impactor samples to yield positive results in respirable diameter ranges. Similar to ^{239}Pu , ^{238}Pu nonrespirable concentrations were greater at Site A than at Site B and also showed an inverse relationship with particle diameter. However, there is fine structure showing deviation around any apparent inverse

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FIGURE 8. ^{239}Pu Concentration on Airborne Soil as a Function of Particle Diameter

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FIGURE 9. ^{238}Pu Concentration on Airborne Soil as a Function of Particle Diameter

relationship. This fine structure indicates there is yet much to be learned about plutonium resuspension and plutonium attachment to nonrespirable as well as respirable host particles.

Both ^{239}Pu and ^{238}Pu concentrations on airborne soil decreased from Site A to Site AB. Site AB/Site A ratios of $\mu\text{Ci/g}$ are shown in Figure 10. Concentrations per gram decreased by a factor of up to about 10^{-3} in the intervening 392 m distance separating sampling Sites A and AB. Concentrations on respirable particles showed ^{239}Pu decreased about 10^{-1} and was independent of particle size. In contrast for nonrespirable particle sizes, ^{239}Pu ratios between sites decreased nearly linearly as particle diameter was increased. ^{239}Pu concentrations on nonrespirable particles decreased at rates greater than the ^{238}Pu decrease. For ^{238}Pu , the $\mu\text{Ci/g}$ ratios for Site AB/Site A were nearly 1 order of magnitude greater than ^{239}Pu . These larger ratios suggest ^{238}Pu resuspended more readily relative to ^{239}Pu .

AIRBORNE AND GROUND Pu $\mu\text{Ci/g}$

There should be some relationship between Pu concentrations on airborne soil and Pu concentrations on ground surface soil. Ground surface soils were characterized⁽⁴⁾ for the same time period these airborne nonrespirable samples were collected. Ground surface sample results are summarized in Table 1 for sampling sites near Sites A, AB, and B in both dpm/g and $\mu\text{Ci/g}$. Similarly, ground surface $^{238}\text{Pu}/^{239}\text{Pu}$ ratios are also shown.

Ground and airborne Pu soil sample results are compared in Table 2 for both ^{239}Pu and ^{238}Pu . Comparisons are for plutonium in ground surface samples 5 cm deep versus airborne nonrespirable particle concentrations in particle diameter ranges. Airborne concentrations were taken from data limits in Figures 8 and 9. Maximum ranges of $\mu\text{Ci/g}$ airborne compared to $\mu\text{Ci/g}$ in the ground surface soil are shown in the last two columns. These ratios range from 1×10^{-4} up to 2. Thus, in hazard evaluations⁽²⁾ one might consider maximum Pu concentrations on airborne soil to be comparable to Pu concentrations on ground surface soils. This is indicated by the ratio 2. However, in most cases Pu concentrations on airborne soil should be significantly less than Pu concentrations on ground surface soils.

Neg. No. PNL-767208-7

FIGURE 10. Decrease in ^{238}Pu and ^{239}Pu Concentrations on Nonrespirable Particles from Site A to Site AB

TABLE 1. Selected Surface Soil Sample Results*

Locations Near Site	HASL-304 Map Sites	^{239}Pu		^{238}Pu		$^{238}\text{Pu}/^{239}\text{Pu}$
		dpm/g	$\mu\text{Ci/g}$	dpm/g	$\mu\text{Ci/g}$	
A	25	6890 $\pm$ 5	3.10×10^{-3}	128 $\pm$ 5	5.77×10^{-5}	0.019
	26	1530 $\pm$ 4	6.89×10^{-4}	27.5 $\pm$ 5	1.24×10^{-5}	0.018
AB	2	171 $\pm$ 6	7.70×10^{-5}	3.47 $\pm$ 7	1.56×10^{-6}	0.020
	4	85.8 $\pm$ 5	3.86×10^{-5}	1.70 $\pm$ 7	7.66×10^{-7}	0.020
B	6	5.91 $\pm$ 3	2.66×10^{-6}	0.113 $\pm$ 7	5.09×10^{-8}	0.019
	7	85.5 $\pm$ 5	3.85×10^{-5}	1.58 $\pm$ 7	7.12×10^{-7}	0.018

