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Plutonium Concentrations in Airborne Soil at Rocky Flats and Hanford Determined During Resuspension Experiments

by
George A. Sehmel

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PLUTONIUM CONCENTRATIONS IN AIRBORNE SOIL AT ROCKY FLATS AND HANFORD DETERMINED DURING RESUSPENSION EXPERIMENTS

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ABSTRACT

Plutonium resuspension results are summarized for experiments[1] conducted by the author at Rocky Flats, onsite on the Hanford reservation, and for winds blowing from offsite onto the Hanford reservation near the Prosser barricade boundary. In each case, plutonium resuspension was shown by increased airborne plutonium concentrations as a function of either wind speed or as compared to fallout levels. All measured airborne concentrations were far below maximum permissible concentrations (MPC).

Both plutonium and cesium concentrations on airborne soil were normalized by the quantity of airborne soil sampled. Airborne radionuclide concentrations in μCi/g were related to published values for radionuclide concentrations on surface soils. For this ratio of radionuclide concentration per gram on airborne soil divided by that for ground surface soil, there are eight orders of magnitude uncertainty from 10^{-4} to 10^{4}. This uncertainty in the equality between plutonium concentrations per gram on airborne and surface soils is caused by only a fraction of the collected airborne soil being transported from offsite rather than all being resuspended from each study site and also by the great variabilities in surface contamination.

Horizontal plutonium fluxes on airborne nonrespirable soils at all three sites were bracketed within the same four orders of magnitude from 10^{-7} to 10^{-3} μCi/(m^2 day) for ^{239}Pu and 10^{-8} to 10^{-5} μCi/(m^2 day) for ^{238}Pu.

Airborne respirable ^{239}Pu concentrations increased with wind speed for a southwest wind direction coming from offsite near the Hanford reservation Prosser barricade. Airborne plutonium fluxes on nonrespirable particles had isotopic ratios, $^{240}Pu/^{239}$ ^{240}Pu, similar to weapons grade plutonium rather than fallout plutonium.

INTRODUCTION

Resuspension occurs when particles on a surface are disturbed and carried up into the air by air currents. Wind-caused resuspension is the process by which wind blows particles from a surface into the air and transports them downwind. For radionuclide contaminated surfaces, wind might cause radionuclide particles to be resuspended and transported to other sites. Resuspension occurs at radionuclide contaminated sites on the Hanford reservation in Washington,[1,2] at Rocky Flats in Colorado,[3-8] at the Nevada test site,[9,10] at the Savannah River Laboratory reservation in Tennessee,[11] and at other sites,[12,13] but with our present knowledge,[14,15] amounts of wind-caused resuspension and its effects cannot be adequately predicted.

The need for such predictions is not new: for many years resuspension has been occurring at nuclear sites. Some of the earliest data were obtained[9,10] at the Nevada test site. Ground radioactivity contours were determined as a function of time after a test detonation. These contours show deposited radionuclide particles were resuspended by prevailing winds, and ground surface concentrations decreased with increasing distance from the test center during the initial time period after the test.[9] Subsequent ground radioactivity contours showed a migration of radionuclides from the test site, indicating that resuspension had occurred. Similarly, aerial surveys at Hanford[16] showed ^{241}Am transport by wind resuspension.

Resuspension is of considerable interest at the Rocky Flats nuclear plant in Colorado where ground surfaces were contaminated with plutonium from leaking storage barrels containing plutonium-contaminated cutting oil.[3-8] After leakage was discovered, barrels were removed and corrective actions were taken, but plutonium resuspension from residually contaminated soil surfaces is still occurring.

More recently, resuspension has been reported at study sites on the Hanford reservation.[1,2] These sites were low-level liquid waste disposal sites.

The objective of this paper is to summarize measured resuspension concentrations and to consider the implications of these results. In these experiments, airborne concentrations were measured[1] as functions of wind speed, airborne particle size, wind direction, and the collected plutonium and other radionuclides per gram of airborne soil. Airborne radionuclides were normalized by the total amount of airborne solids to relate concentration per gram of airborne solid to concentration per gram of radionuclide on the ground.

STUDY SITES

The Hanford Reservation and Rocky Flats were unique sites at which to measure resuspension and determine resuspension physics. At Hanford,[2] nuclear reactors and separation plants have been operated for over 30 years. Fuel elements have been processed and high- and low-level liquid wastes from those separation plant process streams have been stored and accumulated. These include low-level wastes discharged to ponds and trenches to be filtered through soil and sediments. As a result, residual radioactive materials in liquid waste are retained on soil particles or trench bottoms. Several low-level waste disposal areas were resuspension source study sites.[2] In contrast, at Rocky Flats,[3-8] the ground source of plutonium was an area where oil had originally contaminated a relatively small area of soil. This primary contaminated soil was covered with asphalt. Low-level ground contamination[4] was from plutonium resuspended and deposited prior to asphalt covering of the primary source.

EXPERIMENTS

The experiments for measuring particle resuspension reported here have been reported in greater detail in the following references:

- plutonium and americium from resuspension study sites at Hanford[2] (see Figure 1)
- plutonium from contaminated environmental surfaces at Rocky Flats[6,7] (see Figure 2)
- plutonium resuspension results from off-site Hanford near the Prosser barricade at Hanford.[1] (see Figure 1).

PARTICLES

Resuspension was measured for several particle sizes. The plutonium particle size distributions on soils at Rocky Flats and Hanford were those present from the contaminating event and subsequent transformations with time.

AIR SAMPLERS

Airborne resuspended particles were either sampled with total air samplers* or sized while airborne with particle cascade impactors.**

The particle cascade impactor for sampling respirable particles was attached to a rotating cowl, allowing simultaneous sampling of larger nonrespirable particles. The cowl-impactor system[17] is shown in Figure 3. Particles entering the 15-cm-diameter cylindrical sampler inlet of the cowl either settled on the cowl floor or were drawn up into the impactor. Particles settling on the cowl floor are called "nonrespirable" in this report. Respirable particles entering the particle cascade impactor were separated into nominal aerodynamic diameter ranges of 7, 3.3, 2.0 and 1.1 μm, which are impactor stage 50% cutoff diameters for unit-density spheres. Smaller particles were collected on an impactor backup filter.

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

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

RESULTS AND DISCUSSION

Airborne radionuclide concentrations were determined at transuranic resuspension study sites at Rocky Flats, Colorado[6,7] and the Hanford reservation in Washington.[1,2] In addition some cesium resuspension data are reported for Hanford.[2] Both ^{238}Pu and ^{239}Pu resuspension occur onsite at Rocky Flats[6-8] and the Hanford reservation.[2] In addition, plutonium was resuspended at very low concentrations from offsite near the Hanford reservation. All airborne ^{239}Pu concentrations were significantly less than maximum permissible concentrations, MPC, of soluble ^{239}Pu in air for either occupational exposure in a 40-hr work week (2×10^{-12} μCi/cm^3) or nonoccupational exposure in a 168-hr week period (6×10^{-13} μCi/cm^3).[18] Similarly, all airborne ^{238}Pu concentrations were significantly less than the MPC for either occupational exposure in a 40-hr work week (2×10^{-12} μCi/cm^3) or nonoccupational exposure in a 168-hr week (7×10^{-13} μCi/cm^3). In all cases plutonium was deposited on all stages of particle cascade impactors, showing that most plutonium was resuspended while attached to larger host soil particles. Results for each set of experiments will be discussed separately.

