CONF-780212--4

PNL-SA-6771



# Airborne Plutonium-239 and Americium-241 Concentrations Measured from the 125-Meter Hanford Meteorological Tower

by
G. A. Sehmel

January 1978

Pacific Northwest Laboratory
Richland, Washington 99352
Operated for the
U.S. Department of Energy
by

Battelle
Memorial Institute

PNL-SA-6771

## DISCLAIMER

PNL-SA-6771

# AIRBORNE PLUTONIUM-239 AND AMERICIUM-241 CONCENTRATIONS MEASURED FROM THE 125-METER HANFORD METEOROLOGICAL TOWER



Paper Prepared for Presentation at the Nevada Applied Ecology Group Plutonium Information Conference on February 28 - March 2, 1978 at San Diego, CA

by
G. A. Sehmel

This work was prepared for the Department of Energy under Contract EY-76-C-06-1830 with Battelle, Pacific Northwest Laboratories



January 1978



BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

THIS PAGE

WAS INTENTIONALLY

LEFT BLANK

CONTENTS

# AIRBORNE PLUTONIUM-239 AND AMERICIUM-241 TRANSPORT MEASURED FROM THE 125-M HANFORD METEOROLOGICAL TOWER

George A. Sehmel

Pacific Northwest Laboratory
Battelle Memorial Institute
Richland, WA 99352

## ABSTRACT

Airborne plutonium-239 and americium-241 concentrations and fluxes were measured at six heights from 1.9 to 122 m on the Hanford meteorological tower. The data show plutonium-239 was transported on nonrespirable and "small" particles at all heights. Airborne americium-241 concentrations on small particles were maximum at the 91 m height.

## INTRODUCTION

Studies at Rocky Flats and Hanford have shown that plutonium is resuspended. For the time periods investigated in these studies (Krey, et al., 1976; Sehmel, 1977a, 1977b, 1977c, 1978; Sehmel and Lloyd, 1976), airborne plutonium-239 concentrations were above fallout levels but never exceeded 2% of the maximum permissible concentration for individuals even in an uncontrolled area. Plutonium was transported on both respirable and nonrespirable particles. In controlled areas, plutonium-239 transport occurred to at least a 30 m height. The maximum plume height was unknown and the relative transport on respirable and nonrespirable particles could only be estimated.

Americium-241 could also be resuspended from plutonium resuspension sites. Although americium-241 resuspension has been reported at low level waste disposal areas on the Hanford area (Sehmel, 1977b, 1977c), there are few reported data showing airborne americium-241 concentrations. Nevertheless, resuspension of americium-241 could become an inhalation concern a hundred years from now after plutonium-241 has decayed to americium-241.

Simultaneous measurements of airborne plutonium-239 and americium-241 concentrations have not been reported outside of exclusion areas. Resuspension studies to date do not suggest differences in resuspension for different radionuclides, but data are limited. Simultaneous measurements of airborne concentrations of both plutonium-239 and americium-241 would indicate similarities which might be expected for different transuranics. Thus, the

objectives of this study were to measure simultaneously both plutonium-239 and americium-241 airborne concentrations and fluxes on both nonrespirable and "small" particles for sampling heights up to 122 m.

## SAMPLING SITE

The sampling site was at the 125 m meteorological tower located outside the exclusion area on the Hanford area. This site is approximately one km east of the 200W Separation Area fence. Air sampling equipment was located at heights of 1.9, 15, 30, 61, 91 and 122 m.

## EXPERIMENTAL PROCEDURE

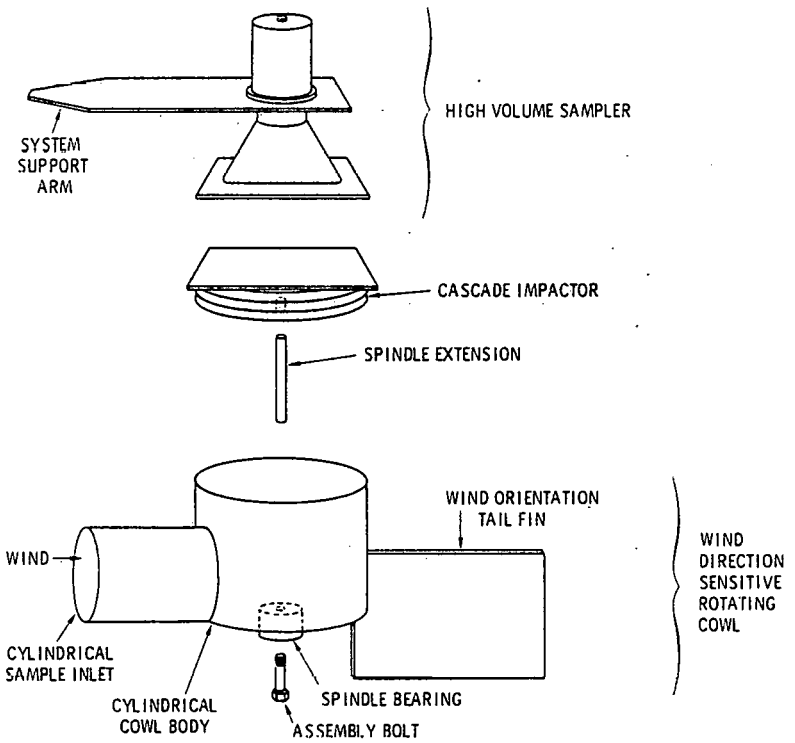
Air sampling was continuous for all wind speeds and directions for the time period between August 13 and November 12, 1976. Air sampling was with the impactor cowl system shown in Fig. 1 (Sehmel, 1973). The 15 cm diameter cowl inlet was continuously directed into the wind by the wind orientation tail fin. Most of the larger particles (greater than about 10 to 20 μm diameter) entering the inlet subsequently settled within the cowl body. Some larger particles were also collected inside the particle cascade impactor (Sehmel, 1973; Willeke, 1975). Interstage loss particles within the cascade impactor which are retrieved by light brushing are of nonrespirable size. It will be assumed all nonrespirable particles entering the impactor are collected as interstage loss particles. The fraction by weight and by plutonium content of these nonrespirable particles on impactor stage collection is considered to have a minimal influence on the general conclusion of this study.