* dpm/g dry soil $\pm$ Stnd. Dev., samples from 2000 cm^2 , 5 cm deep

**TABLE 2. Ratio of Pu Concentration Per Gram of Soil:
Airborne Soil/Ground Surface Soil**

Site	Range, $\mu\text{Ci/g}$				Ratio Range, $\frac{\mu\text{Ci/g airborne}}{\mu\text{Ci/g surface}}$	
	^{239}Pu		^{238}Pu		^{239}Pu	^{238}Pu
	Airborne*	Surface**	Airborne***	Surface**		
A	1.3×10^{-4} to 4.6×10^{-5}	3.10×10^{-3} to 6.89×10^{-4}	7.2×10^{-6} to 3.4×10^{-7}	5.77×10^{-5} to 1.24×10^{-5}	0.04 to 0.2	0.006 to 0.6
AB	3.2×10^{-6} to 8.5×10^{-9}	7.70×10^{-5} to 3.85×10^{-5}	1.8×10^{-6} to 1.4×10^{-9}	1.57×10^{-6} to 7.66×10^{-7}	0.0001 to 0.08	0.0009 to 2

* From Figure 8 as a function of particle diameter

** From Table 1 for total ground surface sample 5 cm deep

*** From Figure 9 as a function of particle diameter

AIRBORNE $^{238}\text{Pu}/^{239}\text{Pu}$ RATIOS

Airborne $^{238}\text{Pu}/^{239}\text{Pu}$ ratios consistently changed from sampling Site A to Site B. Ratios shown in Figure 11 are for total plutonium collected within each cowl at each sampling site and sampling height. At Site A, the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio on airborne nonrespirable soil is comparable to ratios determined from 5 cm deep surface soil samples. The surface soil range is shown as the crosshatched soil sample range. This range⁽⁴⁾ represents surface soil samples taken between the eastern security fence and beyond the eastern cattle fence to Indiana Avenue.

At Sites AB and B, airborne $^{239}\text{Pu}/^{238}\text{Pu}$ ratios on nonrespirable particles are over 1 order of magnitude greater than similar ratios from 5 cm deep surface soil samples. These ratios do not appear to be significantly affected by sampling height between 0.3 to 10 m above ground level. However, data counting statistic error bars at Site A and AB tend to indicate a higher $^{238}\text{Pu}/^{239}\text{Pu}$ ratio closer to the ground.

The $^{238}\text{Pu}/^{239}\text{Pu}$ ratio was determined for all nonrespirable particle diameter ranges at Sites A and AB at the 0.3 m sampling height. These results are shown in Figure 12 along with the total ground surface soil sample range. At Site A, the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio is nearly independent of

particle diameter for all particle diameters above 20 μm . It is only for particles less than 20 μm that the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio is significantly elevated at Site A. The $^{238}\text{Pu}/^{239}\text{Pu}$ ratios on nonrespirable particles greater than 20 μm at Site A are all comparable to ratios in the 5 cm deep surface soil samples. This similarity would be expected if there were no preferential ^{238}Pu to ^{239}Pu separation on soil surface. In contrast to Site A, $^{238}\text{Pu}/^{239}\text{Pu}$ ratios for Site AB are much different and are significantly elevated for all particle diameters above the surface soil sample range.

Neg. No. PNL-767208-8

FIGURE 11. $^{238}\text{Pu}/^{239}\text{Pu}$ Ratio on Nonrespirable Airborne Particles as a Function of Distance

Neg. No. 767208-11

FIGURE 12. $^{238}\text{Pu}/^{239}\text{Pu}$ Ratio on Nonrespirable Airborne Soil as a Function of Particle Diameter

One can only hypothesize as to why there should be a difference in $^{238}\text{Pu}/^{239}\text{Pu}$ ratios at Sites AB and A as compared to soil surface samples. One difference is the surface activity level at Site A is greater than at Site AB. As was shown in Table 1, the ground soil surface activity level at Site A was approximately 10 times greater than at Site AB. If plutonium particles were attacked by microorganisms in the soil, microorganism activity might be decreased by the increased activity level at Site A. If microorganisms preferentially attacked ^{238}Pu at Site AB with a lower Pu contamination, ^{238}Pu on surface soils might become more readily available for resuspension.

Another possibility for increased resuspendability of surface ^{238}Pu at Site AB might be differences in soil chemistry between Sites A and AB. Both possibilities exist of either microorganism activity or differences in soil chemistry, but it is unknown if either is the reason causing elevated $^{238}\text{Pu}/^{239}\text{Pu}$ ratios at Site AB. Additional research is indicated to determine causes of elevated $^{238}\text{Pu}/^{239}\text{Pu}$ ratios at Site AB.

