

Radionuclides and Inorganics in Ambient Aerosols

Introduction

CEMRC collects and analyzes samples of airborne particulate matter ("aerosols") as part of its WIPP Environmental Monitoring (EM) project. Although the term "aerosol" technically applies to both the solid and liquid particles and the gases in which they are suspended, common usage allows "aerosol" to refer to the particles alone, a practice that will be followed here.

These studies of atmospheric aerosols are an important part of the Center's monitoring efforts because if a situation arose in which radioactive or chemical contaminants were released from the WIPP, those materials could be rapidly dispersed through the atmosphere and spread throughout the environment. Furthermore, in the unlikely event of such a release, the inhalation of aerosol contaminants from the WIPP would represent a major route of exposure to radionuclides and other chemicals for the local citizenry.

Aerosol studies for the WIPP EM began prior to the receipt of the first shipment of radioactive waste at the WIPP, which was on March 26, 1999. Data for the samples collected before the receipt of that waste shipment have been used to characterize the baseline concentrations of selected radionuclides in the atmosphere of the WIPP environs. Baseline data for selected inorganic substances extend up to September 9, 2000, when the first shipments of mixed waste arrived at the site. The baseline aerosol data are being compared with results from the ongoing WIPP EM monitoring studies as a means of determining whether activities at the WIPP site have affected radionuclide activities in the atmosphere or the concentrations of nine elements of primary concern (As, Be, Ba, Cd, Cr, Pb, Hg, Se and Ag). These elements are listed as components of the Permitted TRU Mixed Wastes in the WIPP hazardous waste permit (*Waste Acceptance Criteria for*

the Waste Isolation Pilot Plant, DOE/WIPP-069, November 8, 1999).

It bears mention at the outset of this section, that there is no evidence to date that operations at the WIPP have significantly impacted the activities of any radionuclides or concentrations of any trace elements of concern. In the discussion of the aerosol results that follows, we first present a condensed version of a manuscript reporting the results of a study of $^{239,240}\text{Pu}$ and inorganic substances in aerosols from the vicinity of the WIPP site (Arimoto et al., *Health Physics*, in press) and then conclude the discussion with a synopsis of the more recent monitoring data.

Methods

Detailed information regarding the sampling design for the WIPP EM ambient aerosol studies has been presented in the description of the WIPP Environmental Project above and in the CEMRC reports for 1998 to 2000. Briefly, for the aerosol studies, several different types of samples were collected from three sampling stations, On Site, Near Field and Cactus Flats (Fig. 2). For the radionuclide studies, high-volume samples were collected on 20 x 25 cm Gelman A/E™ glass fiber filters, which were changed when the flow rates dropped to 90% of their starting values. Therefore, the time intervals for the radionuclide sample collections were variable, but typically several weeks in length. At Cactus Flats and Near Field, high-volume samples for the radiochemistry studies were collected for both total suspended particles (TSP) and PM_{10} (particles less than 10 μm , aerodynamic equivalent diameter). At On Site, only TSP samples were collected for radionuclide analyses.

Gravimetric measurements were made to determine the amounts of aerosol mass collected on the high-volume filters. Prior to sampling, new filters were weighed without being desiccated, and after exposure and re-weighing, the total mass that accumulated on a filter was divided by the total air volume drawn through it to calculate the aerosol mass concentration. Gravimetric determinations were only made for

the high-volume filters because static charging and other technical problems caused the weights of the other types of filters to vary erratically.

