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## TRANSURANIC RESUSPENSION

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## TRANSURANIC RESUSPENSION

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Characteristics of aged resuspension sources are more uncertain than those of new resuspension sources, which can be investigated using inert-particle controlled-tracer sources. Even though airborne concentrations are low, one aged uniform-area source which can be used for resuspension studies is the accumulated radionuclide fallout in the soil from stratospheric and tropospheric fallout debris. Airborne radionuclide concentrations from this source were investigated at convenient locations on the Hanford site. The objective of this chapter is to summarize plutonium and americium resuspension research conducted by the Pacific Northwest Laboratory from 1977 to 1983.

Airborne plutonium was determined at five sites in the Hanford area, and both plutonium and americium were determined at two Hanford sites. Airborne plutonium and americium were examined as a function of aerodynamic particle diameter, sampling height, wind speed increments, and wind direction increments. The following results are discussed: airborne radionuclide concentrations,  $\mu\text{Ci}/\text{cm}^3$  of sampled air; radionuclide activity densities,  $\mu\text{Ci}/\text{g}$  of airborne solids; airborne plutonium fluxes,  $\mu\text{Ci}/(\text{m}^2 \text{ day})$ ;  $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratios,  $(\mu\text{Ci } ^{241}\text{Am})/(\mu\text{Ci } ^{239+240}\text{Pu})$ ; and airborne solid concentrations,  $\mu\text{g}/\text{m}^3$  of sampled air.

In addition, a relationship based on field data for aged plutonium sources at Bikini Atoll, the Hanford site, and Rocky Flats was developed to estimate the maximum expected plutonium activity density on airborne solids compared to activity densities for bulk surface-soil samples. As a result, it is possible to more accurately predict resuspension factor ranges as a function of the resuspension source activity densities.

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## TRANSURANIC RESUSPENSION

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Introduction

Resuspension is defined as the introduction of surface contamination into the air by wind stresses and human activities. Resuspension is difficult to predict accurately because a number of factors that affect resuspension are not well defined. Pacific Northwest Laboratory is conducting research on plutonium resuspension for the Department of Energy to develop better methods for predicting resuspension that can be used for environmental assessments. Accurate predictions of airborne concentrations of resuspended contaminants from aged sources are important in predicting the long-term effects of contamination. In 1980, a review of the literature (Sehmel 1980a) indicated that methods for predicting resuspension needed to be improved. Predictive capabilities are limited by the need for more data on parameters such as the weathering rates of airborne concentrations, the effects of wind stresses, and resuspension coefficients.

Weathering half-lives, which may be one of the controlling parameters in environmental assessments, were originally assumed to be approximately constant, but are now considered to change with time. Reported weathering half-lives of radionuclides range from 35 days to years. The half-life depends on the frequency of high wind speeds, surface properties such as canopy cover, and particle properties such as size and water solubility.

Another problem in predicting resuspension is that resuspension coefficients from early resuspension experiments are not generic. Results are often applicable only to selected experimental conditions. Commonly used coefficients are resuspension factors,  $m^{-1}$ , and resuspension rates,  $s^{-1}$ . The resuspension factor is defined as the airborne concentration of resuspended contaminant divided by the local surface-contamination-per-unit area. The resuspension rate is defined as the fraction of contamination resuspended per second. Resuspension factors are used for predicting airborne concentrations above a contaminated surface, and resuspension rates are used as source terms for predicting concentrations downwind from meteorological transport and diffusion models.

The resuspension factor describes neither vertical fluxes from the resuspension source nor downwind transport fluxes. The review indicates that resuspension factors range over nine orders of magnitude, from  $10^{-10}$  to over  $10^{-2} \text{ m}^{-1}$ . Furthermore, resuspension factors have ranged two to three orders of magnitude even in a single field experiment.

The resuspension rate describes the contamination flux from local surface contamination. The literature review indicated that resuspension rates ranged over eight orders of magnitude, from  $10^{-12}$  to  $10^{-4}$  fraction/s. If the source is known, the vertical flux from resuspension is the product of the resuspension rate and surface-contamination-per-unit area. Resuspension rates measured for both wind stresses and human activities indicate that human activities can resuspend more contamination than average winds; however, the relative increase is short-lived because human activity stresses usually continue over less time and space.

Resuspension of contamination by wind stresses is difficult to predict because wind patterns and source distributions are not well known. Prior results obtained by Sehmel (1980c) at Hanford and Rocky Flats between 1971 and 1977 were summarized in the first volume of this series (Hanson 1980). Results showed that airborne concentrations of plutonium ( $\mu\text{Ci}/\text{cm}^3$ ) increased with wind speed and enrichment in the activity densities ( $\mu\text{Ci}/\text{g}$ ) as a function of airborne particle size. Activity densities for  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$  were greater on respirable airborne particles compared to larger host particles. Continuing research is improving our understanding of plutonium resuspension at several sites, such as the Hanford site (Sehmel 1983b), Rocky Flats (Hodgin 1983a, 1983b and Langer 1983), and Savannah River (Shinn, Homan and Gay 1983). Accurate predictions of airborne concentrations from aged-plutonium resuspension sources are important in developing accurate environmental assessments that include airborne effects from plutonium resuspension.

The characteristics of resuspension from aged sources are more difficult to predict than resuspension from new sources, which can be investigated using newer inert-particle, controlled-tracer sources (Sehmel 1983a). One aged uniform-area source is the accumulation of radionuclides in the soil from stratospheric and tropospheric fallout debris. Airborne concentrations of plutonium were investigated at convenient operation sites to determine the resuspension characteristics of an aged uniform-area source.

This chapter summarizes plutonium resuspension research conducted by the Pacific Northwest Laboratory from 1977 to 1983. This research compared the similarity of plutonium resuspension to tracer resuspension and evaluated important parameters at tracer sources that need to be investigated in more detail (i.e., to determine if resuspension of fallout debris was reflected by airborne plutonium concentrations increasing with greater wind speed and to determine plutonium distributions as a function of the aerodynamic particle diameters of airborne host-soil particles on which the plutonium was transported).

In addition, a relationship based on field data for aged plutonium sources was developed to predict the maximum expected plutonium activity density on airborne solids compared to activity densities for bulk surface-soil samples. As a result, the resuspension factor ranges as a function of the resuspension-source activity densities can be predicted more accurately.

#### Investigations of Transuranic Resuspension

Experimental results reported here for transuranic resuspension and the relationships developed have been discussed in more detail in the following publications:

- Airborne plutonium concentrations (Sehmel 1978b, 1978c, 1979b, 1979c, 1980b, 1981a, 1981b, 1981c, 1981d, 1983b)
- Airborne americium concentrations (Sehmel 1979b, 1981c)
- Airborne plutonium activity densities (Sehmel 1978b, 1978c, 1979b, 1979c, 1980b, 1981a, 1981b, 1981c, 1981d, 1982a, 1982c, 1983b)
- Activity density enrichments for airborne americium (Sehmel 1979b, 1981c, 1982c, 1982d)
- Plutonium activity densities on airborne and surface soils (Sehmel 1982a)
- Resuspension factors for aged plutonium sources (Sehmel 1982b).

The resuspension of airborne plutonium from aged uniform-area sources was evaluated at five sampling sites on the Hanford site. The locations of the five sampling sites are shown in Fig. 1. All sites are in relatively flat terrain except site 3, which is on the highest mountain within the local area. The elevation of sampling site 3 is 1075 m, which is approximately 875 m above the 200-m elevation of the Hanford site. At site 5, the 125-m Hanford Meteorological Station tower was used as a sampling platform.

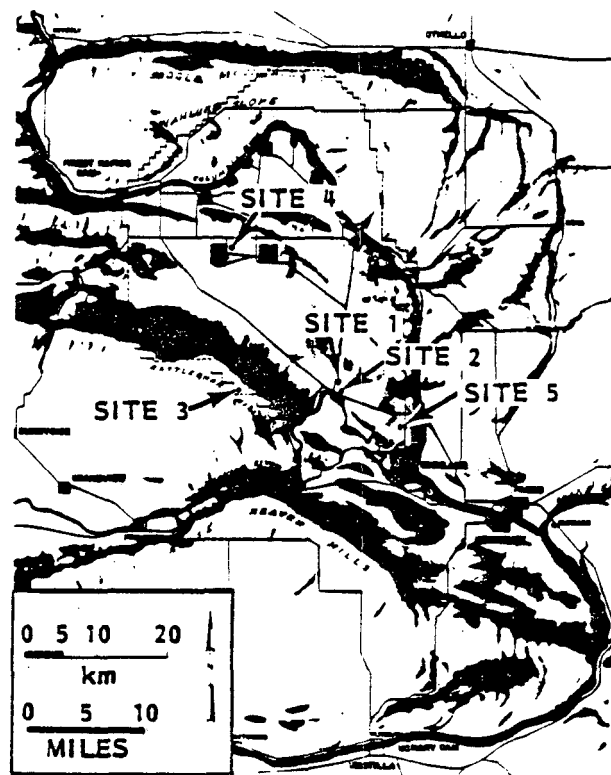


Fig. 1. Map of the Hanford Site showing sampling-site locations.

Airborne plutonium was determined at all five sites, and americium was determined at two sites. Airborne plutonium and americium concentrations and activity densities were determined as functions of aerodynamic particle diameter, sampling height, wind speed increments, and wind direction increments.

In general, samples were collected as a function of aerodynamic particle diameters of respirable and inhalation interest. In some experiments, samples were collected at representative heights where respiration could occur; in other experiments, samples were collected to determine concentration changes as a function of height. The airborne particulate samplers were activated usually as a function of wind direction to maximize the potential for evaluating airborne concentration dependencies on wind speed. Also, samples were taken at selected wind directions to partially control the effects of upwind surface roughness and vegetative canopy. Because resuspension is expected to increase with higher wind speed, the direction was selected for the greatest frequency of high wind speeds. In these experiments, the frequency of high winds was greatest from the southwest ( $225^\circ$ ), so most samples were collected only during southwesterly winds. Also, because wind resuspension rates were



expected to increase with increasing wind speed, above a threshold value, some samples were collected as a function of wind speed increments.

Sampling parameters for each sampling site are summarized in Table 1. Sampling heights ranged from 0.3 to 124 m above ground. Samples were usually collected by automatic sampling with multiple samplers during selected increments of wind speed and direction.

Except at site 2, all samplers at each site were located horizontally within a few meters of each other. At site 2, ambient airborne concentrations were determined using two sampling towers 305 m apart. When wind speed and direction were identical at each tower (i.e., a wind direction of  $233^\circ \pm 45^\circ$  and a wind speed increment of 5 to 7 m/s measured at an elevation of 30 m), airborne solids were collected on filters that were subsequently analyzed for plutonium.

#### Automatic Controllers

Automatic sampler controllers responding to selected wind speed and wind direction criteria were usually used to activate samplers for airborne particulates. Otherwise, samplers were manually activated to sample continuously for all wind speeds and directions. Samplers were turned off during precipitation.

When wind speed and direction criteria were satisfied, the controllers automatically activated designated particulate samplers. The controllers usually were set to activate at wind directions of about  $225^\circ$  (i.e., the wind direction for highest frequency and duration of high wind speeds). Three wind speed increments were selected to determine how airborne concentrations depend on wind speed. Airborne solids were sampled during seven wind speed increments to determine threshold wind speeds for resuspension.