PLUTONIUM PARTICLE RESUSPENSION "ON-SITE"

Airborne plutonium concentrations at Rocky Flats and Hanford were measured as a function of particle diameter, wind speed, and sampling site. Radionuclide concentrations per gram of airborne solid were determined.

Plutonium Resuspension Research at Rocky Flats

Experimental measurements of plutonium resuspension at Rocky Flats were made in July 1973.[6,7] As shown in Figure 2, airborne plutonium concentrations were measured at three sampling sites east of the plant. The first sampling site was along the eastern security fence. This site was called sampling site A. Sampling site B was along the eastern cattle fence, and sampling site AB was between sites A and B. The distance from site A to site AB was 227 m. Airborne plutonium at these sites was sampled and analyzed as a function of sampling height, particle size, and wind speed.

Airborne particles were separated in the sampling process into two main fractions. One sample contained particles collected by gravity settling in the inlet cowl section of the sampler shown in Figure 3. The second fraction contained those particles passing through the inlet section and collected within the high volume cascade impactor. The smallest particles collected in the inlet cowl section were about 10 μm diameter. This fraction was assayed for ^{238}Pu and ^{239}Pu. In some cases, nonrespirable particles were sieved into smaller size fractions and these fractions also assayed for ^{238}Pu and ^{239}Pu. Data for respirable and nonrespirable particles will be discussed separately.

Respirable Plutonium Concentrations at Rocky Flats

Airborne ^{239}Pu concentrations at the three Rocky Flats sampling stations were reported in μCi/cm^3 of air and μCi/g of airborne soil. The maximum airborne ^{239}Pu concentration was 3.7×10^{-15} μCi/cm^3. The maximum ^{239}Pu concentration on the airborne soil was 5×10^{-5} μCi/g total airborne soil and 7×10^{-3} μCi/g for the respirable fraction of airborne soil collected on the 2-μm particle impactor stage.

The general trend of the complete airborne ^{239}Pu concentration data is a decrease in concentration with increasing distance eastward from site A.[6] As might be expected, this decrease in concentration corresponded to increasing distance from the original oil storage area, which was the principal source of ground contamination. However, significant deviations did occur in concentration profiles of airborne ^{239}Pu with both distance and height. These deviations might be attributed to sampling some more-active-than-normal particles or clusters of particles. These increases in average airborne ^{239}Pu concentrations were present at both sites AB and B.

As indicated in Figure 4 for site AB, some more-active-than-normal particles or clusters of particles ("hot") particles may have been present in the 2.0-μm size range. In this case, the concentration at the 1-m height of site AB is 1 to 2 orders of magnitude greater than at other heights for this site. More important to the "hot" particle concept is the concentration at the 10-m height of B. This concentration of 2.3×10^{-16} μCi/cm^3 was the largest ^{239}Pu concentration measured for 2-μm particles. This relatively high concentration was unexpected since this sampling location was the most remote from both the ground and the original oil storage area. This suggests other relatively "hot" particles could also be escaping from the plant boundaries; however, due caution is indicated in

interpreting this "hot" particle concept. The total of 6 dpm collected on the 2-μm stage or 2.3 x 10^{-16} μCi/cm^3 is much less than the maximum permissible air concentration of 2 x 10^{-12} μCi/cm^3 (occupational). It is conceivable that the majority of this plutonium was attached to one soil particle.

In Figure 5, total airborne concentrations are shown for air sampled at all wind speeds (average wind speed of 0.9 m/sec), at wind speeds from 4.1 to 6.3 m/sec, and at wind speeds from 6.3 to 9.8 m/sec. Airborne ^{239}Pu concentrations at wind speeds from 4.1 to 6.3 m/sec are definitely larger than average airborne concentrations for continuous air sampling. However, the 2σ radiochemical counting statistics error limits are too large to determine the wind speed dependency. Nevertheless, an attempt to approximate airborne ^{239}Pu concentrations and consequently the resuspension rate dependency upon wind speed was made for the 7-μm-diameter particles. This approximation was for the 0.3-m height at sampling site AB. For the three data points taken at the 0.3-m height, ^{239}Pu concentrations increased with the 5.9th power of wind speed.

Respirable Plutonium Concentrations at Hanford

Extensive data were obtained on airborne radionuclide concentrations around resuspension sites studied.[2] These concentrations were expressed both in μCi/cm^3 of filtered air and μCi/g of airborne solids. This report summarized ranges of data collected, but did not detail data for each experiment.

Airborne plutonium concentrations for both ^{238}Pu and ^{239}Pu measured[1] in resuspension experiments are shown in Figure 6 and are compared with Hanford Area fallout levels[19] approximately 30 Km distant. The data represented experiments conducted over various time periods. For each data symbol, the vertical line is plotted at the mid-time of the resuspension experiment while the experiment duration is shown by horizontal lines drawn at both maximum and minimum measured airborne concentrations. Airborne peak plutonium concentrations at resuspension study sites were significantly greater than fallout levels[19] and airborne ^{239}Pu concentrations in general were greater than airborne ^{238}Pu concentrations. Measured airborne concentrations were significantly less than maximum permissible concentrations[18] (MPC's).

FLUXES OF PLUTONIUM ON NONRESPIRABLE PARTICLES AT ROCKY FLATS AND HANFORD

Airborne plutonium fluxes on nonrespirable particles were calculated for both ^{238}Pu and ^{239}Pu. The Rocky Flats data[7] are shown in Figures 7 and 8.

In Figure 7, the airborne ^{239}Pu nonrespirable horizontal flux in μCi/(m^2 day) is shown as a function of sampling distance and sampling height. As might be expected, the maximum airborne ^{239}Pu flux transported by nonrespirable particles was at Site A near the original oil storage area. The maximum airborne ^{239}Pu flux was 6 x 10^{-4} μCi/(m^2 day). The airborne flux decreased with both distance and sampling height. At Site A, the ^{239}Pu flux decreased over one 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 one 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 two 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 Rocky Flats 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 show the airborne ^{239}Pu flux did not significantly decrease for heights greater than 1 m up to 10 m. The nonrespirable particle plume above 10 m was not sampled.

Similar results are shown in Figure 8 for total ^{238}Pu flux on nonrespirable particles at Rocky Flats as a function of sampling site and sampling height. The maximum airborne ^{238}Pu nonrespirable flux was 1.2 x 10^{-5} μCi/(m^2 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 was made. Airborne ^{238}Pu fluxes at 2 and 10 m heights at Site B were greater than at Site AB. An explanation for this increase is not apparent, but the increase is supported by comparing plutonium analyses uncertainties. Error bars show a ^{238}Pu flux range for Site B significantly above the flux range around Site AB.

Airborne nonrespirable fluxes at Rocky Flats were greatest near the original oil storage area (source of contaminated leakage) and near ground level. Fluxes of ^{239}Pu ranged from 10^{-7} up to 10^{-3} μCi/(m^2 day). In contrast, fluxes of ^{238}Pu ranged from 10^{-8} to 10^{-5} μCi/(m^2 day).