The high-volume samples were analyzed for selected radionuclides, including ^{238}Pu and $^{239,240}\text{Pu}$, and for these analyses, entire filters were muffled for 4 hr at 500°C and then spiked with Pu tracers. The samples were dissolved using HF, HCl and HClO_4 , and the resulting solutions processed by multiple precipitation, co-precipitation, and ion-exchange and/or extraction chromatography steps. The nuclides of interest were then precipitated with LaF_3 , deposited onto filters, mounted on planchettes, and counted using an Oxford Oasis alpha spectroscopy system. The plutonium data are reported in the following two ways. First, $^{239,240}\text{Pu}$ activity concentrations were calculated as the $^{239,240}\text{Pu}$ activity per unit volume of air samples during each collection period (nBq m^{-3}). Uncertainties in the activity concentrations, not taking into account errors in the volumes of air sampled (which are estimated to be $\sim 10\%$), ranged from 1.4 nBq m^{-3} for the PM_{10} samples from Near Field to 2.2 nBq m^{-3} for the TSP samples from Cactus Flats, with relatively small differences among sites. Second, activity densities were calculated as the activity of $^{239,240}\text{Pu}$ per unit mass of aerosol particles material collected (mBq g^{-1}). Uncertainties in the activity densities, again without propagating errors in the sampling volumes, ranged from 0.049 mBq g^{-1} for the On Site TSP samples to 0.080 mBq g^{-1} for the Cactus Flats PM_{10} .

For the trace element (TE) study, aerosol samples were collected from the same three stations used for the radionuclide studies. These TE samples, in contrast to the ones for the radionuclide studies, were collected using low-volume ($\sim 10 \text{ L min}^{-1}$) systems for TSP, PM_{10} and $\text{PM}_{2.5}$ (particulate matter less than $2.5 \mu\text{m}$ in diameter). From the beginning of the experiment through January 1998, all low-volume samples were collected daily using $2\text{-}\mu\text{m}$ pore-size, 47-mm diameter TefloTM filters. Starting in February 1998, the TE sampling periods

were changed to 2, 2, and 3 days per week (the filters were replaced on Monday, Wednesday, Friday). Beginning with the change in sampling intervals, the sampling substrate used for the TE study was switched from the TefloTM filters to $0.8\text{-}\mu\text{m}$ pore-size, 47-mm diameter cellulose-ester filters. The PM_{10} and $\text{PM}_{2.5}$ sampling at Near Field and Cactus Flats was discontinued in March 2000.

Aerosol filters were prepared for elemental analyses using a mixture of HCl, HNO_3 , HF, and H_2O_2 , with heat and high pressure from a microwave digestion system. The concentrations of major and trace elements were determined in the aerosol samples by atomic absorption spectrometry (AAS) and inductively coupled plasma-emission spectrometry prior to January 1999 and by AAS and inductively coupled plasma-mass spectrometry (ICP-MS) afterwards.

The inorganic data are subject to uncertainties in (a) flow volumes; (b) sample collection and storage, and preparation; (c) analytical methods; and (d) blank corrections. The magnitudes of these uncertainty components, except flow volumes, vary among analytes, and representative values for each range from $< 5\%$ to 15% , resulting in combined uncertainties of 15% to 20% . Representative detection limits for the inorganic analytes are as follows: nitrate = $0.0045 \mu\text{g m}^{-3}$, sulfate = $0.0119 \mu\text{g m}^{-3}$, sodium = $0.02 \mu\text{g m}^{-3}$, aluminum = 17 to 57 ng m^{-3} , and lead = 0.06 to 0.12 ng m^{-3} .

Results and Discussion

The results of the CEMRC aerosol studies drew our attention to the importance of re-suspension as a determinant of Pu activities in aerosols, and those results were thought to be of interest for others working on related topics. As a result, in FY 2001, a manuscript based on the WIPP-EM aerosol studies was prepared and submitted for publication in a peer-reviewed scientific journal. The sampling periods covered in the study were as follows: 2 February 1998 to 10 August 2000 for the radionuclides; 4 November 1997 to 17 August 2000 for the major ions; and 2 February 1998 to 7 September 2000 for the trace elements. Highlights of that study are presented in the next section.

Summary of: Arimoto, et. al, ^{239,240}Pu and inorganic substances in aerosols from the vicinity of the Waste Isolation Pilot Plant: The importance of resuspension (Health Physics, in press)

^{239,240}Pu Activities

^{239,240}Pu was quantified in all but one of the 141 aerosol samples analyzed. The geometric mean, volume-based, activity concentrations for ^{239,240}Pu in the PM₁₀ samples were 6.2 nBq m⁻³ at Near Field and 8.8 nBq m⁻³ at Cactus Flats while the means for the TSP samples collected over the same time periods were 12, 16, and 14 nBq m⁻³ at Near Field, Cactus Flats, and On Site respectively. The geometric mean PM₁₀ ^{239,240}Pu mass-based activity densities were 0.39 mBq g⁻¹ at Near Field and 0.47 mBq g⁻¹ at Cactus Flats. The corresponding average TSP activity densities were 0.48 (Near Field), 0.54 (Cactus Flats), and 0.33 mBq g⁻¹ (On Site).