#### Sampling Method Number 1

Airborne particles were sampled with particle cowl cascade-impactor systems (Sehmel 1973) shown in Fig. 2. The airflow rate was  $0.57 \text{ m}^3/\text{min}$  (20 cfm). The particle cascade impactor\* for sampling respirable-size particles was attached to a rotating cowl, allowing simultaneous sampling of larger nonrespirable particles. Particles entering the 15-cm-dia cylindrical sampler inlet of the cowl either settled on the cowl floor or were drawn up into the impactor. In this report, particles settling within the cowl are called "large" cowl-collected particles. The sum of all particles collected

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\*Model 65-100 High-Volume Sampler Head, Anderson 2000, Inc., Atlanta, GA 30336

TABLE 1 Sampling Parameters for Airborne Plutonium

Site Number	Sampling Period	Sampling Procedures				
		Wind Increments Sampled		Airborne Particles Analyzed for Plutonium Content		
		Direction	Speed Ranges, m/s	Particle Sampling Heights, m	Sampling Procedure by Method Number	Host-Particle Diameter 50%-Stage-Efficiency, $\mu$ m
1	12 April to 29 June 1976	225° ±35°	3-5	0.3	Number 1,	less-than-1.1
			5-7	2.0	4-stage	1.1 and 2.0
			7-11	5.8	impactor	3.3 and 7
	12 Aug. 1976 to 11 Jan. 1977	225° ±35°	3-5	0.3	Number 1,	less-than-1.1
			5-7	2.0	4-stage	1.1 and 2.0
			7-11	5.8	impactor	3.3 and 7
	17 Sept. to 8 Nov. 1979	240° ±15° 240° ±30° 240° ±45°	All	1.4	Number 2,	5.5
					Collec-	diameter
					tion after cyclone preseparator,	50%-collection-efficiency
2	21 July to 20 Nov. 1978	233° ±45°	5-7	0.3	Number 3,	
				1.0	Filter	
				2.0	with	
				3.0	isoki-	
				5.0	netic	
	11 May to 2 July 1979			9.0	sampler	
				20.0	inlet	
					Number 4,	2.1-stage
					2-stage,	plus
					impactor	less-than-2.1 backup
3	27 May to 2 June 1978	240° ±35°	0.5-3	1.7	Number 2,	5.5
			3 -5		Collec-	diameter
			5 -7		tion	50%-
			7 -11		after	collection-
			11 -15		cyclone	efficiency
			15 -17.5		presepar-	
			17.5-31		ator	

TABLE 1 (continued) Sampling Parameters for Airborne Plutonium

Sampling Procedures						
Site Number	Sampling Period	Wind Increments Sampled		Airborne Particles Analyzed for Plutonium Content		
		Direction	Speed Ranges, m/s	Particle Sampling Heights, m	Sampling Procedure by Method Number	Host-Particle Diameter 50%-Stage-Efficiency, $\mu$ m
3	30 June to 17 Nov. 1978	240° ±35°	0.5-3	1.7	Number 2, Collection after cyclone preseparator	5.5 diameter 50%-collection-efficiency
			3 -5			
			5 -7			
	11 May to 2 July 1979		7 -11		Number 4, 2-stage impactor	2.1-stage plus less-than-2.1 backup
			11 -15			
			15 -17.5			
	25 July to 8 Nov. 1979		17.5-31		Number 4, 2-stage impactor	2.1-stage plus less-than-2.1 backup
4	12 April to 25 July 1979	225° ±35°	All	1.9		
				17		
				63		
				124		
		45° ±145°	All	1.4		
				62		
				124		
5	25 July to 8 Nov. 1979				Number 4, Two-stage impactor	2.1-stage plus less-than-2.1 backup

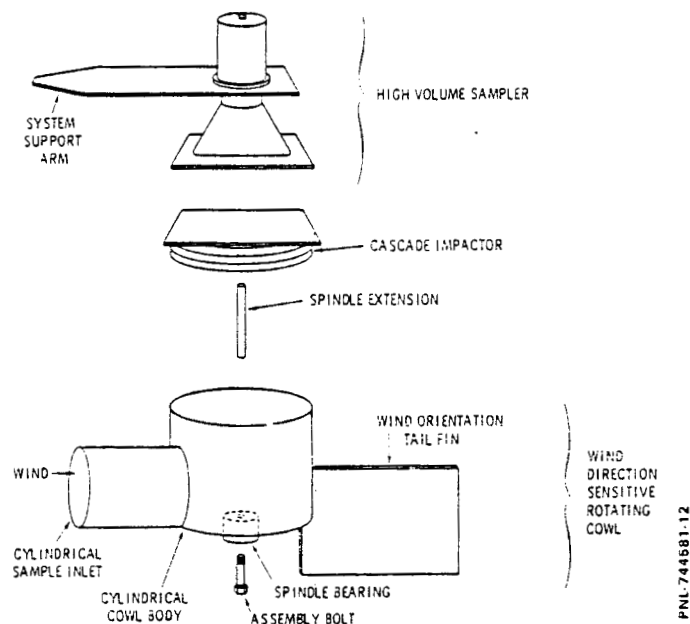


Fig. 2 Sampling system: a wind direction-oriented cowl for collecting "large" particles by gravity settling and a particle cascade impactor for collecting the remaining "small" particles.

on the cascade impactor collection surfaces plus backup filter are called "small" particles. Small particles entering the particle cascade impactor were separated into nominal aerodynamic diameter ranges of 7, 3.3, 2.0 and 1.1  $\mu\text{m}$ , which are impactor-stage, 50%-collection-efficiency diameters for unit density spheres. For some experiments, these systems were operated without impactor stages. In these cases, airborne particles were collected either in the cowl or on a filter attached to the air pump.

#### Sampling Method Number 2

Airborne solids were sampled with a modified cyclone preseparator followed by a cascade particle impactor\*. The airflow rate was 1.1  $\text{m}^3/\text{min}$  (40 cfm). The cyclone preseparator was modified by attaching an inlet closure on the preseparator. The closure was opened with a solenoid when the air pump was activated by the wind speed and direction controller. The 50%-collection-efficiency diameter for the cyclone preseparator was 5.5  $\mu\text{m}$ , and for the last stage of the cascade impactor, it was 0.5  $\mu\text{m}$ .

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\*Model 230CP Cyclone Preseparator, Sierra Instrument Co., Carmel Valley, CA 93924

### Sampling Method Number 3

Airborne solids were sampled with an isokinetic sampling inlet (Sehmel 1978a, 1979a) preceding a filter. The airflow rate was  $1.1 \text{ m}^3/\text{min}$  (40 cfm). The air-sampler inlet was attached to a standard 20 x 25-cm (8 x 10-in.) filter holder. In Fig. 3, a cutaway view of the assembled inlet is shown with the inlet attached. The inlet is 25 cm wide, and its height is adjusted to isokinetic flow for the average wind speed increment sampled. The inlet cross-sectional area is held constant for the first 7.5 cm toward the filter, after which the top of the inlet (16 ga aluminum) is angled upward to fit over the filter holder. The total distance between inlet and filter is 25 cm.

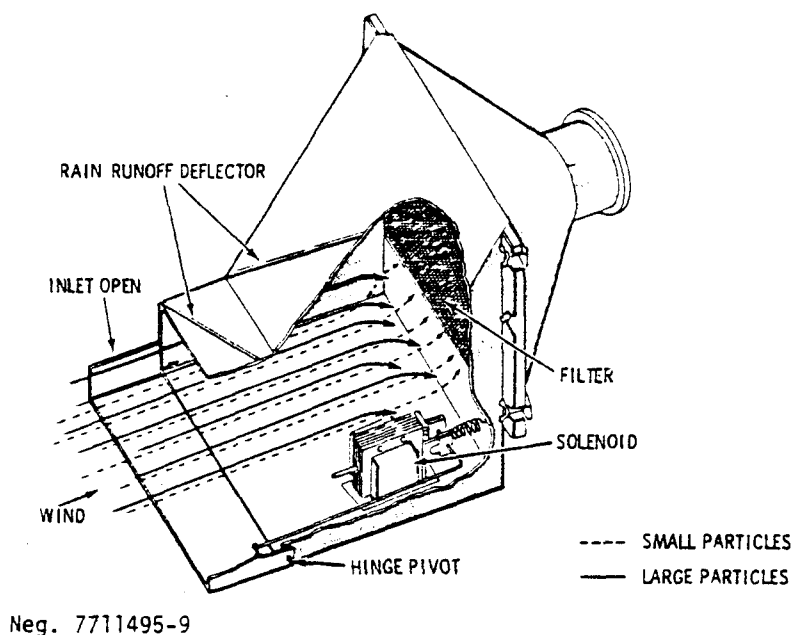


Fig. 3. Cutaway view of the isokinetic sampler -- inlet open.

A solenoid-operated closure uncovers the inlet when the air sampler is activated. The cover is shown opened (see Fig. 3). Also shown schematically are paths for "large" and "small" particles. In this case, large refers to those particles that settle on the bottom of the inlet, and small refers to those particles that collect on the filter.

### Sampling Method Number 4

Airborne solids were collected for all wind speeds and directions using massive-volume air samplers (Mitchell, Henry and Henderson 1978). The airflow rate was  $11 \text{ m}^3/\text{min}$  (390 cfm). These samplers have two cascade-impactor stages

followed by an electrostatic precipitator. The first impactor stage was coated with silicone grease to minimize particle bounce-through. The 50%-collection-efficiency diameter is about 4  $\mu\text{m}$  for the first impactor stage and is calculated to be 2.1  $\mu\text{m}$  for the second impactor stage. Particles collected on the electrostatic precipitator plates were removed for plutonium analysis.

#### Radiochemistry

After sample collection, samples were equilibrated with low-humidity air and weighed. After weighing, samples were analyzed for  $^{239+240}\text{Pu}$  or  $^{241}\text{Am}$  content by a commercial laboratory\*. Plutonium samples were completely dissolved and then electrodeposited onto platinum discs for alpha energy analysis.

#### Results

The maximum airborne concentration of resuspended plutonium is low, even for the highest wind speeds investigated. Average airborne concentrations would be even less during continuous sampling. The maximum concentration shown ( $2 \times 10^{-16} \mu\text{Ci}/\text{cm}^3$  in Fig. 4 for site 1) is only 0.3% of the 168-hr, maximum-permissible-concentration guide (ERDA 1977), which is  $6 \times 10^{-14} \mu\text{Ci}/\text{cm}^3$ .

Plutonium-239,-240 content was evaluated for all samples reported here. In addition, americium contents were determined for some samples. The following results are discussed:

- airborne radionuclide concentrations,  $\mu\text{Ci}/\text{cm}^3$  of sampled air
- radionuclide activity densities,  $\mu\text{Ci}/\text{g}$  of airborne solids
- airborne  $^{239+240}\text{Pu}$  fluxes,  $\mu\text{Ci}/(\text{m}^2 \text{ day})$
- $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratios,  $(\mu\text{Ci } ^{241}\text{Am})/(\mu\text{Ci } ^{239}\text{Pu})$
- airborne solid concentrations,  $\mu\text{g}/\text{m}^3$  of sampled air.

Because of the small radionuclide contents in each sample, uncertainties from non-isokinetic sampling and weighing errors are less than radiochemical-counting statistic uncertainties. In general, radiochemical uncertainties and wind speed increments sampled are indicated for each data point in the following figures. Wind speed increments are shown by horizontal lines, and concentrations corresponding to the one-sigma limits of radiochemical-counting statistics are shown by vertical lines through each data point. However, if radiochemical detection limits include zero, a data point is shown at the upper limit with an arrow pointing toward zero.

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\*LFE Environmental Analysis Laboratory, 230 Wright Ave., Richmond, CA 94804

### Airborne Plutonium at Site 1

Plutonium transport during southwesterly winds was determined at site 1 for two time periods in 1976 to 1977 (Sehmel 1978c, 1980b).

Site 1 Concentrations from April 12 to June 29, 1976. Airborne  $^{239+240}\text{Pu}$  concentrations on small particles and airborne  $^{239+240}\text{Pu}$  fluxes on large particles were determined (see Fig. 4 and Table 2). Data are shown in Fig. 4 for plutonium collection on the 3.3 plus 7- $\mu\text{m}$  50%-collection-efficiency impactor stages, 1.1 plus 2- $\mu\text{m}$  50%-collection-efficiency impactor stages, and the backup filter. Airborne  $^{239+240}\text{Pu}$  concentrations increased about two orders of magnitude, from about  $10^{-18}$  to  $10^{-16}$   $\mu\text{Ci}/\text{m}^3$ . When a power law dependency is assumed,  $U^n$ , the wind speed dependency is shown as an exponent on the wind speed velocity,  $U$ , measured at the 1.5-m height. Airborne  $^{239+240}\text{Pu}$  concentrations increased as powers of wind speed,  $U$ :  $U^{0.8}$  to  $U^{7.8}$ .