Particles collected within the cowl plus impactor interstage loss particles are called "large" cowl collected particles. Cowl-collected particles are assumed to have been collected independent of the air sampling rate.

Although the "small" impactor-collected particles are an index to the respirable airborne particle concentrations, these small particles do contain some nonrespirable particles. These small particles include all particle collection on each cascade impactor stage except for interstage loss particles. Particles were sampled at a flow rate of 0.57 $m^3$/min and those found on the stages were assumed to be without inertia with respect to sampling efficiency. Thus, small particles were assumed to have followed the sampled air into the cascade particle impactor. For radiochemical analysis, particles collected as interstage losses in the impactors were combined with the large cowl particles.

After sample collection, the impactor* Type "A" fiberglass stage collectors were equilibrated with laboratory humidity (approximately 50% relative humidity), weighed, and all stages and backup filter combined into one "small" particle sample. Particles collected within the cowl and interstage loss particles

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




Neg. 784581-12

FIGURE 1. Rotating Cowl and Impactor

were brushed off for collection, weighed, and combined into one sample. These "small" and "large" particle samples were analyzed** for plutonium-239 and americium-241.

## HORIZONTAL FLUX CALCULATION

Average horizontal airborne plutonium-239 and americium-241 fluxes were calculated from the collected small and large particles. Fluxes are in units of $\mu Ci/(m^2 \text{ day})$ for both large and small particles. Two isokinetic sampling assumptions were made for these calculations: 1) for large particles, particle inertia was assumed sufficient to cause particle collection within the cowl inlet for all wind speeds and 2) for small particles, particles were assumed to be without inertia and followed airflow. For large particles, the flux was calculated from the $\mu Ci$ collected in the cowl (plus interstage impactor loss) divided by the product of cowl inlet cross sectional area and sampling time. For small particles, the flux was calculated from the product of measured airborne concentration per unit air volume sampled in the impactor, $\mu Ci/cm^3$, and average wind speed at each sampling height. The average wind speed at each height was: 1.6 m/sec at 1.9 m; 2.9 m/sec at 15 m; 3.4 m/sec at 30 m; 3.7 m/sec at 61 m; 3.8 m/sec at 91 m; and 4.0 m/sec at 122 m.

## RESULTS AND DISCUSSION

Airborne plutonium-239 and americium-241 concentrations per unit air volume, concentrations per gram of airborne solid, and horizontal fluxes were calculated. Calculated results will be shown with the one sigma radiochemical counting statistic limits around each data point. If limits are not shown, the radiochemical counting limits are within the plotting symbol.

### IMPLICATIONS OF CONCENTRATION PROFILES

Airborne concentration profiles are the airborne concentrations in either $\mu Ci/cm^3$ or $\mu Ci/g$ as a function of height. In some cases airborne concentration profiles of plutonium-239 and americium-241 can be interpreted in terms of possible resuspension sources. Concentration profiles are influenced by the extent and variation of the resuspension source, deposition between the resuspension source and sampling location, and by meteorological parameters such as wind speed and atmospheric stability. For example, the concentration profile for an infinite source would show the $\mu Ci/cm^3$ decreasing with increasing height. For an upwind source and simultaneous airborne plume depletion by deposition, the maximum airborne concentration in $\mu Ci/cm^3$ will be at some elevated height. Similarly for airborne concentration profiles in $\mu Ci/g$, the

** LFE Environmental Laboratories, Richmond, California.

airborne concentration is influenced by sampling both the soil from the contaminant resuspension source as well as airborne soil transported from uncontaminated surfaces. An attempt will be made to interpret the experimental concentration profiles in terms of these possible sources. All interpretations of the observed concentration profiles all carry one qualification: that these average profiles are for samples collected for all wind speeds and directions. Thus, sources of airborne plutonium-239 and americium-241 cannot be identified with respect to direction from the air sampling location.