The geometric mean ^{239,240}Pu activity concentrations in the TSP and PM₁₀ samples from Near Field were roughly half of those presented in Lee et al., (*J. Radioanal Nucl Chem*, 1998) who analyzed a more limited set of samples collected from the same site during February-May 1996. While it is possible that the activities of ^{239,240}Pu changed over time, the differences between the two data sets can more simply be explained by the strong temporal variability in ^{239,240}Pu observed in the current study as discussed below.

Temporal Variability and Resuspension

Strong seasonal cycles and interannual differences in the aerosol mass loadings and ^{239,240}Pu activity concentrations are evident for both the TSP and PM₁₀ samples (Fig. 12). The highest ^{239,240}Pu activities tend to occur in the first half of each calendar year, which is the when the samples analyzed by Lee et al., (*op cit.*) were collected, and this likely explains the high activities they observed compared with the average concentrations from the present study. Peaks in the ^{239,240}Pu activity concentrations generally were observed between March and

May each year for both the TSP and PM₁₀ samples, with especially strong springtime peaks evident in 1999. High plutonium activities also were observed at Cactus Flats and On Site in November-December 1999.

Atmospheric studies conducted prior to the end of above-ground nuclear weapons testing era showed that plutonium activities in the atmosphere varied with season, and those temporal patterns were thought to be driven by seasonal cycles in the exchange of material from stratosphere to the troposphere (Perkins and Thomas, U.S. Department of Energy: DOE/TIC-22800, 1980). While the current WIPP EM aerosol data and those collected during the time of above-ground weapons testing both show springtime peaks in aerosol ^{239,240}Pu, the causes for the cycles are likely quite different. This is because the contribution of bomb-derived ^{239,240}Pu from primary stratospheric sources to present day atmospheric loadings is likely to be small (Lee et al., *op cit.*). Atmospheric nuclear weapons testing ended in 1980 and the residence time of plutonium in the atmosphere is on the order of a year (Perkins and Thomas, *op cit.*), and therefore contemporary aerosol Pu activities are likely to be controlled by sources and processes other than weapons testing and stratosphere/troposphere exchange.

The most likely explanation for the present-day seasonal cycles is that they are a consequence of the re-suspension of previously deposited fallout nuclides. As shown in Fig. 12, the aerosol mass loadings followed a seasonal pattern similar to that of the ^{239,240}Pu activity concentrations. In addition, a scatterplot of ^{239,240}Pu activity concentrations versus aerosol masses (Fig. 13) shows a distinct linear trend between the variables, thus demonstrating that the Pu activities varied in proportion to aerosol mass concentrations.

It is apparent in Fig. 13 that the relationship between mass loadings and ^{239,240}Pu activities for the On Site TSP samples is clearly different compared with the other sets of samples. More specifically, the On Site samples generally showed lower ^{239,240}Pu to mass ratios (lower activity densities) than the other sets of samples. This result implies that activities or processes occurring at or near the WIPP site produced aerosols that contributed to the mass loadings but contained less ^{239,240}Pu than ambient

aerosols. A second implication of this finding is that local sources of non-radioactive particulate matter can affect the mass-based plutonium activity densities. Therefore measurements of activity calculated with respect to both particulate mass and air volume are useful in understanding the factors controlling the dynamics of $^{239,240}\text{Pu}$ in the atmosphere. It is also worth noting that the volume-based activity concentrations likely encompass larger error terms (than activity density) in association with air-flow volume measurement uncertainty and possible losses of particulate mass from filters during handling.