Nonrespirable airborne plutonium fluxes around U-Pond on the Hanford Reservation are shown[20] in Figure 9. The ^{238}Pu flux was less than the ^{239}Pu flux. This decrease is similar to the Rocky Flats data. However, the U-Pond data show the nonrespirable plutonium flux extends at least up to 30 m above ground level. Also, there was a greater airborne plutonium flux east of U-Pond than west of U-Pond. This is to be expected since prevailing winds are from the west.

At U-Pond, the airborne ^{238}Pu flux ranged from 10^{-8} to 10^{-7} $\mu Ci/(m^2\ day)$, which is within the midrange of 10^{-8} to about 10^{-5} $\mu Ci/(m^2\ day)$ measured at Rocky Flats. Similarly, the ^{239}Pu flux at U-Pond ranged from about 10^{-6} to 10^{-5} $\mu Ci/m^2\ day)$, which is within the 10^{-7} to 10^{-3} $\mu Ci/(m^2\ day)$ measured at Rocky Flats. The bracketing of the airborne nonrespirable particle fluxes near U-Pond and at Rocky Flats even within three to four orders of magnitude may be coincidental since surface sources and other factors are peculiar to each site.

PLUTONIUM CONCENTRATION PER GRAM OF AIRBORNE SOIL

Airborne plutonium concentrations were normalized to the soil collected with the airborne plutonium. Plutonium concentrations in $\mu Ci/g$ were determined as a function of particle diameter as determined with both particle cascade impactors for respirable diameters as well as sieve sizes for nonrespirable particles. Resuspended plutonium is attached to nonrespirable as well as respirable particles. Hence, nonrespirable soil particles may contribute significantly to downwind airborne plutonium concentrations and represent one mechanism for transporting plutonium to surrounding land.

For Rocky Flats, nonrespirable soil collected at 0.3 m above ground level was sieve sized[7] into twelve different size increments. Each size increment was analyzed for ^{238}Pu and ^{239}Pu. Plutonium concentrations were normalized ($\mu Ci/g$) to the grams of soil collected within each size increment. Results are shown in Figure 10 for ^{239}Pu as a function of particle size at Sites A and AB. Plutonium-239 was associated with all particle sizes. The maximum concentration was about 10^{-4} $\mu Ci/g$ for particle diameters between 10 and 20 μm. For larger particle diameters up to 230 μm, concentrations tended to decrease with an increase in particle diameter. Concentrations at Site A were greater than at Site AB. This is expected since Site A was closer to the original oil storage area at which plutonium leakage occurred.

At each site, plutonium concentrations in $\mu Ci/g$ 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. The relationship is complicated by collection of both contaminated onsite and uncontaminated offsite nonrespirable particles within the cowls.

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

Plutonium-238 concentrations in $\mu Ci/g$ on airborne soil are shown in Figure 11. In this case, only nonrespirable particle diameter ranges are shown. There was insufficient ^{238}Pu collected in the particle cascade impactor samples to yield statistically significant results in respirable diameter ranges. Similar to ^{239}Pu, ^{238}Pu nonrespirable particle concentrations were greater at Site A than at Site B and also showed an inverse relationship with particle diameter.

Both ^{239}Pu and ^{238}Pu concentrations on airborne soil decreased from Site A to Site AB. Site AB/Site A ratios of $\mu Ci/g$ are shown in Figure 12. Concentrations per gram decreased by up to about 10^{-3} in the intervening 392-m distance separating sampling Sites A and AB. Concentration ratios for ^{239}Pu on respirable particles were about 10^{-1} and were independent of particle size. In contrast for nonrespirable particle sizes, ^{239}Pu ratios between sites decreased nearly linearly as particle diameter increased. Plutonium-239 concentrations on nonrespirable particles decreased at rates greater than the ^{238}Pu decrease. For ^{238}Pu, the $\mu Ci/g$ ratios for Site AB/Site A were nearly one order of magnitude greater than ^{239}Pu. These larger ratios suggest ^{238}Pu resuspended more readily relative to ^{239}Pu.

For onsite Hanford experiments, airborne ^{238}Pu and ^{239}Pu concentrations expressed in $\mu Ci/g$ of airborne solids are shown in Figure 13 for collection on filters. Concentrations of ^{239}Pu were somewhat greater than those of ^{238}Pu. Plutonium concentrations on airborne solids ranged from

10^{-9} to 10^{-5} µCi/g. The only exception was the October 1973 single sample described above, for which the concentration was 6 x 10^{-5} µCi/g. Otherwise, plutonium concentrations on airborne solids around U-Pond appeared to be nearly independent of time.

Plutonium concentrations on airborne nonrespirable particles were also determined[20] near U-Pond on the Hanford reservation for sampling heights from 0.3 up to 30 m above ground. Airborne solids were sampled continuously for all wind directions at sites both east and west of U-Pond. The distance between sampling sites was 480 m. Samples were analyzed for both ^{239}Pu as well as ^{238}Pu. Calculated results shown in Figure 14 are for µCi/g of nonrespirable airborne solids collected within cowls. Results show plutonium concentrations on airborne nonrespirable solids were approximately one order of magnitude higher east as compared to west of U-Pond. This increase is caused by prevailing west winds which caused resuspension from this low level liquid waste disposal area. East of U-Pond, plutonium concentrations on airborne nonrespirable solids tended to be uniform with height up to 30 m. The plume height above 30 m is unknown.

Plutonium concentrations on nonrespirable airborne solids are within the range shown in Figure 13 for smaller particles collected on filters. In both cases, ^{239}Pu concentrations ranged from 10^{-7} to 10^{-5} µCi/g. However, ^{238}Pu concentrations on respirable solids collected on filters tended to be greater than on the nonrespirable particles.

Both ^{238}Pu and ^{239}Pu concentrations on nonrespirable airborne solids near U-Pond were less than concentrations determined at Rocky Flats site A. This comparison can be seen by comparing data in Figures 10 and 11 with data in Figure 14. However, plutonium concentrations on airborne nonrespirable solids at Hanford's U-Pond and Rocky Flats site AB tended to be comparable.

AIRBORNE AND GROUND PLUTONIUM µCi/g RATIOS

The previous data showed there was a great variability in the µCi/g ratio for airborne soil at sites A and AB. These µCi/g airborne ratios will now be compared to µCi/g ratios for surface soil in order to determine if any relationship exists between Pu concentrations on airborne soil and Pu concentrations on ground surface soil. Identification of relationships between sites of radionuclide concentrations on airborne solids and contaminated ground solids would be useful in establishing criteria for releasing contaminated areas for other uses.

At Rocky Flats 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 µCi/g. Similarly, ground surface ^{238}Pu/^{239}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 Figures 10 and 11 data limits. Maximum ranges of µCi/g airborne compared to µCi/g in the ground surface soil are shown in the last two columns. These ratios range from 1 x 10^{-4} up to 2. Thus in hazard evaluations[3] 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 were significantly less than Pu concentrations on ground surface soils.