## SMALL PARTICLE CONCENTRATIONS IN $\mu Ci/cm^3$

Airborne concentrations in $\mu Ci/cm^3$ for "small" particles are shown in Fig. 2. The relative plutonium-239 and americium-241 concentrations for "small" particles show distinctly different concentration profiles. The plutonium-239 airborne concentration was $1.4 \times 10^{-17}$ $\mu Ci/cm^3$ (maximum) at 1.9 m and decreased with increasing height up to 91 m. From 91 m to 122 m, the plutonium-239 concentration increased. In contrast to the decrease, there was a maximum americium-241 concentration of $9.6 \times 10^{-17}$ $\mu Ci/cm^3$ at a 91 m height.

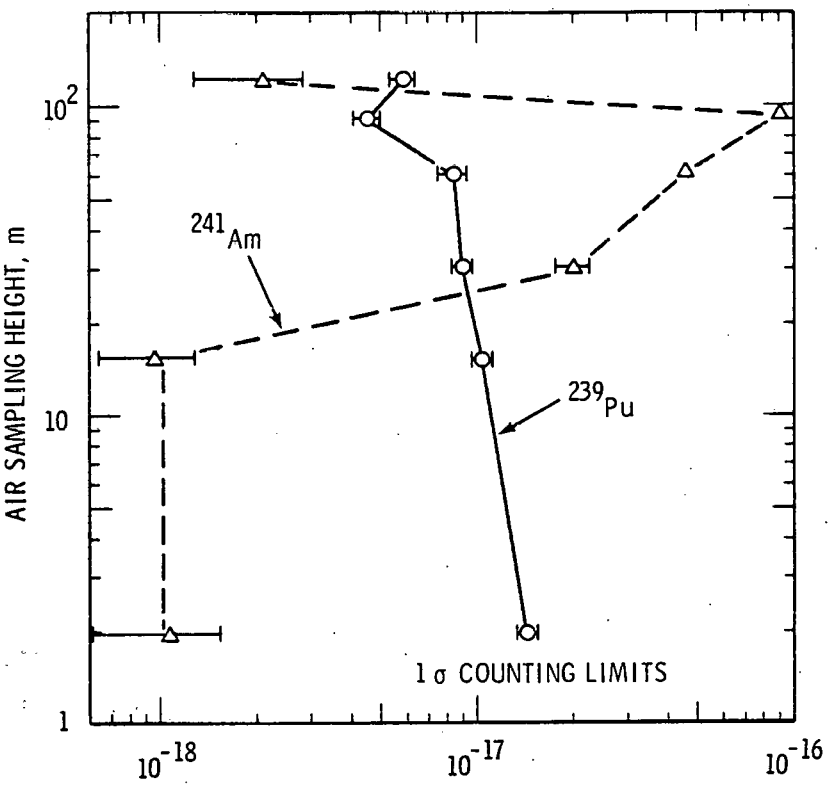
It is unknown why the plutonium-239 airborne concentration in $\mu Ci/cm^3$ for "small" particles at 122 m was greater than at 91 m. The increased airborne concentration at 122 m is attributed in part to a greater plutonium-239 $\mu Ci/g$ at 122 m than at 91 m. Although the increase is unexplained, an increased concentration was also observed, (Sehmel and Lloyd, 1976), at Rocky Flats for sampling heights between 10 and 30 m.

In contrast to the plutonium-239 concentration profile, the americium-241 concentration profile in $\mu Ci/cm^3$ for "small" particles suggests a surface depletion of the airborne plume. This concentration profile indicates either an upwind resuspension source or an elevated release. In any event, the americium-241 source was not determined.

The maximum plutonium-239 concentration was only 0.002% of the maximum permissible airborne concentration (MPC-168hr) for an uncontrolled area (ICRP, 1959). The maximum americium-241 concentration was only 0.005% of the MPC-168hr. For a controlled area, the maximum concentrations were only 0.0002% and 0.002% of the MPC-40hr for plutonium-239 and americium-241 respectively.