Relationships between $^{239,240}\text{Pu}$ and Major Ions and Trace Elements: Further Evidence for Resuspension

The CEMRC WIPP-EM program also involves studies of local soils, and these studies are relevant to the present discussion of aerosols because the entrainment of soil particles into the atmosphere by strong winds would cause the re-suspension of previously deposited fallout nuclides. Studies by Kirchner et al. (*op cit.*) showed that all of the radionuclides quantified in soils for the WIPP EM (^{234}U , ^{235}U , ^{238}U , ^{230}Th , ^{232}Th , ^{228}Th and $^{239,240}\text{Pu}$, ^{137}Cs , ^{208}Tl , ^{212}Pb , ^{212}Bi , ^{214}Pb , ^{228}Ac , $^{234\text{m}}\text{Pa}$, ^{241}Am , ^{40}K , ^{60}Co and ^7Be) were significantly ($p < 0.01$) correlated with soil texture and with aluminum concentrations, that is, the nuclides showed an affinity for relatively small soil particles. In atmospheric studies, aluminum has been shown to be a good indicator of mineral dust (Rahn, Grad. Sch. of Oceanogr.: Univ. of Rhode Island, Kingston, 1976; Uematsu et al., *J. Geophys. Res.*, 1983), and the Earth's upper crust contains approximately 8.04 parts per million aluminum (Taylor and McLennan, *Rev. Geophys.*, 1995).

As mineral matter was expected to be a major component of the aerosol mass, and particle mass loadings were found to be related to $^{239,240}\text{Pu}$ activities, we next focused our attention on the relationship between mineral dust and aerosol $^{239,240}\text{Pu}$. Mineral

aerosol concentrations, as represented by Al, were highly variable over time, and the highest dust concentrations occurred in the spring (Fig. 14), which is when the winds in Carlsbad are often strong and the visibility in the area occasionally impaired by raised dust. As noted above, spring is also when the highest $^{239,240}\text{Pu}$ activities are observed and the implication is that there is an association between aerosol $^{239,240}\text{Pu}$ and soil-derived aerosols.

A simple test of this hypothesis was made by examining the aerosol data for a correlation between $^{239,240}\text{Pu}$ and Al. For this analysis, the aluminum data were averaged over the longer collection intervals used for the high-volume radionuclide sample collections. Even though the radionuclide and trace element samples were not precisely matched, a scatterplot of $^{239,240}\text{Pu}$ activity versus aluminum (Fig. 15) shows the two variables were weakly but significantly correlated ($r = 0.54$, $p < 0.0001$). Furthermore, there were no discernable differences in the relationships among sites as there were for $^{239,240}\text{Pu}$ versus mass loadings.

While the correlation between $^{239,240}\text{Pu}$ and mineral dust does not prove that the activities of $^{239,240}\text{Pu}$ are controlled by resuspension, it does establish a link between plutonium activities and the concentrations of mineral particles in the atmosphere, at least some of which must be mobilized by resuspension of local soils. A comparison of the aerosol and soil data show that the $^{239,240}\text{Pu}$ activity/aluminum concentration ratio for the aerosols (all data = 3.3×10^{-2} nBq ng $^{-1}$) was much higher than for soils (6.9×10^{-5} and 1.1×10^{-5} nBq ng $^{-1}$ at Near Field and Cactus Flats, respectively), (Kirchner et al., *J. Environ. Radioactivity*, *op cit.*). The higher $^{239,240}\text{Pu}$ /aluminum ratio in aerosols relative to soils are likely due to the affinity of $^{239,240}\text{Pu}$ for small soil particles that have large surface area to mass (and volume) ratios. While these small particles contribute little to the soil mass, they tend to have higher aluminum contents than larger soil particles, which include a considerable amount of quartz. Even though small with respect to other soil particles, these Al-rich particles are large compared with most aerosols, and they would tend to remain suspended in the atmosphere much longer than the bulk of the soil-derived aerosol mass, which is quite rapidly re-deposited by gravitational

settling (Sehmel, et al., *Washington, DC: Office of Science and Technical Information CONF-841142*, 1987).

The results of the WIPP-EM study demonstrate that processes affecting the resuspension of $^{239,240}\text{Pu}$ and possibly other substances of concern are of considerable importance in terms of material budgets and must be known and quantified if any impacts of WIPP operations are to be properly evaluated. This is an important consideration because any potential releases of radionuclides or other materials from the WIPP will be superimposed on the variability caused by the winds and other meteorological factors responsible for the resuspension and re-distribution of contaminants deposited independent of WIPP operations.