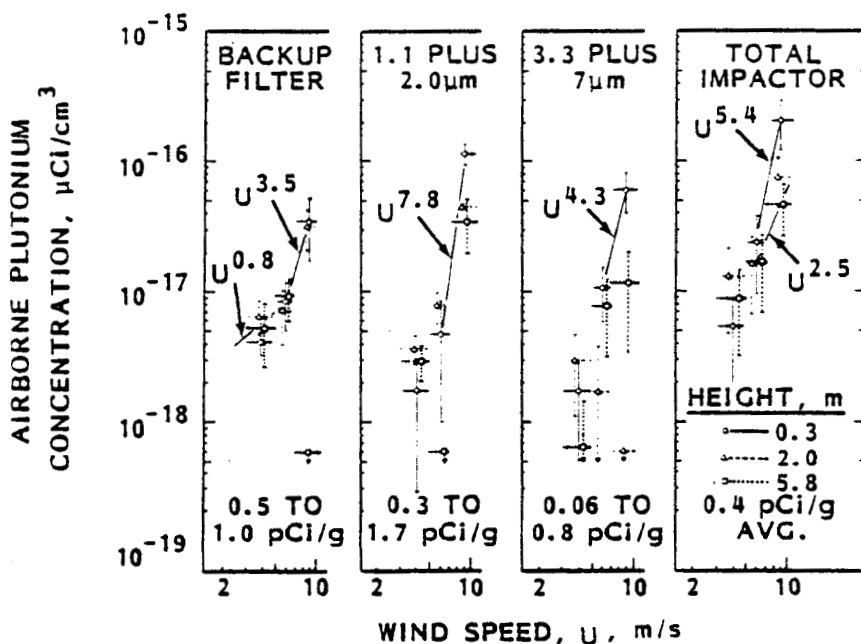


Fig. 4. Airborne  $^{239+240}\text{Pu}$  concentrations at site 1 as a function of wind speed during April 12 to June 29, 1976. Winds were sampled from 190 to 260° and measured at the 1.5-m height.

Horizontal plutonium flux calculations were made for the total time wind speed was between 3 and 11 m/s (and wind direction was between 190 to 260°), and for the total time the cowl samplers were in the field. When the shorter time period (3 to 11 m/s winds) was used for calculating horizontal plutonium fluxes, fluxes ranged from  $3.9 \times 10^{-6}$  to  $1.4 \times 10^{-6}$   $\mu\text{Ci}/(\text{m}^2 \text{ day})$ . Fluxes using the total time were smaller.

Activity densities of  $^{239+240}\text{Pu}$  per gram of airborne solids were determined. For small particles, activity densities ranged from  $6.0 \times 10^{-8}$  to  $1.7 \times 10^{-6} \mu\text{Ci/g}$  with an average of  $4.0 \times 10^{-7} \mu\text{Ci/g}$ . For large particles, activity densities ranged from  $1.3 \times 10^{-7}$  to  $2.1 \times 10^{-7} \mu\text{Ci/g}$ .

Site 1 Concentrations from August 12, 1967 to January 11, 1977. Airborne plutonium concentrations and concentrations of airborne particles are shown in Fig. 5. Data are shown separately for each sampling height of 0.3, 2.0, and 5.8 m. Airborne concentrations ranged from  $4 \times 10^{-18}$  up to a maximum of  $2 \times 10^{-16} \mu\text{Ci/cm}^3$ . At the lowest sampling height of 0.3 m, airborne  $^{239+240}\text{Pu}$  concentrations were nearly constant at about  $5 \times 10^{-17} \mu\text{Ci/m}^3$  for all wind speeds. Although plutonium concentration remained constant, the bottom portion of the figure shows that when a power law dependency is assumed,  $U^n$ , the airborne mass-loading dependency is  $U^{2.1}$ .

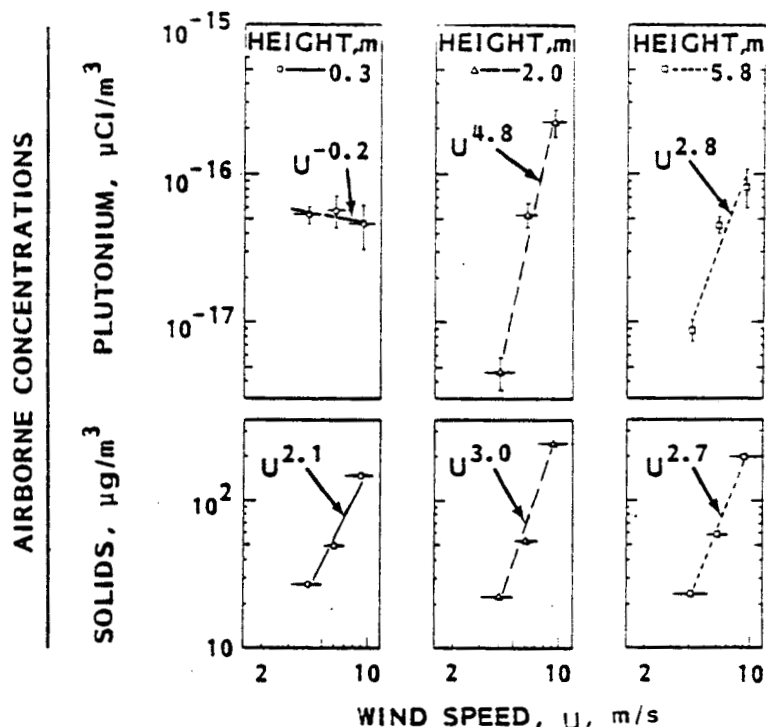


Fig. 5. Airborne  $^{239+240}\text{Pu}$  concentrations and soil loadings collected at site 1 as a function of wind speed for total samples within particle cascade impactors and on backup filters during August 12, 1976, to January 11, 1977. Winds were sampled from 190 to 260° and measured at the 1.5-m height.



In contrast to concentrations at the 0.3-m height, plutonium concentrations at the 2.0 and 5.8-m heights increased with wind speed. Airborne concentrations for the 2.0-m sampling height are shown in the central section of Fig. 5. Airborne plutonium concentrations were  $4.6 \times 10^{-18} \text{ } \mu\text{Ci/cm}^3$  for the 3 to 5 m/s wind speed increment. At this lowest wind speed increment, the airborne plutonium concentration at the 2-m height was one order of magnitude less than plutonium concentrations at the lower height of 0.3 m for all wind speeds. As a possible explanation of the data, a decrease in airborne plutonium concentration with increasing height would be expected for a local resuspension source superimposed on concentrations from a uniform source.

In contrast, for the highest wind speed increment of 7 to 11 m/s, the airborne plutonium concentration at 2-m height is about four times greater than that at the 0.3-m height. This increased airborne plutonium concentration with height indicates that resuspension results during high winds are dominated by upwind source(s).

Airborne plutonium concentrations increased with about the third to fifth power of wind speed. At 2.0 m, airborne plutonium concentrations increased with the 4.8 power of wind speed, while the airborne solids concentration increased with the 3.0 power of wind speed. For the highest point sampled (5.8 m), both airborne plutonium and solids concentrations increased with the wind speed to the 2.7 to 2.8 power.

Plutonium activity densities for airborne solids are shown in Fig. 6 at each sampling height as a function of wind speed. For the lowest sampling elevation level of 0.3 m, activity densities on airborne solids decreased with wind speed to the negative 2.3 power. The underlying cause of this decrease in activity density with increasing wind speed is unknown. A possible explanation could be that either locally uncontaminated soil was resuspended at higher wind speeds, or that uncontaminated soils were transported from upwind. In contrast, at a sampling height of 2.0 m, plutonium activity densities on airborne solids increased with the 1.8 power of wind speed.

A third activity pattern is shown by the data from the highest sampling level of 5.8 m. For this height, activity densities were nearly constant for the lowest and highest wind speed increments, with a maximum activity density occurring at the middle wind speed increment. The reason for this maximum is unknown. However, a maximum might have occurred also for the intermediate wind speed at the 2.0-m height.

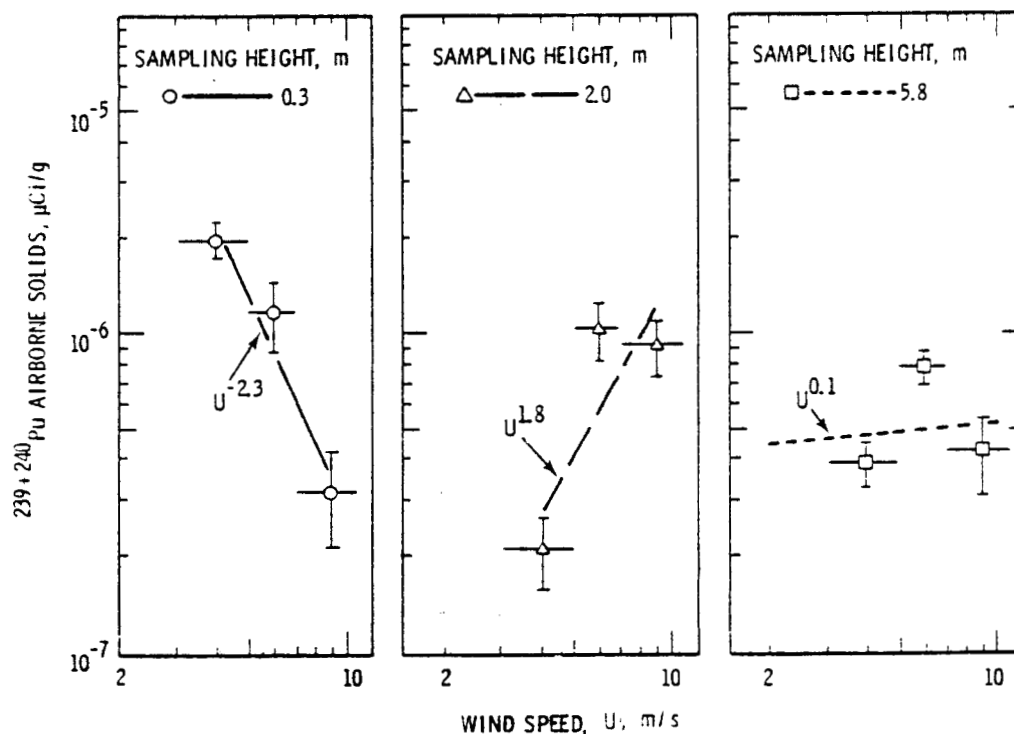


Fig. 6. Plutonium-239,-240 activity densities on airborne soil as a function of wind speed for total samples collected within particle cascade impactors and on backup filters from August 12, 1976, to January 11, 1977. Winds were sampled from 190 to 260° and measured at the 1.5-m height.

For large cowl-collected particles,  $^{239+240}\text{Pu}$  activity densities and fluxes are shown in Table 2. Activity densities for small particles ( $0.6 \times 10^{-7}$  to  $20 \times 10^{-7} \mu\text{Ci/g}$ ) were up to an order of magnitude greater than for large particles ( $1.0 \times 10^{-7}$  to  $2.1 \times 10^{-7} \mu\text{Ci/g}$ ).