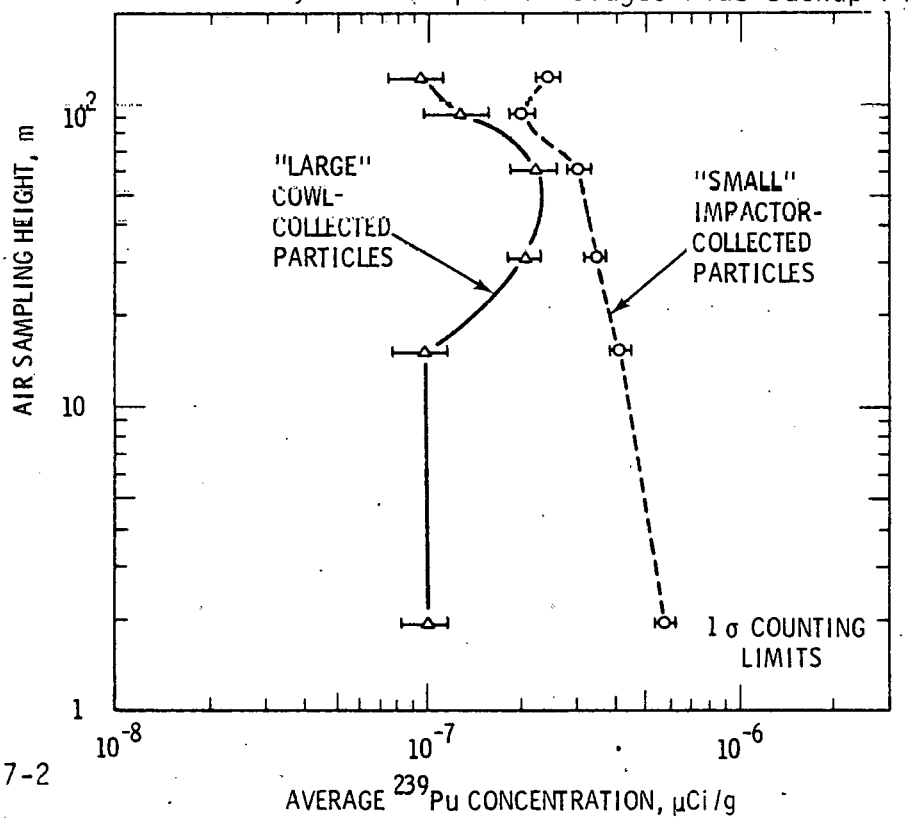
## CONCENTRATIONS IN $\mu Ci/g$ AIRBORNE SOIL

Plutonium-239 and americium-241 concentrations per gram airborne solids were determined for both "small" and "large" particles. Airborne concentrations in $\mu Ci/g$ are shown in Fig. 3 for plutonium-239. The $\mu Ci/g$ for small particles decreased with increasing height up to 91 m. This decrease might be explained by sampling both "more contaminated" locally resuspended particles as well as "less contaminated" soil blowing in from a greater upwind distance. In contrast for large particles, the maximum $\mu Ci/g$ occurred for a sampling height of 30 to 60 m.




Neg. 7802057-6

FIGURE 2. Airborne Plutonium-239 and Americium-241 Concentrations at the Hanford Meteorological Tower During August 13 to November 12, 1976 (Impactor Stages Plus Backup Filter)




Neg. 7802057-2

FIGURE 3. Plutonium-239 on Airborne Solids Collected at the Hanford Meteorological Tower During August 13 to November 12, 1976

This "large"-particle plume concentration profile in μCi/g might be explained by assuming two possible sources of airborne particles were sampled. For one source possibility, contaminated large particles might be resuspending at a distance upwind and the airborne plume be depleted by dry deposition between the resuspension and sampling site. For the second source possibility, many "less contaminated" particles are resuspended near the sampling tower and are sampled. This less contaminated airborne soil being simultaneously sampled with soil from upwind would decrease the μCi/g.

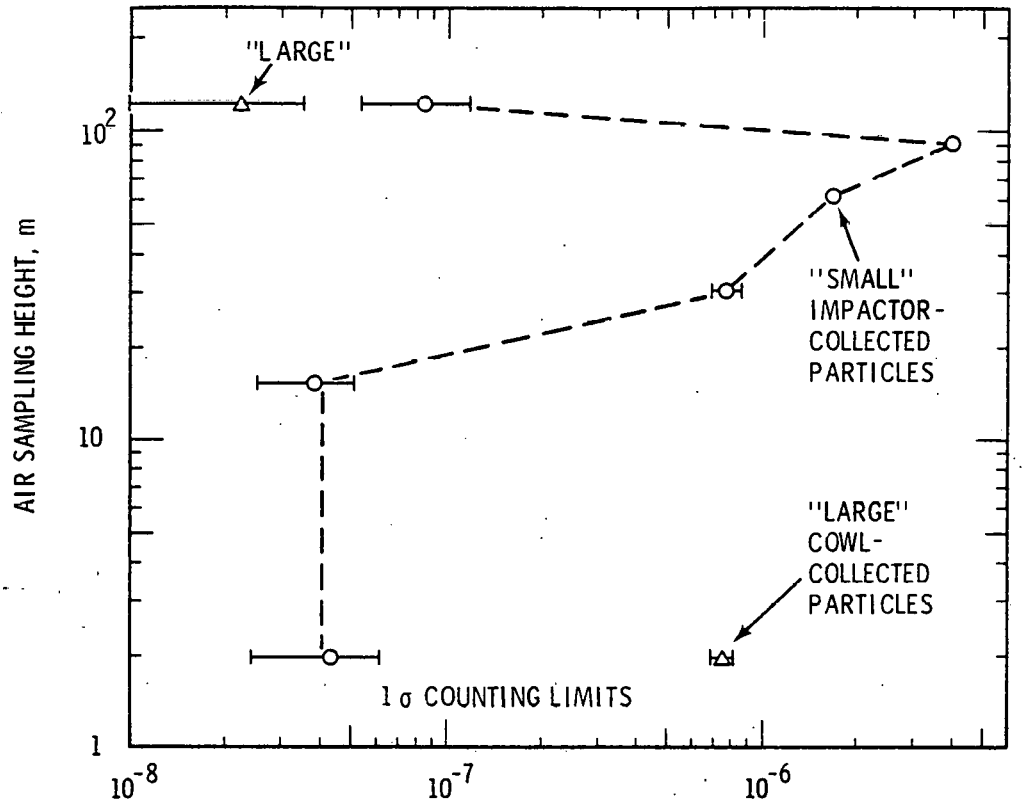
## RELATIONSHIP: PLUTONIUM-239 ON LARGE PARTICLES AND AMERICIUM-241 ON SMALL PARTICLES