The inorganic results serve to highlight the dynamics of the aerosol system in the area around the WIPP. In particular, the aluminum data show that the loadings of mineral dust in the atmosphere were higher during the spring peaks in 1999 and 2000 compared with the baseline period (Fig. 14). Because dust concentrations and $^{239,240}\text{Pu}$ activities are correlated, one can infer that interannual differences in the amounts of resuspended mineral dust would directly translate into variability in the activities of resuspended $^{239,240}\text{Pu}$, again with important implications for any programs designed to detect any potential releases from the WIPP. Such interannual variability in mineral dust concentrations is well documented in the atmospheric sciences literature, and differences of this type are influenced both by conditions in the dust source regions (Prospero and Nees, *Nature*, 1986) and variations in meteorology and transport pathways (Tegen and Miller, *J. Geophys. Res.*, 1998).

Summary of Recent Aerosol Data

Early data from the trace element studies of aerosols (covering the period from 3 February 1998 to 29 July 1999 and summarized in the CEMRC 1999 report) suggested that the sites differed in the percentages of samples with TE

concentrations above detection (Fig. 16, upper panel). As a follow-up to that analysis, the data from the beginning of the program up until July 2001 were replotted for comparison against the earlier results (Fig. 16, lower panel). It is clear from this exercise that the patterns in TE concentrations above detection limits are now comparable at all stations, with most of the changes between the earlier and more recent data occurring at Cactus Flats. In particular, the percentage of samples with Ba and Pb at Cactus Flats have increased over the course of the WIPP-EM while the number of samples with detectable Ag, Cd, and Hg at Cactus Flats has decreased.

The similarity in the percentages of samples with TE concentrations above detection limits at the three stations reinforces the notion that there has been little if any perturbation to the trace elements of concern that can be tied to activities at the WIPP. More rigorous comparisons can be made by comparing the mean concentrations using statistical methods such as the analysis of variance (ANOVAs). Two examples of such tests are presented here, one for aerosol Ba and the other for Pb because these two elements are detected in far more samples than the other elements. For these tests (Tables 3 and 4), the logarithms of the Ba and Pb concentrations were used because the data tend to be more nearly log-normally distributed than normal.

The geometric mean Ba concentrations are 4.50, 3.88 and 5.12 ng m^{-3} for Cactus Flats, Near Field and On Site, respectively. The probability (p) for differences in means as large as these due to chance alone is $p = 0.033$, which we regard as marginally significant. While it is possible that some Ba has been released by the activities at the WIPP, the concentrations at the monitoring stations were not very different from those in the exhaust shaft (i.e., the FAS samples). For comparison, the mean Ba concentration for the FAS samples presented in the 2000 CEMRC report was 7.8 ng m^{-3} , and this only slightly higher than those at the three ambient aerosol stations. Thus it is unlikely that the WIPP is a significant source for Ba especially at Near Field and Cactus Flats.

The situation for Pb is quite different from Ba: first, the Pb concentration in the exhaust shaft FAS samples (6.8 ng m^{-3} , CEMRC 2000 report) is nearly ten times higher than in the

ambient samples (0.768, 0.832 and 0.743 ng m⁻³ at Cactus Flats, Near Field, and On Site respectively). Second, and more revealing is the fact that the differences in the mean log (Pb) concentrations among stations were clearly not significant (the probability for chance occurrence from the ANOVA was $p = 0.544$). Therefore, in the case of Pb, for which there was nearly a ten-fold enrichment in the exhaust shaft aerosols, there is no evidence for a perturbation at the On Site station, which is only 0.1 km from the exhaust shaft.