TABLE 2 Plutonium Transport on Large Particles at Site 1

Sampling Period	Sampling Height, m	$^{239+240}\text{Pu}$ on Airborne Solids		Airborne $^{239+240}\text{Pu}$ Horizontal Flux on Large Particles, $\mu\text{Ci}/(\text{m}^2 \text{ day})$	For Total Field Time
		dpm/g	$\mu\text{Ci/g}$	For 190 to 260° Winds Only and Wind Speeds from 3 to 11 m/s	
April 12	0.3	0.29	$1.3 \times 10^{-7}$	$3.9 \times 10^{-6}$	$6.0 \times 10^{-7}$
to	2	0.46	$2.1 \times 10^{-7}$	$4.0 \times 10^{-6}$	$8.3 \times 10^{-7}$
June 29, 1976	5.8	0.32	$1.5 \times 10^{-7}$	$1.4 \times 10^{-6}$	$2.8 \times 10^{-7}$
August 12, 1976	0.1	0.22	$1.0 \times 10^{-7}$	$1.56 \times 10^{-7}$	$1.30 \times 10^{-8}$
to	2	0.23	$1.1 \times 10^{-7}$	$1.89 \times 10^{-7}$	$1.57 \times 10^{-8}$
June 11, 1977	5.8	0.39	$1.8 \times 10^{-7}$	$3.38 \times 10^{-7}$	$2.82 \times 10^{-8}$

Site 1 Horizontal Fluxes. Plutonium transport fluxes on large cowl-collected particles were not directly measured, since the cowl inlet was open and large particles could be trapped within the cowl for all wind directions from about 190 and 260°. The direct measurement would have required closure of the cowl inlet during nonsampling periods. Nevertheless, an estimate was made for the plutonium transport flux on large particles.

Airborne plutonium fluxes on large particles were calculated for two time periods. Horizontal-flux calculations were made both for the total time that winds were simultaneously between 3 and 11 m/s (and with wind direction between 190 and 260°), and for the total time that cowl air samplers were in the field. When the shorter period (for 3 to 11 m/s and 190 to 260° winds) was used, as shown in Table 2, horizontal plutonium fluxes ranged from  $1.6 \times 10^{-7}$  to  $3.3 \times 10^{-7}$   $\mu\text{Ci}/(\text{m}^2 \text{ day})$ .

#### Airborne Plutonium at Site 2

Airborne plutonium and solids concentrations were determined at site 2 (Sehmel 1981a) at two sampling-tower sites separated by a distance of 305 m. On each tower, three sets of isokinetic air samplers (see Fig. 3) were attached at six sampling heights from 0.3 to 20 m above ground. Samples collected using one set of samplers were analyzed for plutonium. When the wind speed (between 5 to 7 m/s measured at an elevation of 30 m) and wind direction were identical at each tower (a wind direction of  $233^\circ \pm 45^\circ$ ), the isokinetic air samplers were automatically activated at both upwind and downwind towers.

Airborne plutonium concentrations, activity densities, and airborne solid concentrations are shown in Fig. 7 for filter collection, as well as total solids including filter plus inlet collection. Upwind and downwind results are shown by solid and dashed lines, respectively. As shown in Fig. 7a, airborne-solids concentrations ( $\mu\text{g}/\text{m}^3$ ) for both filter collection and total solids collected decreased with downwind distance. As shown in Fig. 7b, plutonium activity densities for filter-collected particles at 2- and 3-m heights of the upwind sampling tower were approximately three times greater than for any other sample at the upwind tower. Airborne plutonium concentrations are shown in Fig. 7c. Again, airborne concentrations at the upwind tower sampling heights of 2 and 3 m are approximately three times greater than for any other height at the upwind tower.

#### Airborne Plutonium and Americium at Site 3

Airborne plutonium and americium were determined during two experiments at site 3 (Sehmel 1979b, 1981c), the mountaintop site. Experiments were conducted from May 27 to June 2 and from June 30 to November 17, 1978.

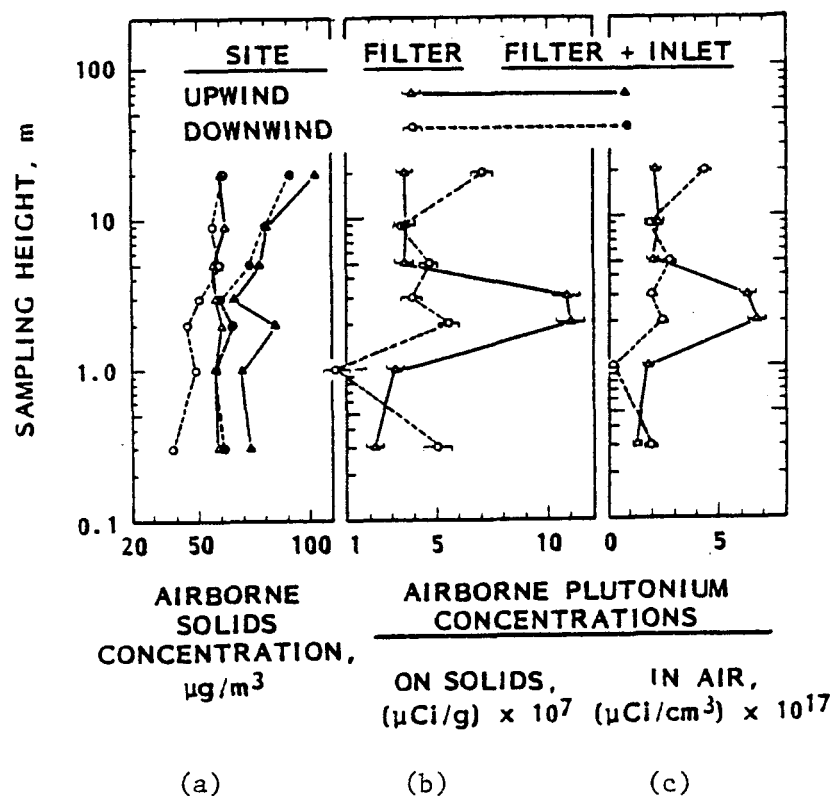


Fig. 7. Average airborne solids and  $^{239+240}\text{Pu}$  concentrations as a function of height at site 2 from July 21 to November 20, 1978, at both an upwind site and a sampling site 305 m downwind along  $233^\circ$ . Winds were sampled during simultaneous wind speed and wind direction increments at both sites. Wind speed increments of 5 to 7 m/s and direction  $233^\circ \pm 45^\circ$  were measured at a height of 30 m.

Airborne  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  Concentrations. Airborne  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  concentrations are shown in Fig. 8 for the first experiment and in Fig. 9 for the second experiment. Shown in the upper and lower portions of each figure are airborne concentrations and activity densities.

In comparing data from these two experiments, results are significantly different. As shown in Fig. 8, for the first experiment, airborne plutonium concentrations were independent of wind speed. In contrast, for the second experiment (shown in Fig. 9), airborne plutonium concentrations were a function of wind speed.

As shown in Fig. 9, airborne plutonium concentrations were a function of wind speed for the second experiment. For five wind-speed-sampling increments between 0.5 and 15 m/s, airborne plutonium concentrations decreased with increasing wind speed. Above a wind speed of 15 m/s, airborne plutonium concentrations increased rapidly with wind speed.

Airborne americium concentrations as a function of wind speed were also different for the two experiments. As shown in Fig. 8, airborne americium

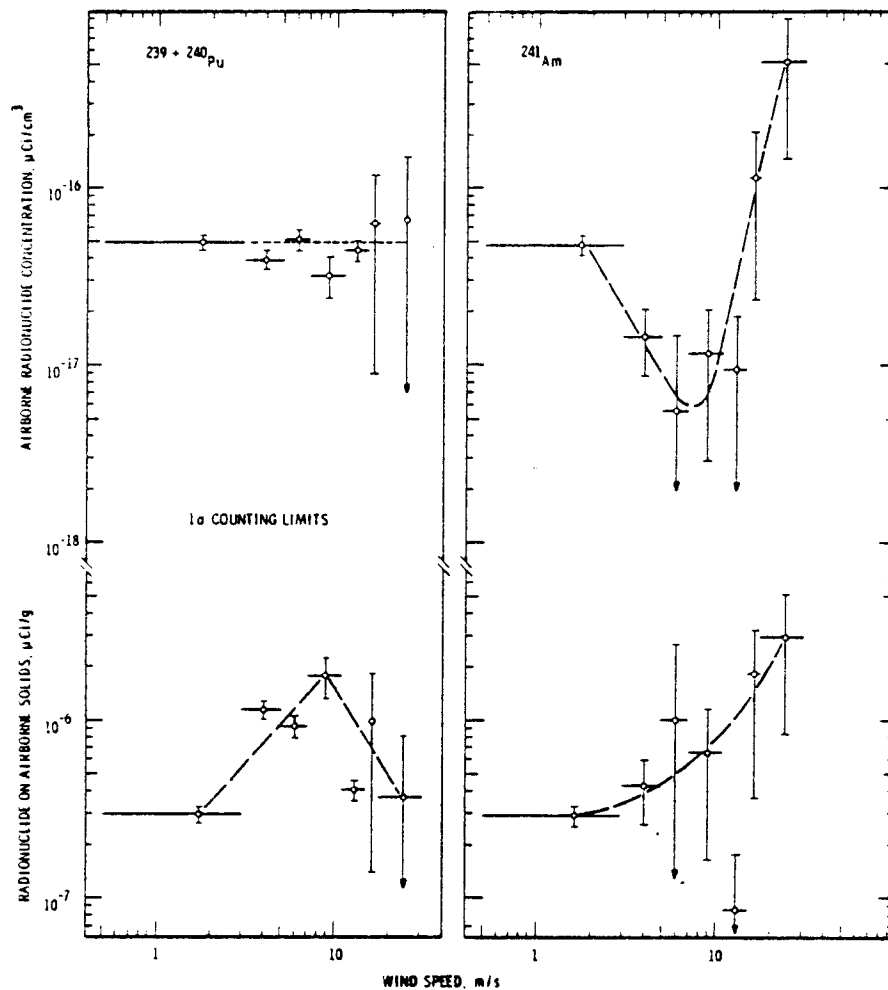


Fig. 8. Airborne  $^{239} + ^{240}\text{Pu}$  and  $^{241}\text{Am}$  concentrations and activity densities on airborne solids collected at site 3 from May 22 to June 22, 1978, when sampling 205 to 275° winds.

concentrations were lowest at an intermediate wind speed of about 7 m/s for the first experiment. For higher wind speeds, concentrations increased with the 4.6 power of wind speed,  $U^{4.6}$ . However as shown in Fig. 9, for the second experiment, americium concentrations were nearly constant and only increased at the highest wind speed increment, 17.5 to 31 m/s.

Activity Densities for Airborne Solids. Plutonium and americium activity densities on airborne solids were calculated from the combined weights of solids collected on impactor stages and backup filter.

For the first sampling period, plutonium activity densities were maximum at an intermediate wind speed increment of 7 to 11 m/s. The reason for this maximum is unknown. However, one postulate is that the relative resuspension

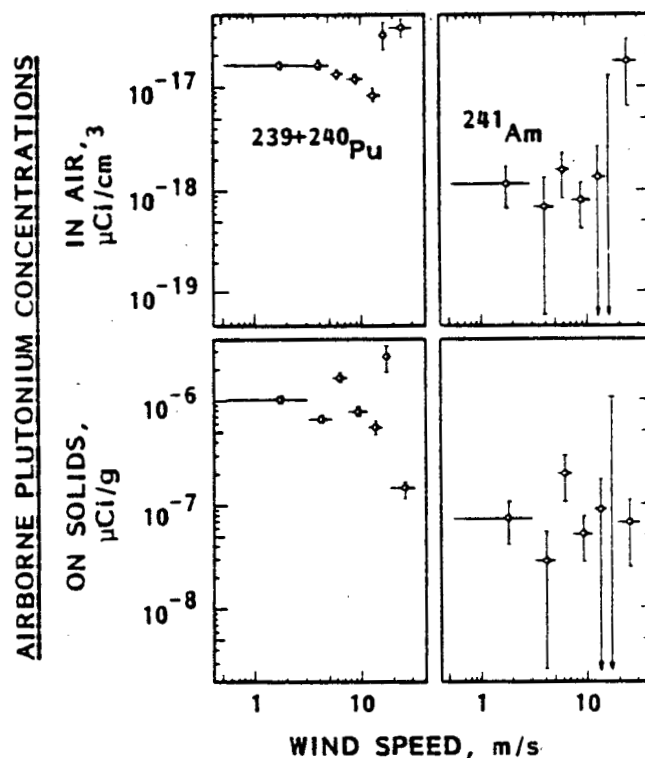


Fig. 9. Airborne  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  concentrations and activity densities on airborne solids collected at site 3 as a function of wind speed during June 30 to November 17, 1978, when sampling  $205$  to  $275^\circ$  winds.

availability of plutonium on surface soils, as compared to the availability of uncontaminated soil, increases as wind speed increases from  $0.5$  up to approximately  $9$  m/s. For higher wind speeds, the relative resuspension availability of uncontaminated surface soils must have increased compared to the availability of plutonium on surface soils. In contrast, during the second experiment as shown in Fig. 9, plutonium activity densities were inconsistent as a function of wind speed.