Airborne concentration profiles in μCi/g are shown in Fig. 4 for americium-241 collected on both "small" and "large" particles. Only two data points are shown for americium-241 transported on large particles. The maximum observed americium-241 concentration on "large" particles was $8 \times 10^{-7}$ μCi/g at the lowest sampling height of 1.9 m. At the highest sampling height of 122 m, the americium-241 concentration on "large" particles had decreased over one order of magnitude to $2 \times 10^{-8}$ μCi/g. At intermediate sampling heights of 15 to 91 m, americium-241 concentrations were not significantly different from zero due to the radiochemical counting statistic uncertainties.

The americium-241 μCi/g on "small" particles was minimum and constant at 1.9 and 15 m sampling heights. For greater heights, a maximum of $4 \times 10^{-6}$ μCi/g was determined at a sampling height of 91 m. A possible explanation for this americium-241 on "small" particles is that resuspension occurs at a distance upwind and the airborne plume is depleted by particle dry deposition between the resuspension and sampling site. Also, many less-contaminated americium-241 small particles might be resuspended near the sampling tower and be collected in the sample. These less contaminated "small" particles would be simultaneously sampled with "small" particles arising from upwind. Consequently, the combined sample of "small" particles would have an average μCi/g value caused by particle dilution from two sources.

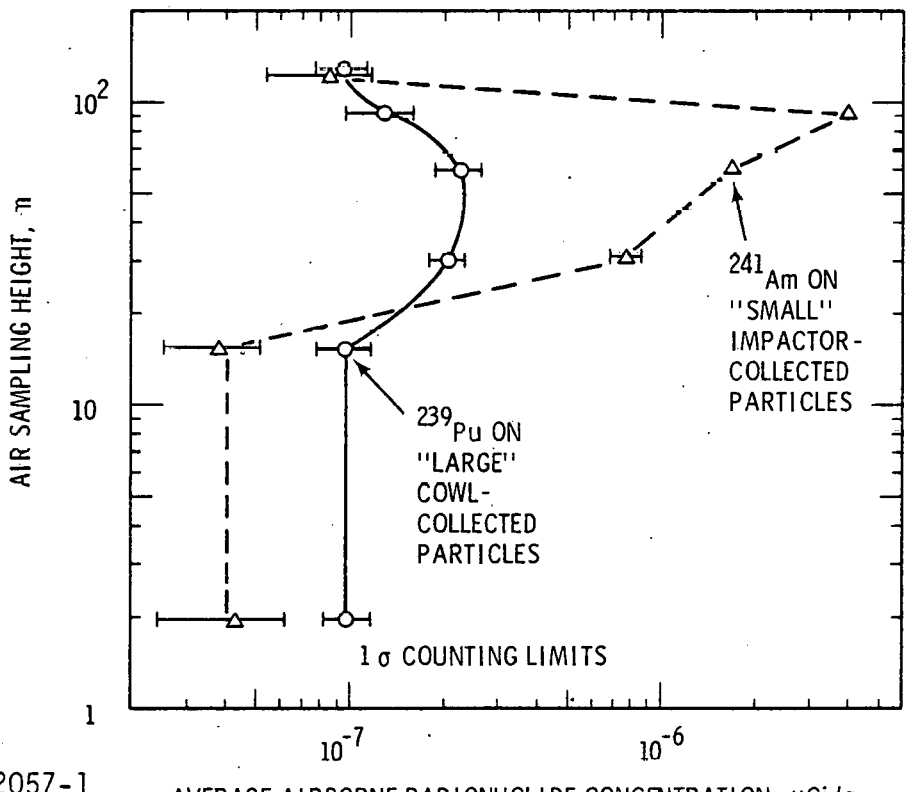
In order to direct attention to a possible relationship between plutonium-239 and americium-241 resuspension, a cross comparison of data from Fig. 3 and Fig. 4 is shown in Fig. 5. In this case, the μCi/g for plutonium-239 on "large" particles is shown along with the μCi/g for americium-241 on "small" particles. In both cases, the μCi/g was constant between 1.9 and 15 m sampling heights. For greater heights, the μCi/g increased for both plutonium-239 on "large" particles and americium-241 on "small" particles. This increase was greater for americium-241 on "small" particles than for plutonium-239 on "large" particles. Nevertheless, for both plutonium-239 on "large" particles and americium-241 on "small" particles, the μCi/g decreased rapidly between sampling heights of 91 m and 122 m.