Concentrations on Hanford ground surfaces obtained from the literature[21-25] are shown in Table 3 for ^{239}Pu as well as ^{137}Cs in µCi/g of solids. The range for ^{239}Pu was from less than radiochemical detection limits to 6.9 x 10^{-7} µCi/g of surface solids. The maximum reported ^{239}Pu concentrations for surface solids were within the 200 areas. The minimum ^{239}Pu concentrations on surface solids reported[21] in the literature occurred at 19 to 34 km from Hanford. Surface contamination levels were reported only out to 34 km. The ^{137}Cs concentrations ranged from 3 x 10^{-8} to 2.5 x 10^{-2} µCi/g. These contamination levels are used in Table 3.

Table 4 is a summary of Hanford airborne solids concentrations[2] in µCi/g for ^{239}Pu and ^{137}Cs. Plutonium concentrations were obtained from Figure 13. Table 4 also shows ground surface contamination levels for ^{239}Pu and ^{137}Cs, expressed in µCi/g. From these, ratios of µCi/g of airborne solids to µCi/g surface soil were determined. The last column shows maximum ratio ranges. Ratios of µCi/g airborne to µCi/g surface soil vary from 10^{-3} to 8 x 10^{3}, indicating that plutonium levels on airborne solids can be either much less than or much greater than plutonium levels on contaminated surface solids.

Caution should be used in interpreting these plutonium data. The ground surface plutonium contamination data are limited in quantity and were not necessarily obtained in the same areas where resuspension experiments were performed. Consequently airborne particle and ground contamination levels are also shown for BC-Crib area in the last two columns of Table 4. In this case, BC-Crib area was sampled[25] in ten 1-m^2 areas. Data from these ten squares indicated surface contamination levels varied by about a factor of 100. Ratio ranges of µCi/g airborne to µCi/g surface soil calculated from these data may be more representative than ranges calculated for ^{239}Pu. The ratio of µCi/g airborne soil to µCi/g surface soil for ^{137}Cs data for BC-Crib area ranged from 10^{-2} to 30. The magnitude of this range is important. The range indicates that the µCi/g airborne soil is usually not equal to µCi/g surface soil. This nonequality is caused in part by the great variability and hence uncertainty in local surface contamination levels within any small area and also by dilution of the plutonium concentration on airborne soil by simultaneously sampling less (or more) contaminated soil resuspended upwind.

PLUTONIUM RESUSPENSION FROM OFFSITE NEAR HANFORD

Plutonium on offsite locations near nuclear facilities has deposited from fallout as well as from onsite low level releases. Resuspension of this offsite plutonium was studied near the Prosser barricade on the Hanford reservation. The Prosser barricade is located about 19 to 20 km southeast (130 to 160°) from the fuel processing areas. Both respirable and nonrespirable airborne solids were collected by sampling with particle cascade impactors and rotating cowl systems. Air sampling was only when wind was blowing from 190-260°. This range of southwest (225°) winds came from offsite towards the Hanford reservation. All southwest winds were continuously sampled with rotating cowl systems for nonrespirable particles while respirable particles were sampled with particle cascade impactors for wind speed increments of 3 to 5, 5 to 7, and 7 to 11 m/sec at a height of 1.5 m.

Airborne respirable plutonium concentrations blowing in from offsite are shown in Figure 15 for the particle cascade impactor data. Both airborne concentrations in µCi/cm^3 air and µCi/g collected solid are given. Airborne plutonium concentrations determined with particle cascade impactors are shown as a function of wind speed increments for plutonium collected on the impactor backup filter, the 1.1 plus 2.0 µm stages, and the 3.3 plus 7 µm stages.

Airborne respirable plutonium concentrations increased with increasing wind speed. Concentrations increased up to about 2 orders of magnitude as wind speed increased from 3 to 5 up to 7 to 11 m/sec. Straight lines are drawn through data in order to direct attention to the wind speed dependency of the data. For the plutonium collected on the cascade impactor backup filter, lines proportional to wind speed to the 1.1 and 4.2 power are shown. For wind speeds below abut 5 m/sec, airborne plutonium concentrations tended to increase nearly linearly with wind speed. However, above 5 m/sec, plutonium concentrations increased with wind speed to the 4.2 power. At the present time, it is unknown whether these indicated relationships would suggest a threshold windspeed of 5 m/sec for resuspension or whether a smooth curve should be drawn through all data points.

In other portions of this figure, selected straight lines suggesting wind speed dependencies of respirable airborne plutonium concentrations are shown only for wind speeds above 5 m/sec. For the 1.1 plus 2.0 µm impactor stages, airborne concentrations increased with wind speed to the 9.3 power. For the 3.3 plus 7 µm impactor stages, airborne concentrations increased with wind speed to the 5.2 power. For total plutonium collection within particle cascade impactors, a range of wind speed dependency is shown on the right side of the figure. For a sampling height of 0.3 m, air concentrations increased with wind speed to the 4.4 power. However, at a sampling height of 1.8 m, airborne concentrations increased with wind speed to the 3.0 power.

These data for airborne plutonium concentration in Figure 15 show plutonium is resuspended and show a very high wind speed dependency for this offsite plutonium respirable resuspension. However, there are other plutonium resuspension data which show a different wind speed dependency. West of U-Pond on the Hanford reservation, airborne plutonium concentrations increased[2] with wind speed to only the 1.5 power. Reasons for these differences in the wind speed dependency of onsite versus offsite plutonium resuspension are unknown. Possibly a threshold wind velocity above which resuspension increases rapidly with wind speed was not exceeded at U-Pond.

Nonrespirable airborne plutonium fluxes blowing from offsite onto the Hanford reservation were also measured. In this case, sampling direction was controlled by placing stops which allowed the rotating cowl (Figure 3) inlet to rotate only with the range of 190° to 260°. Plutonium analysis was for the total nonrespirable solids collection in cowls at each height, rather than as a function of particle size as was done for Rocky Flats (See Figures 10 and 11.)

Plutonium-239 concentrations in μCi/g and fluxes in μCi/(m^2 day) for nonrespirable particles blowing from offsite near the Hanford Prosser barricade are shown in Table 5. Plutonium concentrations on nonrespirable airborne solids ranged from 1.3×10^{-7} up to 2.1×10^{-7} μCi/g. These concentrations are similar to those shown in Figure 14 for airborne nonrespirable particles collected west of U-Pond.

Airborne nonrespirable ^{239}Pu fluxes were calculated using two sampling times for these Prosser barricade samples. Horizontal flux calculations were made for both the total time wind was between 3 and 11 m/sec and 190° to 260° and for the total time cowl air samplers were in the field. The fluxes are shown in the last two columns of Table 5. When the shorter time period (3 to 11 m/sec winds) is used for calculating the horizontal plutonium flux, fluxes range from 3.9×10^{-6} to 1.4×10^{-6} μCi/(m^2 day).