Americium concentrations on the airborne solids also varied. However, absolute values are less certain than those for plutonium because of radiochemical counting uncertainties. During the first experiment, americium activity densities tended to increase with increasing wind speed. As shown in Fig. 8, only the  $11$  to  $15$  m/s wind speed increment shows a significant deviation from this increase. In contrast, although activity densities for the second experiments showed some variation as a function of wind speed, the variation was a nonuniform function (Fig. 9).

Airborne Solids Concentrations. Airborne solids concentrations were different functions of wind speed for the two experiments. As shown in the upper portion of Fig. 10, for the first experiment airborne solids concentrations ranged from about 18 to 180  $\mu\text{g}/\text{m}^3$ . Unexpectedly, concentrations were the largest, and nearly identical, for both the lowest (0.5 to 3 m/s) and highest (17.5 to 31 m/s) wind speed increments. The minimum concentration was for the 7 to 11 m/s intermediate wind speed increment.

The upper portion of Fig. 11 shows that during the second experiment concentrations ranged from 8 to 20  $\mu\text{g}/\text{m}^3$  for wind speeds below 17.5 m/s. This low concentration reflects concentrations expected in a pristine region. However, for the highest wind speed increment investigated, airborne concentrations dramatically increased to 200  $\mu\text{g}/\text{m}^3$ . For the larger particles collected within the cyclone preseparators, the average concentration was 5.6  $\mu\text{g}/\text{m}^3$ .

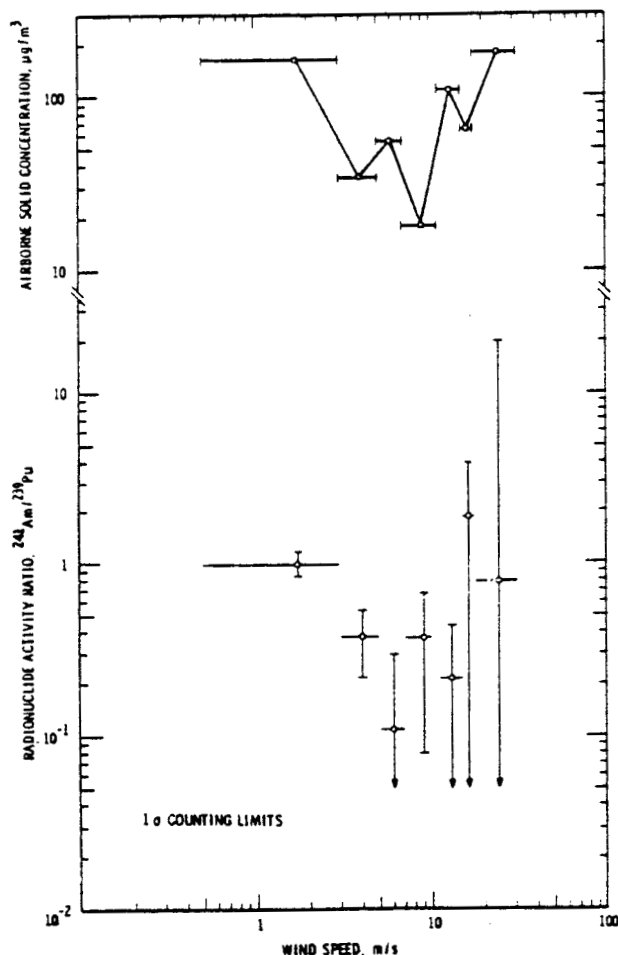


Fig. 10. Airborne solids concentrations and  $^{241}\text{Am}/^{239+240}\text{Pu}$  radionuclide ratios for airborne solids collected at site 3 from May 22 to June 22, 1978, when sampling 205 to 275° winds.

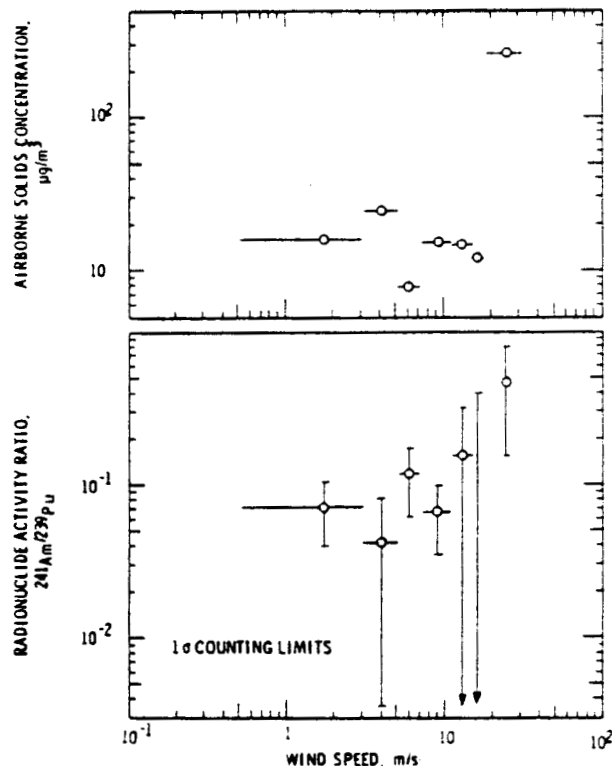


Fig. 11 Airborne solids concentrations and  $^{241}\text{Am}/^{239+240}\text{Pu}$  radionuclide ratio at site 3 for airborne solids collected as a function of wind speed during June 30 to November 17, 1978, when sampling 205 to 275° winds.

Americium-241/Plutonium-239,-240 Activity Ratios. Because airborne plutonium and americium concentrations show different trends as functions of wind speed, the ratios of  $^{241}\text{Am}/^{239+240}\text{Pu}$  concentrations were evaluated. The  $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratio is not constant as a function of wind speed. The ratio ranges from about 0.04 to 1.8. As shown in Fig. 10, for the first experiment the range is from about 0.1 to 1.8, with a minimum in the wind speed range of 5 to 15 m/s. Ratios were smaller for the second experiment. As shown in Fig. 11,  $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratios are about  $10^{-1}$  for wind speeds less than 10 m/s. Only for the highest wind speed increment is there a significant increase in the activity ratio to about 0.5.

#### Airborne Plutonium and Americium at Site 4

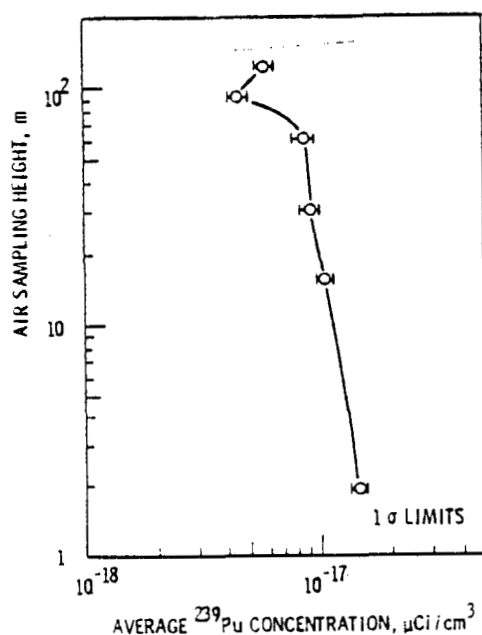
Airborne concentrations of plutonium and americium were determined (Sehmel 1978b, 1982c) during two experiments at site 4. Here concentrations were determined for heights up to 124 m. Sampling heights were changed slightly for the second experiment. Experiments were conducted from August 13 to November 12, 1976, and from April 12 to July 25, 1979. Airborne plutonium



was determined during the first experiment, and both airborne plutonium and americium were determined during the second experiment. The following results are discussed: 1) airborne radionuclide concentrations,  $\mu\text{Ci}/\text{cm}^3$  of sampled air, and 2) radionuclide activity densities,  $\mu\text{Ci}/\text{g}$  of airborne solids.

Site 4 from August 13 to November 12, 1976. Plutonium concentrations and fluxes were determined for sampling heights up to 122 m. Samples were collected for all wind speeds and directions using cowl cascade-impactors (see method number 1 illustrated in Fig. 2).

Airborne plutonium concentrations on small particles are shown in Fig. 12. Concentrations were highest at the lowest sampling height of 1.9 m and decreased with increasing height to 91 m. This concentration profile (concentration as a function of height with a maximum concentration near ground) reflects that airborne concentrations are affected significantly by resuspension from upwind. Nevertheless, it is unclear why the concentration at 91 m was less than the concentration at 122 m.



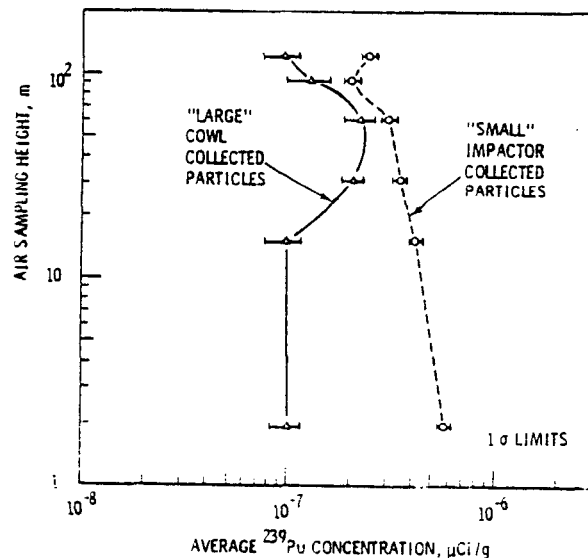
Neg. 7711495-8

Fig. 12. Airborne  $^{239+240}\text{Pu}$  concentrations on small particles at site 4 from August 13 to November 12, 1976 (impactor stages plus backup filter).

Plutonium-239,-240 activity densities as a function of height are shown in Fig. 13 for both small and large particles. Possible explanations for these profiles have at least one qualification: these average profiles are

for samples collected for all wind speeds and directions rather than from only one source. Activity densities for small particles decreased with increasing height up to 91 m and for large particles are maximum at a height between 17 and 63 m. This decrease for small particles might be explained by sampling both "more contaminated" locally resuspended particles and "less contaminated" soil transported from greater distances. For large particles, the maximum activity density occurred for a height between 30 to 60 m. This airborne plume concentration profile might be explained if plutonium on large host particles were resuspended at distances upwind. If redeposition was significant between the resuspension and sampling sites, and if "less contaminated" local-soil particles were resuspended, the average of both (upwind and local soil) decreased the average activity density.

Site 4 from April 12 to July 25, 1979. In general, little was known about enrichment processes, enrichments for different radionuclides, or for different airborne particle diameters. Hence, plutonium and americium concentrations and activity densities were investigated simultaneously as a function of host-particle diameter and sampling heights up to 124 m at site 4 (Sehmel 1982c).



Neg. 7711495-12

Fig. 13. Plutonium-239,-240 on airborne solids collected at site 4 from August 13 to November 12, 1976.

Airborne concentrations and activity densities are shown as two subfigures in Figs. 14 and 15. Airborne concentrations for  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  are shown in Figs. 14b and 15b, respectively, for sampling during a  $225^\circ \pm 35^\circ$  wind direction. Activity densities are shown in the left subfigure. Results for all other wind directions,  $45^\circ \pm 145^\circ$ , are tabulated in Table 3. These samples were collected during significantly different time periods because the equipment malfunctioned. Because samples were collected at significantly different time periods, comparing changes with height is not significant.