It is unknown why there appears to be a relationship between americium-241 transport on "small" particles at elevated heights as compared to plutonium-239 transport on "large" particles at elevated heights. There are at least




Neg. 7802057-5

FIGURE 4. Americium-241 on Airborne Solids Collected at the Hanford Meteorological Tower During August 13 to November 12, 1976




Neg. 7802057-1

FIGURE 5. Comparison of µCi/g for Plutonium-239 Transport on "Large" Particles and Americium-241 Transport on "Small" Particles Collected at the Hanford Meteorological Tower During August 13 to November 12, 1976

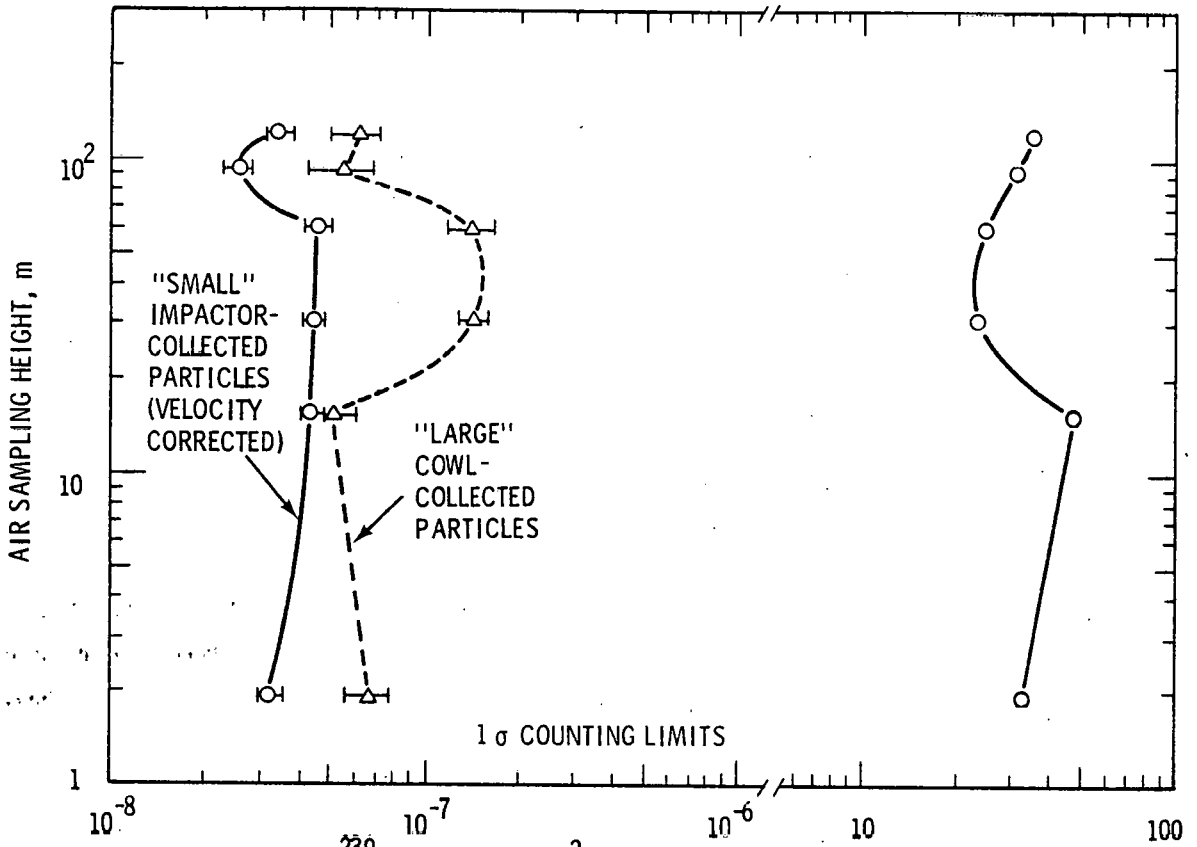
three possible explanations for this relationship. The americium-241 and plutonium-239 were originally deposited at the resuspension source in two different size ranges. A second possibility is that plutonium-239 attaches to soil particles more readily than americium-241. The third possibility is there may be some mechanisms by which americium-241 is released from plutonium-239-241 resuspension sources.