NONRESPIRABLE AIRBORNE SOIL FLUX

Airborne nonrespirable horizontal soil fluxes in $\text{g}/\text{m}^2/\text{day}$ were calculated. These airborne soil fluxes were determined for both the total soil flux and soil flux as a function of particle diameter for samples analyzed for Pu.

Airborne nonrespirable soil fluxes as a function of distance from Site A are shown in Figure 13. Fluxes are shown for Site A near the eastern security fence, Site B near the eastern cattle fence, and Site A between these two sites. Fluxes are shown both as a function of distance as well as sampling height. As might be expected, the largest average airborne flux was near the ground surface. In this case, the maximum flux was $14 \text{ g}/\text{m}^2/\text{day}$. This maximum flux was nearly constant at both Sites A and AB. The airborne soil flux decreased with sampling height. The lower lines show the soil flux decreased almost two orders of magnitude between sampling heights of 0.3 m up to a sampling height of 2 m. The soil flux between 2 and 10 m did not show a consistent change with height. As shown by the 10 m height line, the flux at 10 m was somewhat greater than the flux at 2 m.

Airborne nonrespirable soil fluxes as a function of particle diameter at 0.3 m above ground level are shown in Figure 14. The two data symbols in the upper right-hand corner indicate the flux for particle diameters greater than $210 \mu\text{m}$. This was the largest sieve size used for screening. Similarly in the lower left portion of the figure, arrows indicate the flux for particles smaller than $20 \mu\text{m}$ in diameter. The data between 20 and $210 \mu\text{m}$ are shown by horizontal lines. Line lengths correspond to diameter increments between sieving screen sizes.

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FIGURE 13. Airborne Nonrespirable Soil Flux as a Function of Distance

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FIGURE 14. Airborne Nonrespirable Soil Flux as a Function of Particle Diameter at 0.3 m Height

The airborne nonrespirable soil flux as a function of particle diameter ranged from 10^{-1} g/m²/day for 20 μm particles up to almost 10 g/m²/day for 210 μm particles. Comparisons of the solid lines for Site A and the dotted lines for Site AB indicate reasonable agreement between airborne soil fluxes between sampling sites. In general, the flux increased as particle size increased. The only exceptions are for particle diameters between 20 and 37 μm and between 37 and 53 μm. For these two diameter ranges, airborne nonrespirable soil fluxes are significantly greater than would be expected by a simple smooth curve through the other data points. At the present time,

it is unknown whether this variation in these two diameter ranges might be generalized for other conditions and other resuspension sites. Additional data will be required to make generalizations for the flux as a function of particle diameter range.

ESTIMATION OF RELATIVE RESPIRABLE AND NONRESPIRABLE Pu FLUXES

A direct comparison between airborne fluxes on respirable and nonrespirable particles was not measured since respirable and nonrespirable samples were sampled differently. Respirable particles were sampled at a constant flow rate of $0.57 \text{ m}^3/\text{min}$ while nonrespirable particles were collected by inertial collection within cowls. In order to calculate the relative flux on respirable and nonrespirable particles, one needs to know (1) the average wind speed to determine the average flux on respirable particles, and (2) how particles were collected within the cowl by inertial impaction. In this case inertial impaction means particles would enter the cowl as if the cowl sampling were isokinetic. In actuality, there is flow divergence around the cowl inlet. Consequently correction factors are needed for calculating the true airborne particle fluxes for nonisokinetic sampling. Nonisokinetic correction factors are not available. Even with these qualifications, one might still be interested in an approximation of the relative Pu fluxes between respirable and nonrespirable particles. Consequently, a simple calculation will be made to illustrate the relative orders of magnitude for respirable and nonrespirable Pu fluxes.

The horizontal plutonium flux can be estimated from airborne soil fluxes and plutonium concentrations on airborne soil. Since soil fluxes have already been discussed, plutonium concentrations on airborne soil will be discussed prior to discussing plutonium fluxes. Plutonium concentrations in pCi/g of airborne respirable and nonrespirable soil are shown in Table 3. This table summarizes both total plutonium concentrations per gram of soil collected within cowls, and also respirable concentrations per gram for cases in which air was sampled continuously with particle cascade impactors. Respirable ^{239}Pu concentrations ranged from 2 to 62 pCi/g airborne soil. Concentrations of ^{238}Pu on respirable soil were less than radiochemical analytical limits.