A possible explanation of this offsite plutonium resuspension was sought. Experimental results for μCi/g airborne soil and plutonium isotopic ratios were determined. As shown in Table 5 the range of ^{239}Pu concentrations on airborne soil was from 1.3×10^{-7} up to 2.1×10^{-7} μCi/g. This range is greater than fallout levels in soil surface samples. As shown in Table 3, reported ^{239}Pu concentrations in surface samples 19 to 34 km from Hanford had a range from 3.6×10^{-9} to 7.6×10^{-8} μCi/g. These last values are similar to a fallout level of 3.8×10^{-8} μCi/g measured at North Eastham, MA.[26]

Most plutonium collected appears not to have originated from fallout. Rather, most plutonium collected on these airborne nonrespirable particles near the Prosser barricade resembles weapons grade plutonium.[27,28] Plutonium isotopic ratios ($^{240}Pu/^{239+240}Pu$) in atom percent for these nonrespirable samples were 6.10 ± 0.02 at 0.3 m height, 6.31 ± 0.02 at 2 m height, and 6.28 ± 0.03 at 5.8 m height. In comparison, the isotopic ratio determined from a sample of forest fire smoke plume near Mt. St. Helens, WA, was 13.82 ± 0.05. Isotopic ratios for respirable particles sampled near the Prosser barricade were not determined. Although plutonium was blowing from offsite near the Prosser barricade, airborne respirable plutonium concentrations were far below maximum permissible concentrations as was shown by Figure 15.

CONCLUSIONS

Both ^{238}Pu and ^{239}Pu resuspension occurred onsite at Rocky Flats and the Hanford reservation, but all airborne plutonium concentrations were significantly below maximum permissible concentrations in air. In addition, plutonium was resuspended from offsite near the Hanford reservation. In all cases plutonium was deposited on each stage of particle cascade impactors, showing that most plutonium was resuspended while attached to larger host soil particles.

Plutonium concentrations per gram of both respirable and nonrespirable airborne soils are summarized in Table 6. Agreement is within several orders of magnitude from 2×10^{-8} to 6×10^{-5} μCi/g for respirable ^{239}Pu and from 1×10^{-7} to 3×10^{-4} μCi/g for nonrespirable ^{239}Pu. Plutonium concentrations on collected airborne soil at Rocky Flats 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[8] surface soil concentration was 3×10^{-3} μCi/g. Similarly at the Hanford resuspension study sites,[2] maximum concentrations per gram of airborne solid were for ^{239}Pu, 6×10^{-5} μCi/g and for ^{238}Pu, 1×10^{-6} μCi/g. There is much uncertainty (several orders of magnitude) in the relationships between radionuclide concentrations per gram of airborne solid and per gram of surface soil.

Radionuclide particles may be resuspended[1,2] either as individual particles or, more probably attached to host soil or solid particles. An average or "normal"-activity radionuclide particle distribution is usually collected on sampling filters. However, at both Rocky Flats and Hanford, one filter sample collected in each case showed significantly greater plutonium concentration than the maximum for all other samples collected during the same time period. These anomalous higher concentrations are attributed to one or more plutonium particles of unusually higher activity than these normally, or most frequently, resuspended.

There are many variables such as wind speed, vegetative cover, upwind source contamination levels, the amount of airborne soils, and others which could influence fluxes of plutonium on nonrespirable particles. At each site these variables were different. Nevertheless, it is instructive to summarize the measured range of fluxes. The plutonium fluxes on nonrespirable particles offsite at Hanford, onsite at Hanford U-Pond, as well as onsite at Rocky Flats for the time periods investigated are summarized in Table 7. The Prosser barricade flux range is within the range measured near Site A at Rocky Flats which was shown in Figure 7. However, if the total time cowl air samplers were in the field is used for calculation, Prosser barricade airborne nonrespirable offsite plutonium fluxes were lower and within the range measured at Rocky Flats Sites AB and B (See Figure 7). These data and the

decrease of flux with distance[1,2] represent, as far as we can determine, the present knowledge on this subject. Agreement is only within several orders of magnitude for nonrespirable airborne plutonium fluxes. The range of this flux may be caused in part by the interplay of relatively more soil transport with a lower plutonium-on-soil concentration and lower soil transport and concurrent higher plutonium-on-soil concentration.

Airborne ^{239}Pu concentrations increased[1,2] as a function of wind speed to the 1.5 to 6 power for on-site resuspension study sites. Airborne plutonium concentrations for off-site resuspension increased as a power function of wind speed. Above a wind speed of about 5 m/sec, plutonium air concentrations increased with wind speed to the 3 to 5th power.

ACKNOWLEDGMENT

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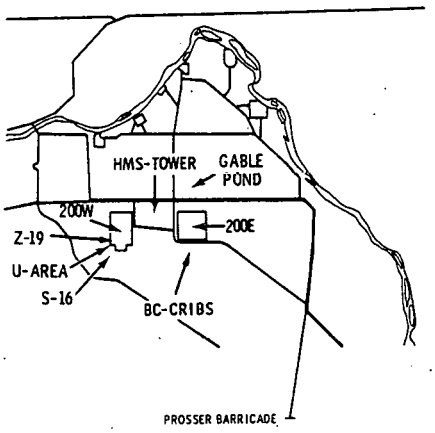

FIG. 1. Location of Hanford Study Sites

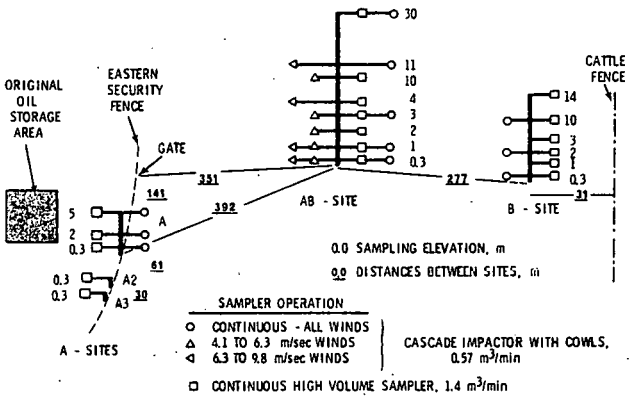

FIG. 2. Schematic of Rocky Flats Resuspension Tower Locations

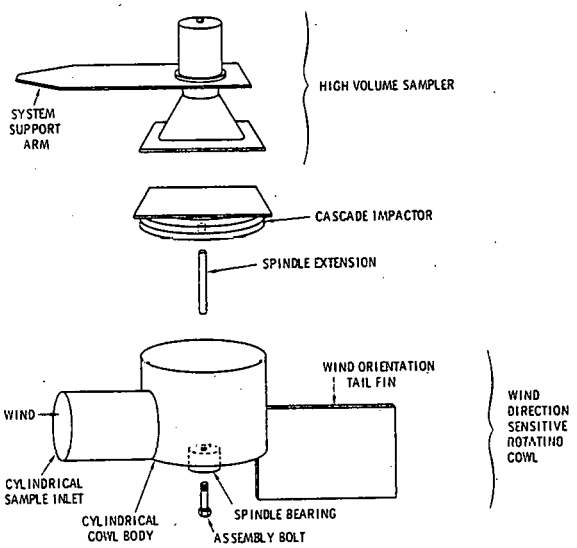

FIG. 3. Rotating Cowl and Impactor

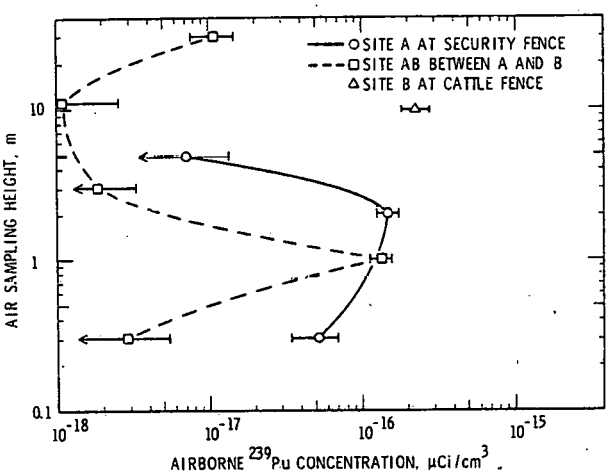

FIG. 4. Airborne ^{239}Pu Concentrations from Impactor 2.0 μm Stage Collections at Rocky Flats for all Winds