Possibly for an aged plutonium resuspension source, sufficient daughter americium-241 has grown in by decay of plutonium-241. During decay, small americium-241 particles could be ejected from the parent plutonium. After ejection, the americium-241 small particles no longer retain their identity with the parent plutonium which may be attached to large particles. Thus in being resuspended, americium-241 is in a small particle diameter range as compared to the parent plutonium on large particles. Additional plutonium-239 and americium-241 data concerning transport on both large and small soil particles are needed in order to clarify the physics explaining these compared observations.

## AVERAGE HORIZONTAL FLUXES

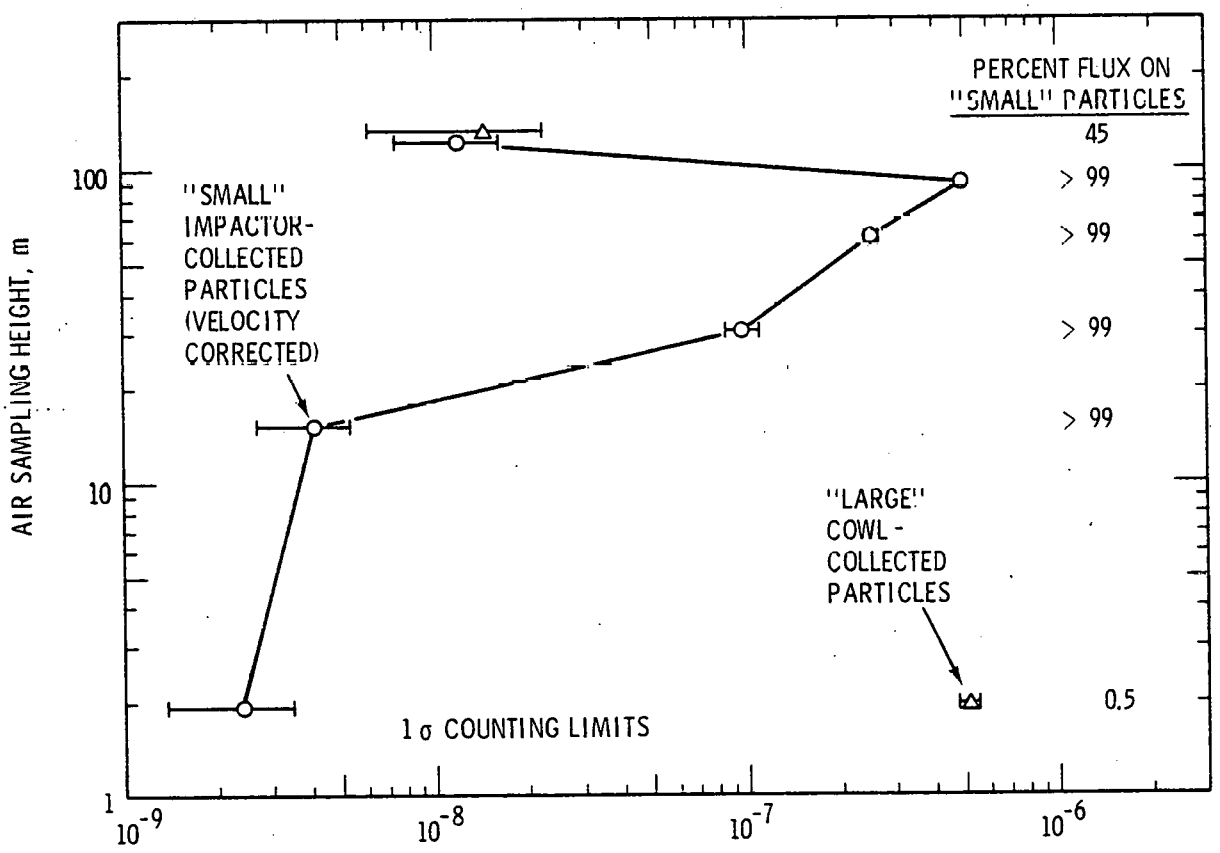
Average horizontal fluxes in $\mu Ci/(m^2$ day) were calculated for both small and large particles. Average horizontal plutonium-239 fluxes are shown in Fig. 6. For large particles, the maximum flux was at a sampling height of 60 m. This maximum flux reflects in part the larger $\mu Ci/g$ at this height. The flux on small particles was less than on large particles. On the right side of the figure is shown the percent of the airborne flux on small particles. The plutonium-239 flux on small particles ranged from 32 to 46% of the total calculated flux.

Average horizontal fluxes for americium-241 are shown in Fig. 7. For transport on small particles, the flux was maximum at $5 \times 10^{-7}$ $\mu Ci/(m^2$ day) at an elevation of 91 m. The flux decreased to $1.2 \times 10^{-8}$ $\mu Ci/(m^2$ day) at an elevation of 122 m. At this highest sampling elevation, the flux on large particles was $1.4 \times 10^{-8}$ $\mu Ci/(m^2$ day). Thus, the percent of total americium-241 flux on small particles was 45% at an elevation of 122 m. This percent flux on small particles is shown on the right side of the figure. Similarly, the americium-241 flux on large particles was $5 \times 10^{-7}$ $\mu Ci/(m^2$ day) at a sampling height of 1.9 m. At this lowest sampling elevation, the percent of total americium-241 flux on small particles was 0.5%. Americium-241 fluxes on large particles are not shown between 1.9 and 122 m sampling height since the radiochemical results were less than radiochemical counting detection limits.