As noted in the CEMRC report for 2000, other benchmarks used for interpreting the aerosol data were the maximum trace element concentrations observed during the baseline period. Table 5 presents the maximum concentrations for each trace element for the baseline and operational periods. For the elements of primary interest (As, Be, Ba, Cd, Cr, Pb, Hg, Se and Ag),

there were several cases in which the operational maxima exceeded the baseline maxima, but in most of these cases, the percentage increases were considered to be small, less than 20%. The one notable exception was Pb for which the baseline maximum at On Site was 2.05 ng m⁻³ and the operational maximum there was 8.62 ng m⁻³. Further analysis of the On Site Pb data showed that the operational maximum occurred in the first sample collected after the mixed waste was received and that the Pb concentrations in subsequent samples decreased. However, a second On Site sample (from late April 2001) had a Pb concentration of 4.36 ng m⁻³, which was also higher than the baseline maximum. As a result of these observations, the concentrations of Pb will be given special scrutiny in future studies.

Tables presenting the aerosol data summarized herein are available on the CEMRC web site at <http://www.cemrc.org>.

Table 3. Means for One-way ANOVA (LogBa)

Station	^a Number	Mean	Std Error
Cactus Flats	110	0.6531	0.0301
Near Field	96	0.5892	0.0322
On Site	95	0.7091	0.0324
		F-ratio = 3.45, p = 0.033	

^a Number = number of samples.

Table 4. Means for One-way ANOVA (LogPb)

Station	^a Number	Mean	Std Error
Cactus Flats	102	-0.1145	0.0301
Near Field	90	-0.0799	0.0320
On Site	87	-0.1287	0.0326
		F-ratio = 0.61, p = 0.544	

^a Number = number of samples.

Table 5. Maximum Trace Element Concentration for Baseline and Operational Aerosol Samples

Element	Parameter	Cactus Flats	Near Field	On Site
Ag	^a N ^b Baseline	19	19	12
	Baseline Max	4.14E-01	1.56E-01	6.23E-02
	N Operational	10	6	10
	Operational Max	3.96E-02	3.15E-02	5.24E-02
	Percent Increase	None	None	None
Al	N Baseline	82	70	66
	Baseline Max	1.86E+03	2.86E+03	1.44E+03
	N Operational	19	16	18
	Operational Max	8.04E+02	1.54E+03	1.53E+03
	Percent Increase	None	None	6.3
As	N Baseline	4	1	1
	Baseline Max	5.47E+00	3.77E+00	7.21E-01
	N Operational	0	0	0
	Operational Max	^c NA	NA	NA
	Percent Increase	None	None	None
Ba	N Baseline	81	69	65
	Baseline Max	3.33E+01	2.30E+01	2.71E+01
	N Operational	19	16	18
	Operational Max	8.08E+00	1.50E+01	2.87E+01
	Percent Increase	None	None	5.9
Be	N Baseline	0	0	1
	Baseline Max	NA	NA	9.32E-02
	N Operational	0	0	1
	Operational Max	NA	NA	1.14E-01
	Percent Increase	None	None	22.3
Ca	N Baseline	75	68	65
	Baseline Max	2.23E+03	4.55E+03	5.30E+03
	N Operational	18	14	18
	Operational Max	8.35E+02	1.10E+03	2.66E+03
	Percent Increase	None	None	None
Cd	N Baseline	19	19	11
	Baseline Max	1.39E+00	2.21E+00	1.56E+00
	N Operational	9	6	9
	Operational Max	4.60E-01	2.15E-01	8.32E-01
	Percent Increase	None	None	None

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Table 5. Maximum Trace Element Concentration for Baseline and Operational Aerosol Samples (Cont.)