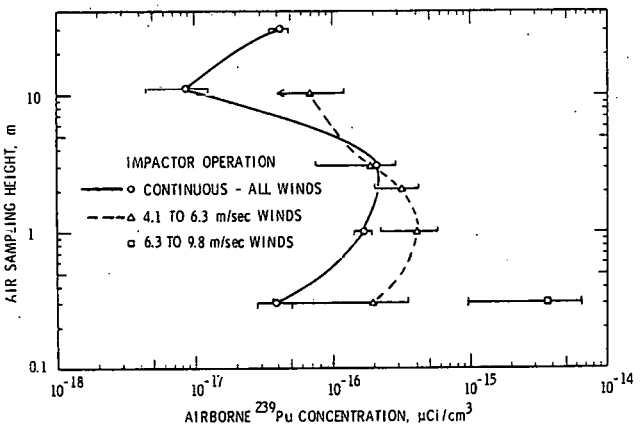

FIG. 5. Total Airborne ^{239}Pu Concentration at AB Site at Rocky Flats as a Function of Wind Speed

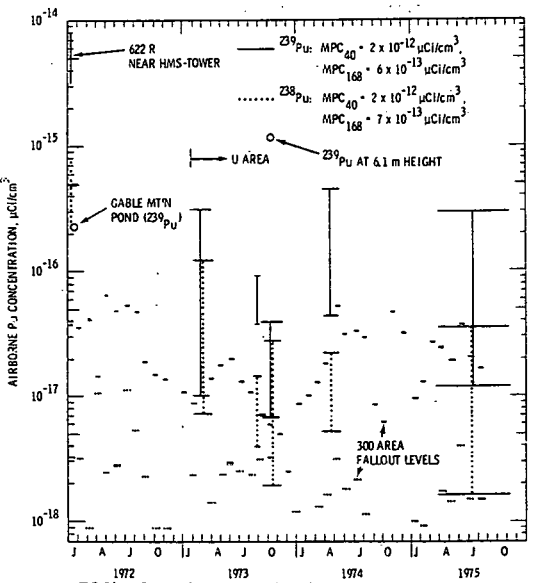

FIG. 6. Range of Airborne Plutonium Concentrations at Onsite Hanford Resuspension Sites Compared to Fallout Levels

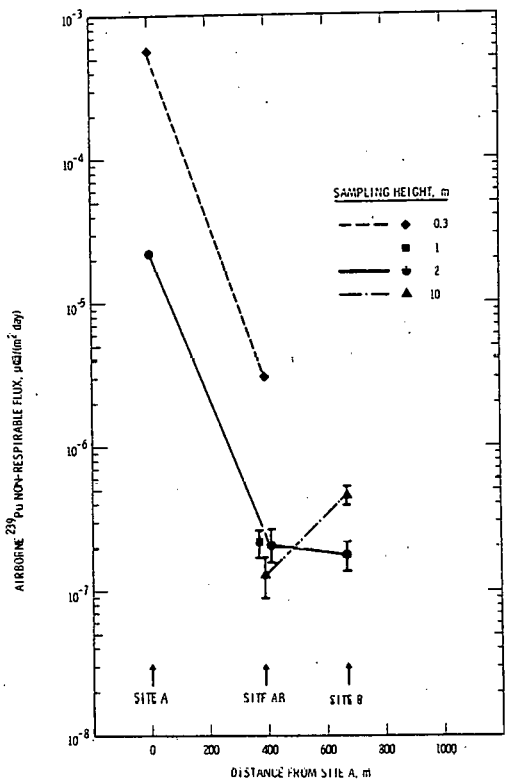

FIG. 7. Decrease with Distance of Total ^{239}Pu Flux on Nonrespirable Particles at Rocky Flats

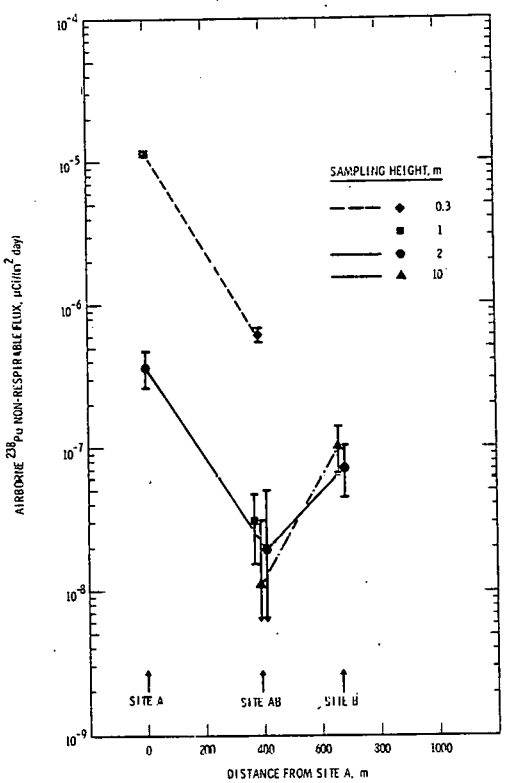

FIG. 8. Decrease with Distance of Total ^{238}Pu Flux on Nonrespirable Particles at Rocky Flats

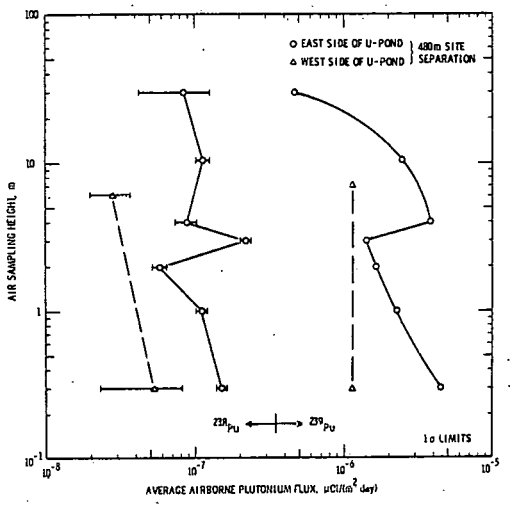

FIG. 9. ^{239}Pu and ^{238}Pu Airborne Fluxes on Nonrespirable Particles at Hanford U-Pond during February 27 - November 10, 1975 (Particles Collected in Cowls)