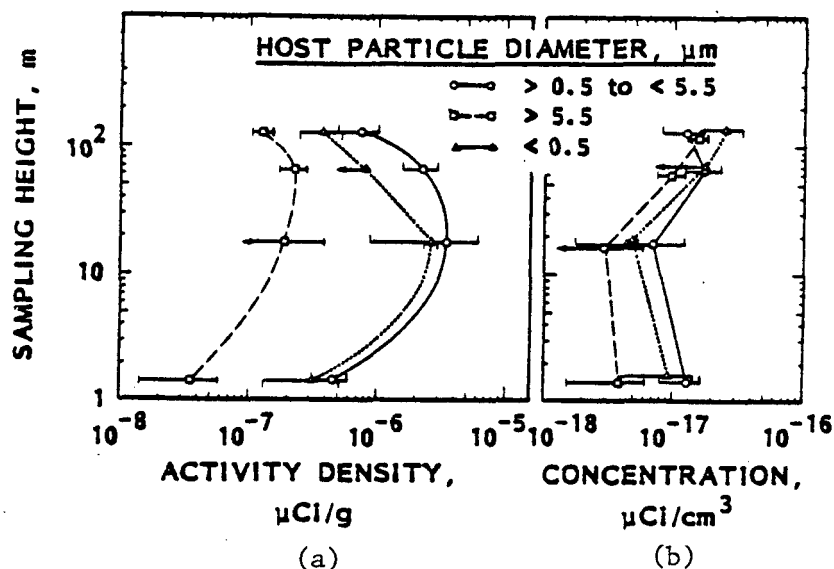


Fig. 14. Airborne  $^{239+240}\text{Pu}$  activity densities and concentrations as a function of height at site 4 from April 12 to July 25, 1979, when sampling during winds from  $225^\circ \pm 35^\circ$ .

Activity densities and airborne concentrations were a function of both sampling height and host-particle diameter. For both plutonium and americium, activity densities were maximum near the 17-m sampling height. These maximum activity densities may reflect upwind resuspension with subsequent dry deposition occurring between resuspension sources and site 4. For plutonium, activity densities were maximum for the intermediate size range,  $>0.5$  to  $<5.5$ - $\mu\text{m}$  aerodynamic particle diameter. The particle-size range for maximum activity densities for  $^{241}\text{Am}$  is less certain because americium analyses were made for only two particle diameter ranges of  $<0.5$  and  $>5.5$   $\mu\text{m}$ . Similar to plutonium for these two size ranges, the maximum americium activity density occurred, with one exception, at the  $<0.5$ - $\mu\text{m}$  size. At the 62-m sampling height, the americium activity density on the  $>5.5$ - $\mu\text{m}$  size was larger than on the  $<0.5$ - $\mu\text{m}$  size.

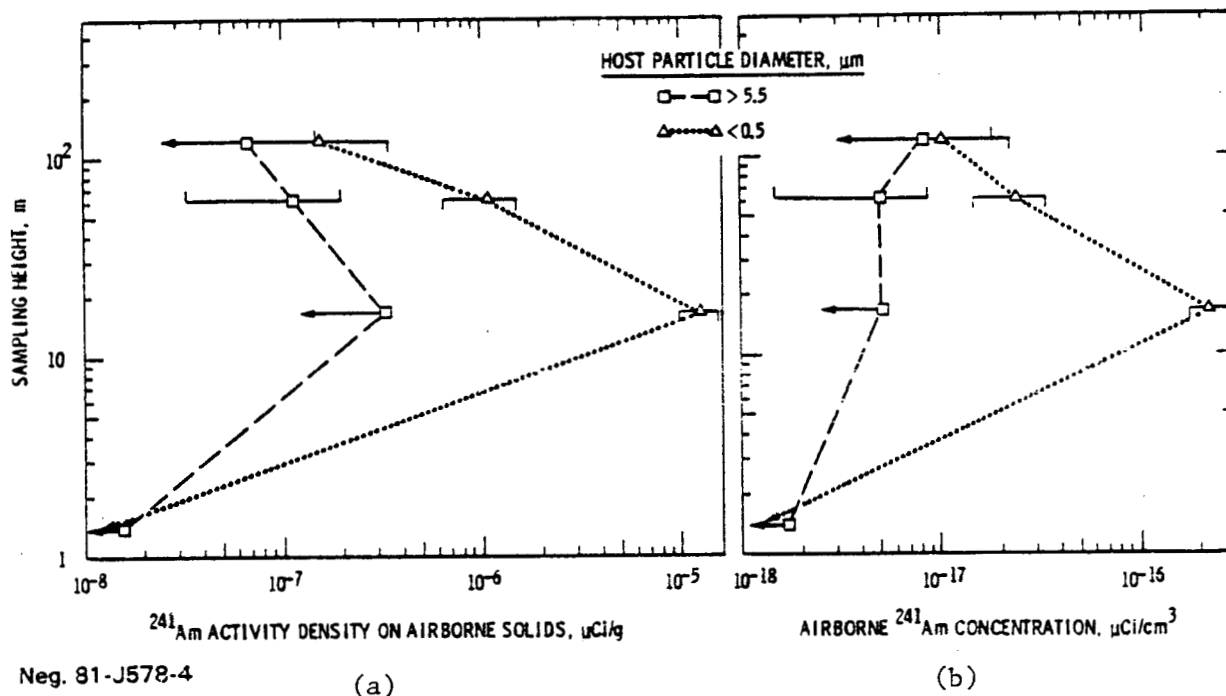


Fig. 15. Airborne  $^{241}\text{Am}$  activity densities and concentrations as a function of height at site 4 from April 12 to July 25, 1979, when sampling during winds from  $225^\circ \pm 35^\circ$ .

TABLE 3 Airborne  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  Activity Densities and Concentrations as a Function of Height at Site 4 from April 12 to July 25, 1979, when Sampling During Winds from  $45^\circ \pm 145^\circ$

Sampling Height (m)	Host-Particle Diameter ( $\mu\text{m}$ )	$^{239+240}\text{Pu}$			$^{241}\text{Am}$		
		Activity Density ( $\mu\text{Ci/g}$ )	Airborne Concentration ( $\mu\text{Ci/cm}^3$ )	1 $\sigma$ Radiochemical Error ( $\pm\%$ )	Activity Density ( $\mu\text{Ci/g}$ )	Airborne Concentration ( $\mu\text{Ci/cm}^3$ )	1 $\sigma$ Radiochemical Error ( $\pm\%$ )
124	$> 5.5$	$2.1 \times 10^{-7}$	$3.7 \times 10^{-16}$	28	$< 5.2 \times 10^{-6} \text{ (a)}$	$< 8.9 \times 10^{-19} \text{ (a)}$	(a)
	$> 0.5 \text{ to } < 5.5$	$2.2 \times 10^{-6}$	$8.5 \times 10^{-16}$	19	NA	NA	NA
	$< 0.5$	$6.7 \times 10^{-7}$	$5.0 \times 10^{-16}$	33	$1.6 \times 10^{-6}$	$1.2 \times 10^{-17}$	42
62	$> 5.5$	$7.7 \times 10^{-7}$	$3.9 \times 10^{-16}$	12	$2.8 \times 10^{-7}$	$1.1 \times 10^{-18}$	25
	$> 0.5 \text{ to } < 5.5$	$1.1 \times 10^{-6}$	$5.9 \times 10^{-16}$	10	NA	NA	NA
	$< 0.5$	$7.6 \times 10^{-7}$	$7.0 \times 10^{-16}$	12	$7.9 \times 10^{-6}$	$7.2 \times 10^{-19}$	75
1.4	$> 5.5$	$6.8 \times 10^{-8}$	$3.1 \times 10^{-18}$	11	$1.2 \times 10^{-8}$	$5.4 \times 10^{-19}$	29
	$> 0.5 \text{ to } < 5.5$	$1.2 \times 10^{-6}$	$7.4 \times 10^{-18}$	7	NA	NA	NA
	$< 0.5$	$3.5 \times 10^{-7}$	$2.2 \times 10^{-17}$	8	$4.2 \times 10^{-6}$	$2.6 \times 10^{-18}$	17

(a) This is the radiochemical analysis error when the radiochemical result is zero.  
NA = not analyzed for americium content.

### Simultaneous Spatial Variation at Two Sites

Airborne plutonium concentrations were determined (Sehmel 1981d) at sites 1 and 2 by simultaneously collecting airborne solids only during southwesterly winds. The sampling time period was September 17 to November 8, 1979. Also, airborne solid samples were collected simultaneously at sites 3 and 5 by continuous sampling from July 25 to November 8, 1979. Reported here are 1) airborne plutonium and americium concentrations and 2) plutonium activity densities on airborne small particles.

Calculated airborne plutonium concentrations are shown in Figs. 16 and 17. Figures 16a and 17a correspond to samples collected as a function of wind direction with cyclone preseparators at sites 1 and 2. Figures 16b and 17b correspond to samples collected for all wind directions, using massive-volume air samplers at sites 3 and 5.

Airborne plutonium data are shown as a function of size; the host airborne particle size is reflected in the solid collection sites within each sampler. For the cyclone-preseparator systems, data are shown for solids collected in the cyclone (a 5.5- $\mu\text{m}$  50%-collection-efficiency diameter), on the backup filter, and the total of cyclone plus backup filter collections. For the massive-volume air samplers, data are shown for 1.7- $\mu\text{m}$ -diameter particles (the 50%-collection-efficiency diameter for the second impactor stage) and for solids collected on the backup electrostatic precipitator collections. Data symbol offsets are used in Figs. 16 and 17 to decrease the visual superposition of data symbols.

Airborne Plutonium Concentrations. Airborne  $^{239+240}\text{Pu}$  concentrations are shown in Fig. 16. Most of the plutonium concentration was on the small particle size. Airborne concentrations are low; the maximum is only  $1 \times 10^{-17} \mu\text{Ci}/\text{cm}^3$ .

Activity Density on Airborne Solids. The  $^{239+240}\text{Pu}$  activity density on airborne solids,  $\mu\text{Ci}/\text{g}$ , is shown in Fig. 17. Activity densities range from about  $1 \times 10^{-7}$  to  $3 \times 10^{-7} \mu\text{Ci}/\text{g}$ , except for two activity densities that are smaller. The two exceptions are the large particles collected near sites 3 and 5. For these exceptions, activity densities range from about  $2 \times 10^{-8}$  to  $6 \times 10^{-6} \mu\text{Ci}/\text{g}$ , a factor of about three less than the average activity densities in the other samples.