Element	Parameter	Cactus Flats	Near Field	On Site
Ce	N Baseline	82	70	67
	Baseline Max	1.90E+00	3.99E+00	1.61E+00
	N Operational	19	16	18
	Operational Max	8.02E-01	1.50E+00	1.47E+00
	Percent Increase	None	None	None
Co	N Baseline	56	48	51
	Baseline Max	1.29E+01	3.10E+00	8.03E+01
	N Operational	10	7	14
	Operational Max	7.62E-01	1.10E+00	2.67E+00
	Percent Increase	None	None	None
Cr	N Baseline	10	11	10
	Baseline Max	53.7	60.1	40.9
	N Operational	6	4	5
	Operational Max	8.66	8.24	6.71
	Percent Increase	None	None	None
Cu	N Baseline	65	57	56
	Baseline Max	1.30E+01	4.89E+01	1.34E+01
	N Operational	13	12	17
	Operational Max	3.79E+00	3.45E+00	1.94E+01
	Percent Increase	None	None	44.8
Dy	N Baseline	79	61	56
	Baseline Max	1.41E-01	2.94E-01	1.15E-01
	N Operational	16	11	15
	Operational Max	5.63E-02	1.03E-01	8.74E-02
	Percent Increase	None	None	None
Er	N Baseline	66	47	46
	Baseline Max	8.81E-02	1.66E-01	7.40E-02
	N Operational	7	6	12
	Operational Max	2.97E-02	5.60E-02	6.00E-02
	Percent Increase	None	None	None
Eu	N Baseline	54	38	39
	Baseline Max	4.39E-02	7.06E-02	4.19E-02
	N Operational	4	2	8
	Operational Max	1.56E-02	2.59E-02	3.04E-02
	Percent Increase	None	None	None

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Table 5. Maximum Trace Element Concentration for Baseline and Operational Aerosol Samples (Cont.)

Element	Parameter	Cactus Flats	Near Field	On Site
Fe	N Baseline	66	58	53
	Baseline Max	8.73E+02	1.52E+03	6.88E+02
	N Operational	16	12	17
	Operational Max	4.02E+02	7.54E+02	8.13E+02
	Percent Increase	None	None	18.2
Gd	N Baseline	79	64	60
	Baseline Max	2.60E-01	3.54E-01	2.34E-01
	N Operational	16	11	15
	Operational Max	8.42E-02	1.74E-01	1.49E-01
	Percent Increase	None	None	None
Hg	N Baseline	18	15	14
	Baseline Max	2.25E-01	2.10E-01	1.39E-01
	N Operational	4	3	2
	Operational Max	3.77E-02	5.32E-02	4.29E-02
	Percent Increase	None	None	None
K	N Baseline	77	68	63
	Baseline Max	6.30E+02	1.23E+03	1.14E+03
	N Operational	17	15	17
	Operational Max	2.17E+02	4.36E+02	6.09E+02
	Percent Increase	None	None	None
La	N Baseline	68	56	54
	Baseline Max	1.97E+00	7.61E+00	1.46E+00
	N Operational	8	7	9
	Operational Max	3.92E-01	7.39E-01	7.32E-01
	Percent Increase	None	None	None
Li	N Baseline	28	19	20
	Baseline Max	2.72E+00	1.16E+00	1.08E+00
	N Operational	6	6	10
	Operational Max	4.64E-01	1.11E+00	9.19E-01
	Percent Increase	None	None	None
Mg	N Baseline	79	69	67
	Baseline Max	3.64E+02	6.69E+02	6.10E+02
	N Operational	18	15	18
	Operational Max	1.82E+02	3.43E+02	4.56E+02
	Percent Increase	None	None	None

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Table 5. Maximum Trace Element Concentration for Baseline and Operational Aerosol Samples (Cont.)

Element	Parameter	Cactus Flats	Near Field	On Site
Mn	N Baseline	80	68	65
	Baseline Max	1.89E+01	3.70E+01	1.65E+01
	N Operational	18	16	18
	Operational Max	8.48E+00	1.60E+01	1.98E+01
	Percent Increase	None	None	20.0
Mo	N Baseline	10	6	9
	Baseline Max	8.79E-01	3.57E-01	4.75E-01
	N Operational	6	8	6
	Operational Max	2.03E-01	1.98E-01	9.75E-01
	Percent Increase	None	None	105.3
Na	N Baseline	50	53	50
	Baseline Max	3.23E+03	2.40E+03	7.34E+02
	N Operational	16	15	18
	Operational Max	3.44E+02	2.74E+02	4.11E+02
	Percent Increase	None	None	None
Nd	N Baseline	82	70	67
	Baseline Max	8.40E-01	1.75E+00	6.64E-01
	N Operational	19	16	18
	Operational Max	3.10E-01	5.95E-01	6.76E-01
	Percent Increase	None	None	1.8
Ni	N Baseline	34	29	34
	Baseline Max	4.07E+01	2.08E+01	5.93E+01
	N Operational	4	4	9
	Operational Max	3.62E+00	3.45E+00	2.08E+01
	Percent Increase	None	None	None
Pb	N Baseline	74	62	58
	Baseline Max	2.95E+00	4.91E+00	2.05E+00
	N Operational	18	16	17
	Operational Max	3.44E+00	2.12E+00	8.62E+00
	Percent Increase	16.6	None	320.5
Pr	N Baseline	81	69	65
	Baseline Max	2.29E-01	7.57E-01	1.89E-01
	N Operational	19	16	18
	Operational Max	9.54E-02	1.77E-01	1.67E-01
	Percent Increase	None	None	None