TABLE 3. Pu pCi/g on Airborne Soil Collected During Continuous Air Sampling

Sampling Site	Sampling height, m	pCi/g					
		"Respirable*"		Nonrespirable		Total Assuming Isokinetic Sampling	
		^{239}Pu	^{238}Pu	^{239}Pu	^{238}Pu	^{239}Pu	^{238}Pu
A	0.3	62.3	LAL	203	4.2	188	3.8
	2	4.6	LAL	313	5.1	98.3	1
AB	0.3	5.0	LAL	1	0.2	1.6	0.2
	1	*	*	11.5	1.6		
	2	*	*	12.4	1.6		
B	2	2.1	LAL	10.2	2.4	3.1	0.2
	10	38.2	LAL	10.0	4.2	36.4	0.2

"Respirable" is all material collected with a particle cascade impactor

LAL, less than radiochemical analytical limit

* Respirable samples were collected for wind speed increments rather than continuous sampling

1 dpm/g = 0.45 pCi/g

On nonrespirable soil, ^{239}Pu concentrations ranged from 1 up to 313 pCi/g and ^{238}Pu concentrations ranged from 0.2 up to 5 pCi/g. In order to estimate the average plutonium concentration on airborne soil, respirable and nonrespirable concentrations were combined in the last two columns. For this calculation, isokinetic sampling was assumed. This assumption will be discussed further in a later section. Total Pu concentrations on airborne soil ranged from 1 up to 188 pCi/g for ^{239}Pu , and from 0.2 up to 3 pCi/g for ^{238}Pu . These concentrations can be easily converted to dpm/g by multiplying indicated values by 0.45.

Average airborne plutonium fluxes entering cowl inlets were calculated from airborne soil fluxes and plutonium concentrations on airborne soil. As shown in Table 4, plutonium fluxes on both respirable and nonrespirable particles were calculated in units of $\text{pCi/m}^2/\text{day}$. The calculation was based on collected plutonium, the sampling time, and the cross sectional area of the

TABLE 4. Average Pu Flux Entering Cowl-Impactor System During Continuous Air Sampling

Sampling Site	Sampling height, m	pCi/m ² /day			
		"Respirable"		Nonrespirable	
		²³⁹ Pu	²³⁸ Pu	²³⁹ Pu	²³⁸ Pu
A	0.3	0.406	LAL	10.2	0.211
	2	0.243	LAL	0.403	6.68x10 ⁻³
AB	0.3	3.22x10 ⁻²	LAL	5.66x10	1.13x10 ⁻²
B	2	1.16x10 ⁻²	LAL	8.23x10 ⁻³	1.85x10 ⁻³
	10	0.193	LAL	3.18x10 ⁻³	1.32x10 ⁻³

"Respirable" is all material collected with a particle cascade impactor LAL, less than radiochemical analytical limit

1 pCi = 2.22 dpm

cowl inlet. The cowl inlet diameter was 15.2 cm. The maximum ²³⁹Pu flux was 10 pCi/m²/day and the minimum flux was 3 x 10⁻³ pCi/m²/day. Based on these limited data, these calculated fluxes might be used for estimating total plutonium fluxes over larger integrated areas. However, such estimates should be made with caution since the flux variability at other sites and elevations are unknown.

Average plutonium fluxes entering the cowl impactor system can be used to estimate the respirable fraction of airborne plutonium. Estimates are given in Table 5 which are based on the isokinetic sampling assumption. Respirable fractions ranged from 3 to 98% of total airborne plutonium. However, one should use these numbers with caution. Plutonium collected within particle cascade impactors contained particles of 7, 3.3, 2.0, and 1.1 μm diameter (which are impactor stage 50% cutoff diameters) as well as smaller particles collected on the impactor backup filter. From the inhalation standpoint, particles collected on the 7 μm state should not be included within the respirable size range. Only the smaller particles are usually

TABLE 5. Estimated Respirable Percent of
Total Airborne ^{239}Pu Flux

Sampling Site	Sampling Height, m	Airborne ^{239}Pu Respirable Percent	
		Assumed Isokinetic	Assumed Correction
A	0.3	3.8	9
	2	37.7	60
AB	0.3	36.3	59
B	2	58.5	78
	10	98.4	99

Where Res = dpm/min collected in impactor

Non = dpm/min collected in cowl

U_s = average flow rate through cowl inlet, 0.36 m/sec

U_a = average windspeed, 0.9 m/sec

Isokinetic = $\frac{\text{Res}}{(\text{Res}) (U_a/U_s)}$

Correction = $\frac{(\text{Res}) (U_a/U_s)}{(\text{Res}) (U_a/U_s) + \text{Non}}$

considered respirable. However, in the present comparison between cowl collected particles and the impactor collected particles there is much uncertainty in calculating the relative $7\ \mu\text{m}$ concentration as compared to the cowl collection particles.