### Americium-241 on Airborne Solids Compared to Surface Soil Samples

Americium-241 enrichments on airborne versus surface soil are calculated from airborne samples (Sehmel 1982d) and soil surface samples (Price, Gilbert

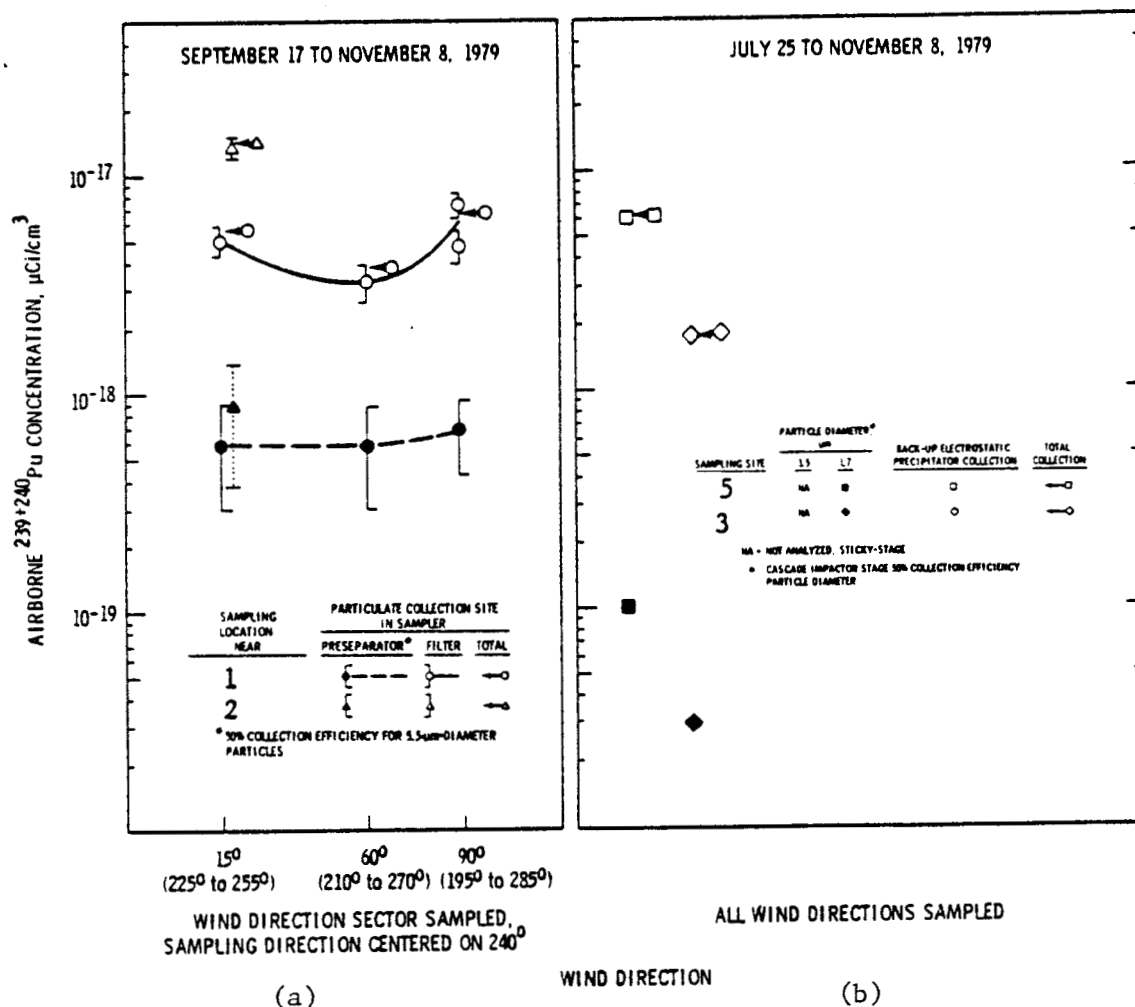


Fig. 16. Airborne  $^{239+240}\text{Pu}$  concentrations at sites 1, 2, 3, and 5 as a function of wind direction sampled for winds measured at the 1.5-m height during 1979.

and Gano 1981). Samples were collected at different locations. The airborne samples were collected at site 4. Surface samples were collected at two sites, about 6 km east-southeast and approximately 60 km west of site 4. Samples were collected either as a function of particle diameter or for all particle diameters. The airborne particles were collected with cyclone preseparator cascade-impactor collection systems (sampling method number 2). Americium-241 activity densities were obtained for the  $>5.5\text{-}\mu\text{m}$ -diameter airborne size fractions. The soil-surface samples were collected for sampling depths up to 2.5 cm.

Reported americium activity densities ranges are summarized in Table 4. Activity densities range from less than radiochemical detection limits to  $1.2 \times 10^{-5} \mu\text{Ci/g}$ . Resuspended americium-241 is enriched on airborne soils. However, enrichment differences in activity densities as a function of

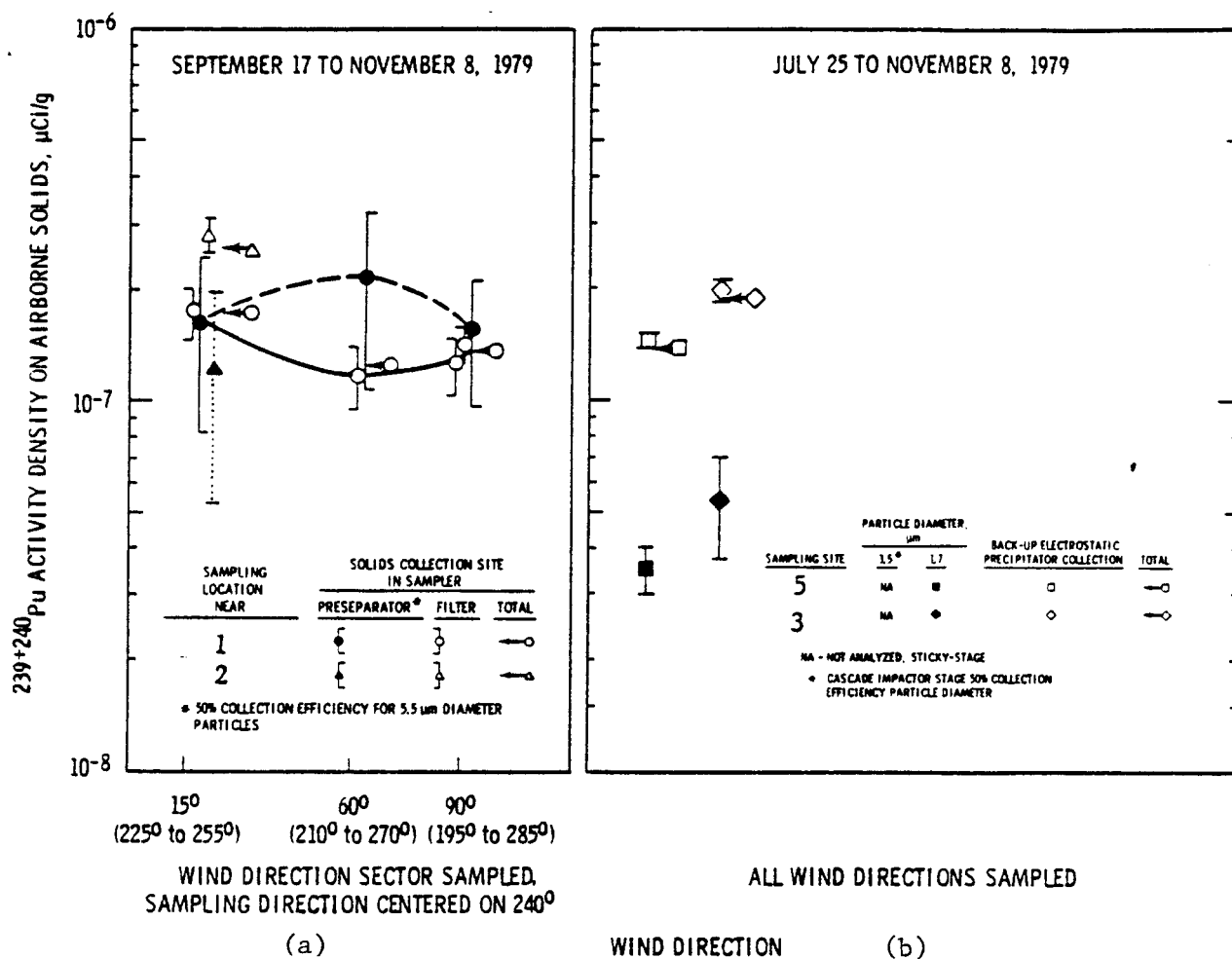


Fig. 17. Plutonium-239,-240 activity densities on airborne solids at sites 1, 2, 3, and 5 as a function of wind direction sampled for winds measured at 1.5-m height during 1979.

TABLE 4 Americium-241 Activity Densities

Sample Source	Particle Diameter, $\mu\text{m}$	Activity Density Range, $\mu\text{Ci/g}$
Airborne at site 4	>5.5	LAL <sup>a</sup> to $1.1 \times 10^{-7}$
Airborne at site 4	<0.5	LAL <sup>a</sup> to $1.2 \times 10^{-5}$
Soil surface 6 km ESE of site 4	all sizes	LAL <sup>a</sup> to $9.5 \times 10^{-8}$
Soil surface 60 km W	all sizes	LAL <sup>a</sup> to $9.8 \times 10^{-8}$

<sup>a</sup>LAL = Less than radiochemical analytical limits.

particle size are evident. The upper limit of each activity density range is largest for the <0.5- $\mu\text{m}$ -diameter airborne particles. The upper limit for 0.5- $\mu\text{m}$  particles,  $1.2 \times 10^{-5} \mu\text{Ci/g}$ , is  $1.2 \times 10^2$  times the upper limit for surface soils,  $9.8 \times 10^{-8} \mu\text{Ci/g}$ . In contrast, the upper limit for 5.5- $\mu\text{m}$  particles,  $1.1 \times 10^{-7} \mu\text{Ci/g}$ , is only 1.1 times the upper limit for surface soils.

#### Relationship Between Plutonium Activity Densities on Airborne and Surface Soils

Although only plutonium activity densities on airborne solids were evaluated in these experiments, a method was sought to relate activity densities on airborne solids to activity densities on surface soils. A predictive correlation was developed (Sehmel 1983c) based on published data for Bikini Atoll, Hanford, and Rocky Flats. The predictive correlation is applied in the mass-loading approach for prediction of airborne plutonium concentrations from surface concentrations.

Airborne concentrations from resuspension have been predicted based on considerations of airborne mass loading and activity densities on airborne soil (Anspaugh, Shinn and Wilson 1974; Anspaugh et al. 1975). The mass-loading approach was developed based on an assumption of equality; i.e., the activity density for airborne soil ( $A_{\text{AIR}}$ ) is identically equal to the activity density of local soil surface  $A_{\text{SURFACE}}$ . In this case, airborne pollutant concentrations are predicted from Equation 1:

$$C = (ML)(A_{\text{AIR}}) \quad (1)$$

where ML = airborne soil mass loading per unit volume of air

$A_{\text{AIR}}$  = activity density for airborne soil.

The method was based on data for plutonium. In developing this method, predicted airborne concentrations,  $\mu\text{Ci/cm}^3$ , were based on an assumed airborne mass loading of  $100 \mu\text{g/m}^3$  and an equality of  $A_{\text{AIR}}$  and  $A_{\text{SURFACE}}$ . The activity density on airborne solids was not determined.

An improved but conservative high-value relationship was developed (Sehmel 1982a) for using the mass-loading calculational approach for predicting airborne plutonium concentrations for wind resuspension. The improvement was to include the observed activity densities on airborne soils over regions of different surface-contamination levels. The relationship was developed from



plutonium activity densities on airborne,  $A_{AIR}$ , and surface soils,  $A_{SURFACE}$ , at Bikini Atoll, Hanford, and Rocky Flats. Activity density ranges are shown in Fig. 18 for fCi/g (i.e.,  $10^{-15}$  Ci/g) of collected solids. The activity density range on airborne soils is one order of magnitude less than the activity density range on surface soils. All plutonium activity density data on surface soils were determined for total soil depth samples, rather than as a function of host-soil particle diameters to which plutonium was attached. Thus, the reported plutonium activity densities are for both respirable and nonrespirable-size host-soil particles. In contrast, most plutonium activity densities for airborne solids were determined for particle sizes that were respirable or near respirable.

Maximum activity densities on airborne soils as a function of activity densities on surface soils are represented by Equations 2 and 3. These equations represent maximum observed plutonium activity densities on airborne soils as functions of minimum plutonium activity densities on surface soils. These equations intersect at an activity for surface soils of  $2 \times 10^3$  fCi/g. For surface-soil activity densities "less" than at the intersection, the equation is:

$$A_{AIR} = 3.3 \times 10^3 (A_{SURFACE})^{0.35} \quad (2)$$

For surface-soil activity densities "higher" than at the intersection, the equation is

$$A_{AIR} = 1.3 \times 10^4 (A_{SURFACE})^{0.17} \quad (3)$$

These correlations do not address changes in resuspension as function of wind speed, surface cover, or contaminant parameter other than the activity density for surface soils. The activity density on airborne soil is not equal to the activity density on surface soils for sites investigated. The inequality is caused in part by the relative contributions to the airborne mass loading from soil transported from upwind as compared to soil resuspended near the airborne-particulate sampling site.