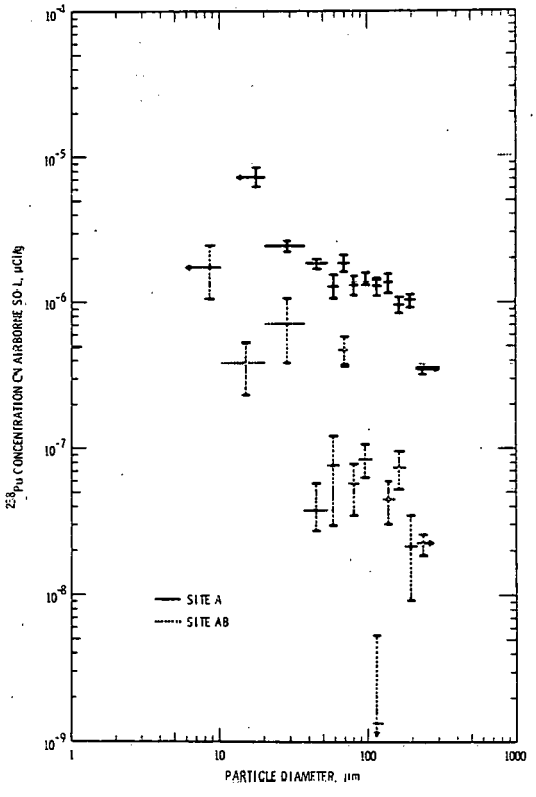

FIG. 11. ^{238}Pu Concentration on Airborne Soil as a Function of Particle Diameter at Rocky Flats

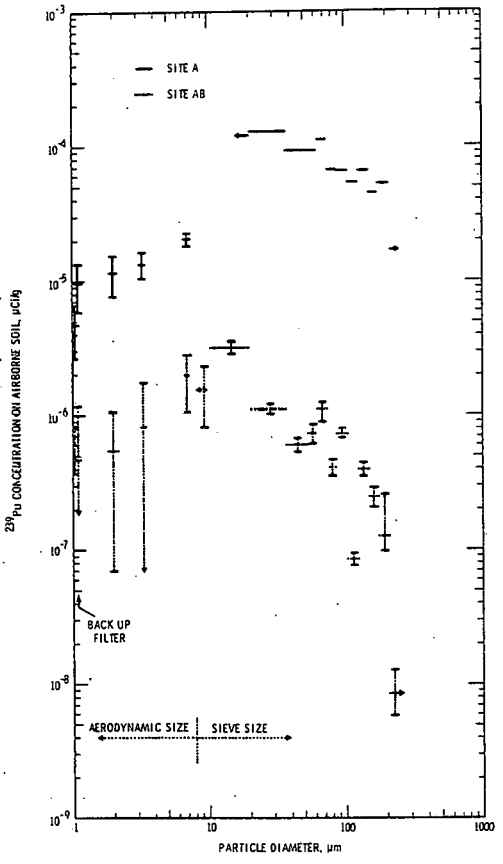

FIG. 10. ^{239}Pu Concentration on Airborne Soil versus Particle Diameter at Rocky Flats

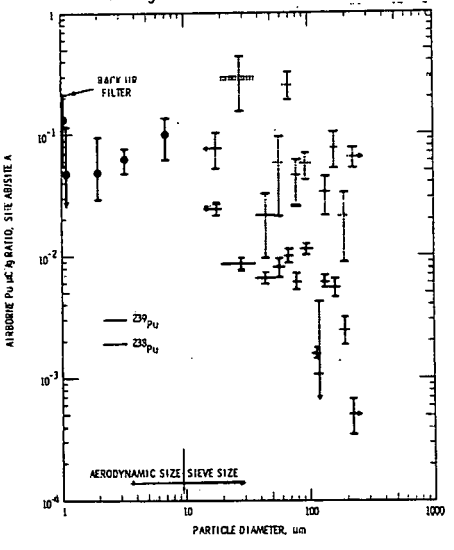

FIG. 12. Decrease in ^{238}Pu and ^{239}Pu Concentrations per Gram of Airborne Particles: 392 m from Site A to Site AB at Rocky Flats

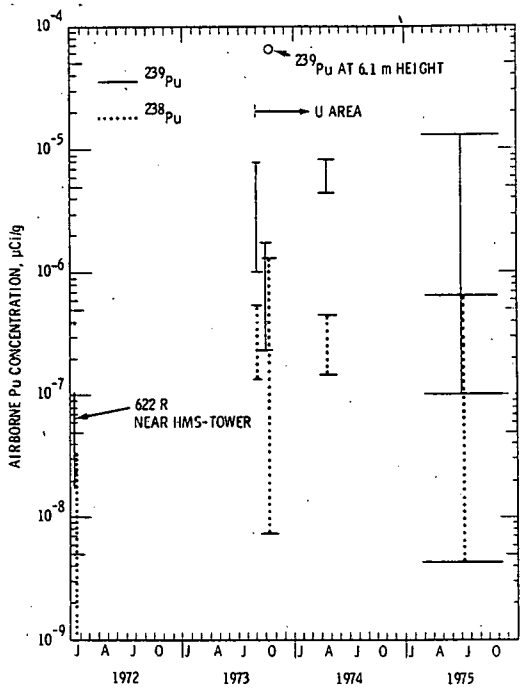

FIG. 13. Range of Plutonium Concentration on Airborne Solids at Onsite Hanford Resuspension Sites

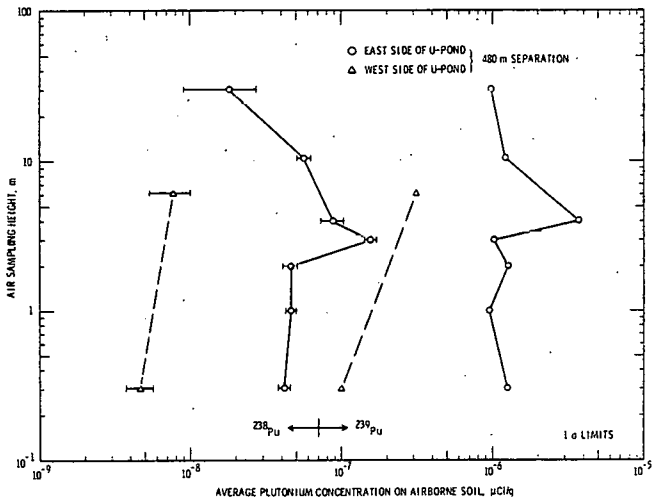

FIG. 14. Concentration of ^{238}Pu and ^{239}Pu on Non-respirable Airborne Solids at Hanford U-Pond during February 27 - November 10, 1975 (Particles Collected in Cowls)