A better estimate of the respirable Pu fraction is shown in the last column of Table 5. The equation used for the assumed correction is shown as the last line of the table notes. Based on the assumed correction factor, the respirable fraction of airborne plutonium increased from 3 to 98% for an isokinetic sampling assumption up to 9 to 99% of the total airborne plutonium. In actuality, true respirable fractions should be between these limiting values.

Respirable fractions have been previously reported⁽⁶⁾ as 25% for plutonium collected within particle cascade impactors. Since the present results indicate plutonium is also attached to the much larger particle size ranges

collected in cows, published respirable fractions are probably indicative of maximum limits rather than a true respirable limit.

Even if all Pu collected in the cowl-impactor sampling systems were respirable, airborne Pu concentrations were still below maximum permissible⁽⁷⁾ concentrations (MPC). The MPC_{168Hr-air} for ^{239}Pu is 600 fCi/m^3 (femto = 10^{-15}). The smallest respirable fraction in Table 5 is 3.8%. This sample at 0.3 m for Site A also represents the largest airborne Pu concentration. For this sample, the real respirable ^{239}Pu concentration ($\leq 3.3 \mu\text{m}$ diameter) was 0.25 fCi/m^3 while the concentration was 0.50 fCi/m^3 if the $7 \mu\text{m}$ particles of the particle cascade impactor are included. If all airborne Pu is assumed respirable, the maximum ^{239}Pu concentration is $(0.50/0.038) 13 \text{ fCi/m}^3$, which is still significantly less than the MPC of 600 fCi/m^3 .

CONCLUSIONS

In July 1973 at Rocky Flats plutonium was transported on both respirable and nonrespirable airborne soil particles. Nonrespirable airborne particles are normally not sampled due to sampling bias in most air sampling systems. Consequently, total airborne plutonium fluxes could be greater than normally reported with existing sampling systems. However, those systems are a conservative estimate (high concentration) of the airborne respirable plutonium concentrations. Nevertheless, plutonium transport on nonrespirable particles may be a significant factor in plutonium transport near Rocky Flats. Larger than respirable particles probably are resuspended and may not travel too far downwind before redepositing again. In contrast, respirable particles remain airborne for a much longer distance. Additional research is needed to clarify the relative significance of plutonium transport on respirable as compared to nonrespirable particles.

Plutonium concentrations on collected airborne soil ranged from a maximum of twice the concentration on ground surface soil to as low as 10^{-4} of the concentration on surface soils. The maximum reported⁽⁴⁾ surface soil concentration was 3×10^3 pCi/g, which corresponds to 6890 dpm/g. Even at this relatively high plutonium surface soil concentration, airborne respirable Pu concentrations were significantly below maximum permissible airborne concentrations (MPC) on a yearly basis. Maximum permissible concentrations are based on dose calculations over a lifetime. One should be able to breathe concentrations significantly above MPC for even extended shortened time periods without undue bodily stress. Consequently, measured airborne respirable plutonium concentrations at Rocky Flats are considered to be no health hazard based on existing biological knowledge.

Relative $^{238}\text{Pu}/^{239}\text{Pu}$ ratio data on nonrespirable particles indicate ^{238}Pu can be resuspended significantly differently than ^{239}Pu . This conclusion was obtained by comparing airborne $^{238}\text{Pu}/^{239}\text{Pu}$ ratios on nonrespirable soil near the eastern security fence and the eastern cattle fence. At the eastern security fence, the airborne $^{238}\text{Pu}/^{239}\text{Pu}$ ratios were similar to ground surface ratios. However, at the eastern cattle fence, the $^{238}\text{Pu}/^{239}\text{Pu}$

ratios were significantly greater than measured on local surface soils (see Figure 11). Consequently, an explanation is needed to explain the increased $^{238}\text{Pu}/^{239}\text{Pu}$ ratio at distances from the original oil storage area. Possible explanations include both biodegradation of ^{238}Pu preferentially to ^{239}Pu as well as differences in the soil chemistry between the eastern security and the eastern cattle fence. These possibilities exist, but are not definitive. Further research is indicated in order to explain the higher relative air-borne $^{238}\text{Pu}/^{239}\text{Pu}$ concentrations near the eastern cattle fence.

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