#### Resuspension Factors for Aged Plutonium Sources

Resuspension factor ranges were calculated (Sehmel 1982b) from the upper limits for activity densities on airborne solids, i.e., from Equations 3 and 4.

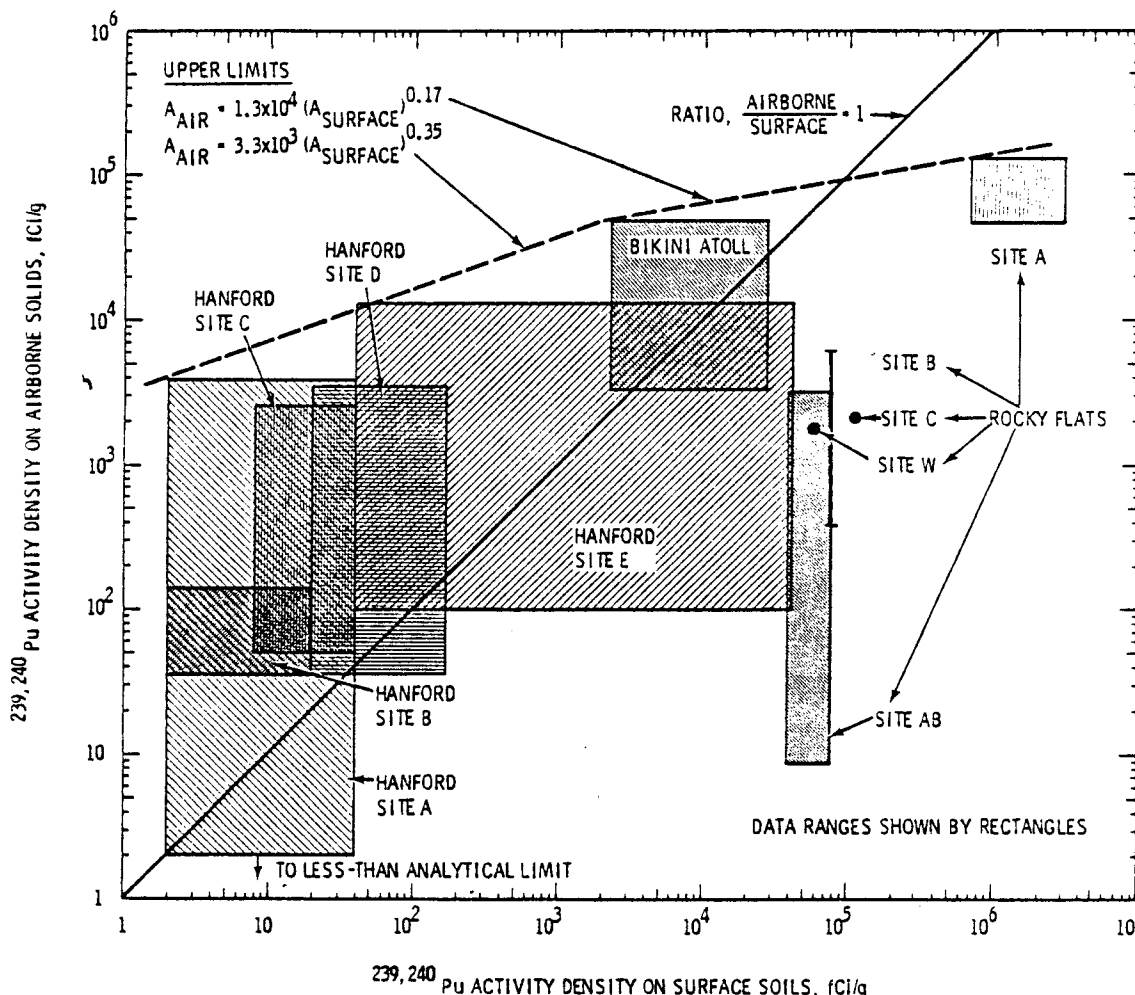


Fig. 18. Plutonium-239,-240 activity densities on airborne and surface soils. For these calculations, ranges of airborne mass loadings,  $C$ , and resuspension-surface-source depths,  $\Delta z$ , were assumed. Next, activity densities were applied to the corresponding calculated-soil masses. Thus, resuspension factors were calculated from:

$$RF = \frac{A_{AIR} (C, \mu\text{g}/\text{m}^3) (\text{g}/[10^6 \mu\text{g}])}{A_{SURFACE} (\Delta z, \text{cm}) (10^4 \text{ cm}^2/\text{m}^2) (1.5 \text{ g}/\text{cm}^3)} =$$

$$RF = 6.7 \times 10^{-11} \frac{(A_{AIR})}{(A_{SURFACE})} \frac{(C)}{(\Delta z)} \quad (4)$$

where a soil density of  $1.5 \text{ g}/\text{cm}^3$  was assumed. Calculations were made for resuspension-source depths,  $\Delta z$ , of 0.1 and 1.0 cm and for airborne mass load-

ings of 10 and 260  $\mu\text{g}/\text{m}^3$ . These ranges are considered realistic. The resuspension source depth that is actually resuspended is unknown, but probably is on the order of 0.1 cm for nonepisodic resuspension events. The 10- $\mu\text{g}/\text{m}^3$  airborne-mass loading corresponds to clean air, while the 260- $\mu\text{g}/\text{m}^3$  airborne mass corresponds to the 24-hr maximum National Ambient Air Quality Standard (NAAQS). Calculations were made for surface-soil activity densities of 2 fCi/g and  $3 \times 10^6$  fCi/g (corresponding to the minimum and maximum values shown in Fig. 18) and also at an intermediate value of  $9 \times 10^4$  fCi/g for which airborne and surface activities are equal. Results are shown in Table 5.

TABLE 5 Resuspension Factors for Aged Plutonium Sources

Parameters	Surface Contamination		
	Low	Intermediate*	High
Assumptions:			
Activity density, $A_{\text{surface}}$ , fCi/g	2	$9 \times 10^4$	$3 \times 10^6$
Activity density, $A_{\text{air}}$ , fCi/g	$4.2 \times 10^3$	$9 \times 10^4$	$1.6 \times 10^5$
Enrichment factor, $A_{\text{air}}/A_{\text{surface}}$	$2.1 \times 10^3$	1.0	$5.3 \times 10^{-2}$
Resuspension factor, range, $\text{m}^{-1}$ , for the following conditions:	$10^{-6}$ to $10^{-4}$	$10^{-9}$ to $10^{-7}$	$10^{-11}$ to $10^{-8}$
for 0.1 cm soil depth			
Airborne mass loadings C of:			
C = 10 $\mu\text{g}/\text{m}^3$	$1.4 \times 10^{-5}$	$6.7 \times 10^{-9}$	$3.6 \times 10^{-10}$
C = 260 $\mu\text{g}/\text{m}^3$	$3.7 \times 10^{-4}$	$1.7 \times 10^{-7}$	$9.2 \times 10^{-9}$
for 1.0 cm soil depth			
Airborne mass loadings C of:			
C = 10 $\mu\text{g}/\text{m}^3$	$1.4 \times 10^{-6}$	$6.7 \times 10^{-10}$	$3.6 \times 10^{-11}$
C = 260 $\mu\text{g}/\text{m}^3$	$3.7 \times 10^{-5}$	$1.7 \times 10^{-8}$	$9.2 \times 10^{-10}$

\*Intermediate corresponding to an enrichment factor of unity.

Resuspension factors are a function of the surface contamination and airborne mass loadings. Calculated resuspension factors range six orders of magnitude from  $10^{-11}$  to  $10^{-4} \text{ m}^{-1}$ , a range almost comparable to the eight orders of magnitude from  $10^{-10}$  to  $10^{-2} \text{ m}^{-1}$  range of experimentally measured resuspension factors (Sehmel 1980a). The similarity is encouraging and lends credence to the resuspension factor predictions. Predicted resuspension factors range

only two orders of magnitude from  $10^{-6}$  to  $10^{-4} \text{ m}^{-1}$  for low-contamination surfaces, activity densities that might be applicable for the general population. In comparison, resuspension factors range three orders of magnitude from  $10^{-11}$  to  $10^{-8} \text{ m}^{-1}$  for the highest reported activity densities for which controls might be applicable.

### Conclusions

Wind resuspension of surface contamination continues for many years. Continued resuspension was shown from investigations of airborne plutonium and americium concentrations ( $\mu\text{Ci}/\text{cm}^3$ ) and activity densities on airborne solids ( $\mu\text{Ci}/\text{g}$ ) resuspended from aged-area sources. Results indicate airborne concentrations and activity densities for aged resuspension sources are functions of host-particle diameter, sampling height, wind speed, and wind direction (direction of highest frequency of high wind speeds). This data base is important for directing predictor development for long-term environmental assessment effects. In general, predictors need to consider the following dependencies:

- Compared to activity densities ( $\mu\text{Ci}/\text{g}$ ) for bulk soil samples, radionuclides are enriched on airborne respirable-particle sizes.
- Most resuspension sources are nonuniform in surface-contamination-per-unit area. For nonuniform resuspension sources, airborne concentrations are functions of height and sampling location. Depending on the sampling location and height, maximum airborne concentrations can be near the surface or greater heights.
- The apparent wind speed dependency of airborne concentrations can be influenced by upwind terrain variations. For sites in relatively flat terrain, plutonium concentrations tend to increase according to a power function of wind speed. When a power law dependency is assumed,  $U^n$ , the exponent  $n$  ranges from 0.8 to 7.8. For a mountaintop site, airborne concentrations are more complex functions of wind speed.
- Americium/plutonium activity ratios may not be unity, and can be a function of wind speed.

Enrichment of activity densities on airborne solids versus bulk soil-samples, and predicted resuspension factors from a correlation of maximum enrichments at several sites, are discussed further.

Resuspension-source surface contamination can be estimated from bulk soil samples. However, if activity densities for these bulk surface samples are low, significant resuspension can occur if surface contamination is preferentially attached to small particles (i.e., enrichment). This is expected because plutonium on resuspended airborne host particles is enriched,  $\mu\text{Ci/g}$ , in the respirable versus the nonrespirable host particle-size range. Also, americium is enriched on airborne respirable particles. Although enrichments do occur for plutonium and americium, and probably most radionuclide and toxic substances, enrichments can be only imprecisely predicted. An increasing enrichment data base is needed for general predictions.

Enrichments on the respirable particle-size range in bulk surface-soil samples may be even greater than enrichments evaluated from airborne particles. This potential increased enrichment reflects that collected airborne solids did not contain the largest soil particles (soil particles of lowest activity density), but the largest mass-fraction of surface soils. Truncation in the airborne-size distribution is caused by preferential transport of the largest surface-soil particles, which tend to move by surface creep and saltation. Most of these particles are usually transported below 0.3 m, the lowest sampling height investigated.

An activity density relationship was developed from activity densities on airborne solids and surface soils at Bikini Atoll, Hanford site, and Rocky Flats, for improved predictions of airborne concentrations from wind resuspension. The maximum expected plutonium-activity density on airborne solids can be estimated from activity densities for bulk surface-soil samples. The relationship can be used to decrease the ultraconservatism in estimating airborne plutonium concentrations above more "highly" contaminated surface areas and to more realistically predict airborne concentrations over surfaces of "lesser" surface contamination (i.e., much closer to surface-contamination levels attributed solely to fallout debris). The relationship is useful for predicting airborne concentrations above contaminated areas but has less utility for predicting airborne concentrations downwind. Resuspension rates (Sehmel 1980a) are needed for predicting airborne concentrations downwind.

An important predictive improvement from the correlation is that resuspension-factor ranges can now be predicted more accurately as a function of the resuspension-source activity densities. These predicted resuspension factors could be used in long-term assessments for aged resuspension sources.

In comparison, resuspension from freshly deposited sources are expected to be more rapid. Less confidence should be placed in resuspension-factor predictions for freshly deposited sources.

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