Neg. 7802057-4

FIGURE 6. Airborne Plutonium-239 Fluxes at the Hanford Meteorological Tower During August 13 to November 12, 1976




Neg. 7802057-3

FIGURE 7. Airborne Americium-241 Fluxes at the Hanford Meteorological Tower During August 13 to November 12, 1976

## CONCLUSIONS

Both plutonium-239 and americium-241 concentrations were measured for sampling heights up to 122 m. Results are reported in $\mu Ci/cm^3$, $\mu Ci/g$ of airborne soil, and average fluxes in $\mu Ci/(m^2\ day)$ for both large and small particles. The small particles are in the respirable diameter range. Since results were determined for heights up to 122 m, these experimental data are the first to show plutonium-239 and americium-241 concentration profiles to those heights. However, even at a sampling height of 122 m, the airborne plutonium-239 and americium-241 plumes were probably not contained.

Some correlation is suggested between americium-241 transport on small particles and plutonium-239 transport on large particles. This apparent correlation needs further validation at other sites and time periods.

Americium-241 has been studied even less than plutonium-239 resuspension. Nevertheless, americium-241 resuspension could become a potential inhalation concern of the future when americium-241 grows in as a daughter of plutonium-241 at aged plutonium resuspension sources. In addition to showing americium-241 transport by resuspension, these data also show plutonium-239 and americium-241 are transported on both small and large particles. Although transport on large particles does not present an immediate inhalation concern, transuranics transported on large particles could present a subsequent inhalation concern. After these large particles are deposited, small particles can be released from the larger host soil particles, (Sehmel, 1978). If transuranics, these released small particles could then be sources for subsequent resuspension in the inhalation diameter range. Experimental data are needed to develop and validate transport models for both respirable and nonrespirable particles as well as plutonium-239 and americium-241 detachment from large particles.

## ACKNOWLEDGMENT

This paper is based on work performed under U.S. Department of Energy Contract EY-76-C-06-1830.

## REFERENCES


1. ICRP, International Commission on Radiological Protection. 1959. Report of Committee II on Permissible Dose for Internal Radiation. ICRP Publication 2, Recommendations of the International Commission on Radiological Protection, Pergamon Press, New York, pp. 82-83.

2. Krey, P. W., R. Knuth, T. Tamura, and L. Toonkel. 1976. "Interrelations of Surface Air Concentrations and Soil Characteristics at Rocky Flats." In: Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants. R. J. Engelmann and G. A. Sehmel, Coordinators, CONF-740921, National Technical Information Service, Springfield, VA 22151, pp. 744-756.

3. Sehmel, G. A. 1973. "An Evaluation of a High-Volume Cascade Particle Impactor System." Proc. of the Second Joint Conference on Sensing of Environmental Pollutants. Washington, DC, pp. 109-115, Instrument Society of America, Pittsburgh, PA.

4. Sehmel, G. A., and F. D. Lloyd. 1976. "Resuspension of Plutonium at Rocky Flats." In: Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants. R. J. Engelmann and G. A. Sehmel, Coordinators, CONF-740921, National Technical Information Service, Springfield, VA 22151, pp. 757-779.

5. Sehmel, G. A. 1977a. "Airborne Plutonium Transport on Nonrespirable Particles," "Weathering of Surfaces as Measured by Airborne Radioactive Particle Concentrations on the Hanford Reservations." Pacific Northwest Laboratory Annual Report for 1976: Atmospheric Sciences, BNWL-2100-3, Pacific Northwest Laboratory, operated by Battelle Memorial Institute, Richland, WA, pp. 65-73, 85-88.

6. Sehmel, G. A. 1977b. "Radioactive Particle Resuspension Research Experiments on the Hanford Reservation," BNWL-2081, Pacific Northwest Laboratory, Operated by Battelle Memorial Institute, Richland, WA.

7. Sehmel, G. A. 1977c. "Transuranic and Tracer Simulant Resuspension." BNWL-SA-6236, Pacific Northwest Laboratory, Operated by Battelle Memorial Institute, Richland, WA.

8. Sehmel, G. A. 1978. "Plutonium Concentrations in Airborne Soil at Rocky Flats and Hanford Determined During Resuspension Experiments." BNWL-SA-6720, Pacific Northwest Laboratory, Operated by Battelle Memorial Institute, Richland, WA.

9. Willeke, W. 1975. "Performance of the Slotted Impactor." Amer. Ind. Hyg. Assoc. J., 36:683-691.