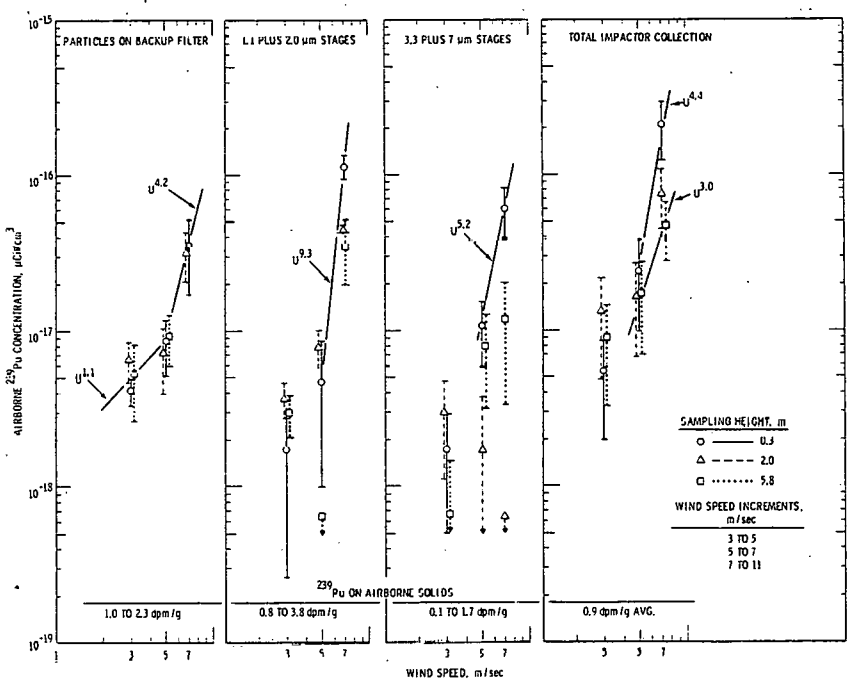

FIG. 15. Airborne ^{239} Pu Concentrations Near Prosser Barricade at Hanford from April 12 to June 29, 1976 when Sampling when Winds were from 190° to 260°

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TABLE 1. Selected Surface Soil Sample Results at Rocky Flats

Locations Near Site	^{239}Pu, µCi/g	^{238}Pu, µCi/g
A	3.10×10^{-3} to 6.89×10^{-4}	5.77×10^{-5} to 1.24×10^{-5}
AB	7.70×10^{-5} to 3.86×10^{-5}	1.56×10^{-6} to 7.66×10^{-7}
B	2.66×10^{-6} to 3.85×10^{-5}	5.09×10^{-8} to 7.12×10^{-7}

TABLE 2. Ratio of Plutonium Concentration per Gram of Soil: Airborne Soil/Ground Surface Soil at Rocky Flats

	Range, µCi/g				Ratio Range, µCi/g Airborne / µCi/g Surface	
	^{239}Pu		^{238}Pu			
Site	Airborne*	Surface**	Airborne***	Surface**	^{239}Pu	^{238}Pu
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 10 as a function of particle diameter
** From Table 1 for total ground surface sample 5 cm deep
*** From Figure 11 as a function of particle diameter

TABLE 3. Concentrations of Plutonium and Cesium in Hanford Ground Surface Solid Samples Reported in the Literature

		μCi/g		
		^{239}Pu	^{137}Cs	
Location	Year	Range	Average	Range
Onsite[22]	1973	*LAL to 8×10^{-9}	9.7×10^{-7}	3×10^{-8} to 2.4×10^{-6}
Inside 200 Areas[21]	1971	2.6×10^{-8} to 6.9×10^{-7}		NR
BC Area[21-24]	1974			2.1×10^{-5} to 2.5×10^{-2}
Within Hanford[22] Site Boundary (3 to 26 km)	1971	9.9×10^{-9} to 1.2×10^{-7}		NR
Site Perimeter[22]	1973	*LAL to 6×10^{-9}	7×10^{-7}	1.4×10^{-7} to 1.5×10^{-6}
Offsite[21] (19 to 34 km)	1971	3.6×10^{-9} to 7.6×10^{-8}		NR

*LAL, less than analytical limit
NR, not reported

TABLE 4. Ratio of Airborne to Ground Surface Radioactivity Concentrations per Gram of Solids at Hanford

	Concentration, μCi/g Solids				Maximum Ratio Range, (μCi/g)air
	Airborne Solids		Ground Surface Contamination		
Material	Minimum	Maximum	Minimum	Maximum	(μCi/g)surface
^{239}Pu[22]	2×10^{-8}	6×10^{-5}	*LAL	8×10^{-9}	2×10^{0} to 8×10^{3}
^{137}Cs at BC-Crib Area[29]	3×10^{-5}	7×10^{-4}			
10 areas 1 m^2			2.1×10^{-5}	1.2×10^{-3}	1×10^{-3} to
Maximum[24]				2.5×10^{-2}	3×10^{1}

* LAL, less than analytical limit

TABLE 5. Plutonium Transport on Nonrespirable Particles from Offsite Near the Prosser Barricade on the Hanford Reservation

Sampling Height, m	^{239}Pu on Airborne Solids		Airborne ^{239}Pu Nonrespirable Flux, $\mu Ci/(m^2\ day)$ for	
	dpm/g	$\mu Ci/g$	Only for 190 to 260° Winds, 3 to 11 m/sec	Total Field Time
0.3	0.29	1.3×10^{-7}	3.9×10^{-6}	8.0×10^{-7}
2	0.46	2.1×10^{-7}	4.0×10^{-6}	8.3×10^{-7}
5.8	0.32	1.5×10^{-7}	1.4×10^{-6}	2.8×10^{-7}

TABLE 6. Summary of Plutonium Concentrations on Total Airborne Solids

Site	Total Airborne $\mu Ci/g$ Range			
	^{238}Pu		^{239}Pu	
	Respirable*	Nonrespirable	Respirable*	Nonrespirable
Rocky Flats[6,7]	NR	2×10^{-7} to 5.1×10^{-6}	2.1×10^{-6} to 6.2×10^{-5}	1×10^{-6} to 3.1×10^{-4}
Hanford Reservation				
On-site[2]	1×10^{-9} to 1×10^{-6}	4×10^{-9} to 1×10^{-7}	2×10^{-8} to 6×10^{-5}	1×10^{-7} to 4×10^{-6}
From Off-site near Prosser Barricade	NR	NR	5×10^{-8} to 1×10^{-6}	1.3×10^{-7} to 2.1×10^{-7}

NR, no radiochemical results.

* "Respirable" as used in this report are those particles which are found on all filter and impactor stages as constrasted to "nonrespirable" particles collected by gravity in rotating cowls.

TABLE 7. Summary of Plutonium Total Transport Fluxes on Nonrespirable* Particles

Site	Ranges of Total Plutonium Fluxes, μCi/(m^2 day)	
	^{238}Pu	^{239}Pu
Rocky Flats[7]	1×10^{-8} to 1×10^{-5}	1×10^{-7} to 6×10^{-4}
Hanford Reservation		
On-site	2×10^{-8} to 2×10^{-7}	4×10^{-7} to 4×10^{-6}
From Off-site near Prosser Barricade for 190 to 260° Winds	NR	1.4×10^{-6} to 3.9×10^{-6}

NR, no radiochemical results.

* Nonrespirable as used in this report are those particles collected within the rotating cowl shown in Figure 3.