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Table 5. Maximum Trace Element Concentration for Baseline and Operational Aerosol Samples (Cont.)

Element	Parameter	Cactus Flats	Near Field	On Site
Sb	N Baseline	28	26	25
	Baseline Max	1.38E+01	5.85E-01	7.65E-01
	N Operational	19	16	18
	Operational Max	6.05E-01	3.60E-01	3.02E-01
	Percent Increase	None	None	None
Sc	N Baseline	14	12	12
	Baseline Max	1.59E+00	1.19E+00	1.32E+00
	N Operational	8	6	10
	Operational Max	4.82E-01	8.74E-01	8.75E-01
	Percent Increase	None	None	None
Se	N Baseline	2	0	1
	Baseline Max	7.94E-01	NA	2.51E+00
	N Operational	2	2	2
	Operational Max	1.32E+00	1.55E+00	1.31E+00
	Percent Increase	66.2	None	None
Sm	N Baseline	76	63	56
	Baseline Max	1.90E-01	3.42E-01	1.44E-01
	N Operational	16	14	16
	Operational Max	8.26E-02	1.31E-01	1.13E-01
	Percent Increase	None	None	None
Sn	N Baseline	3	3	7
	Baseline Max	3.15E-01	1.01E+02	6.32E+01
	N Operational	0	0	0
	Operational Max	^d ND	ND	ND
	Percent Increase	None	None	None
Sr	N Baseline	73	60	61
	Baseline Max	8.72E+00	2.07E+01	9.82E+00
	N Operational	19	16	18
	Operational Max	3.51E+00	6.36E+00	7.05E+00
	Percent Increase	None	None	None
Th	N Baseline	81	68	60
	Baseline Max	3.45E-01	6.28E-01	2.70E-01
	N Operational	17	13	16
	Operational Max	1.44E-01	2.69E-01	2.71E-01
	Percent Increase	None	None	0.4

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Table 5. Maximum Trace Element Concentration for Baseline and Operational Aerosol Samples (Cont.)

Element	Parameter	Cactus Flats	Near Field	On Site
Ti	N Baseline	78	64	58
	Baseline Max	9.49E+01	1.71E+02	8.11E+01
	N Operational	15	12	16
	Operational Max	3.38E+01	6.36E+01	6.93E+01
	Percent Increase	None	None	None
Tl	N Baseline	10	8	11
	Baseline Max	9.04E-02	2.53E-01	6.20E-01
	N Operational	8	7	5
	Operational Max	7.41E-02	7.20E-02	5.23E-02
	Percent Increase	None	None	None
U	N Baseline	73	56	52
	Baseline Max	1.00E-01	1.28E-01	7.40E-02
	N Operational	16	11	16
	Operational Max	3.81E-02	6.66E-02	6.44E-02
	Percent Increase	None	None	None
V	N Baseline	26	21	24
	Baseline Max	3.08E+01	1.03E+01	2.14E+01
	N Operational	8	4	7
	Operational Max	2.69E+00	3.88E+00	4.46E+00
	Percent Increase	None	None	None
Zn	N Baseline	20	20	23
	Baseline Max	1.84E+02	7.19E+01	1.62E+02
	N Operational	12	12	15
	Operational Max	9.93E+00	1.22E+01	4.89E+01
	Percent Increase	None	None	None

^aN = number of samples

^bBaseline samples were collected prior to 9 September 2000; operational samples were collected through June 2001

^cNA = not analyzed

^dND = no data

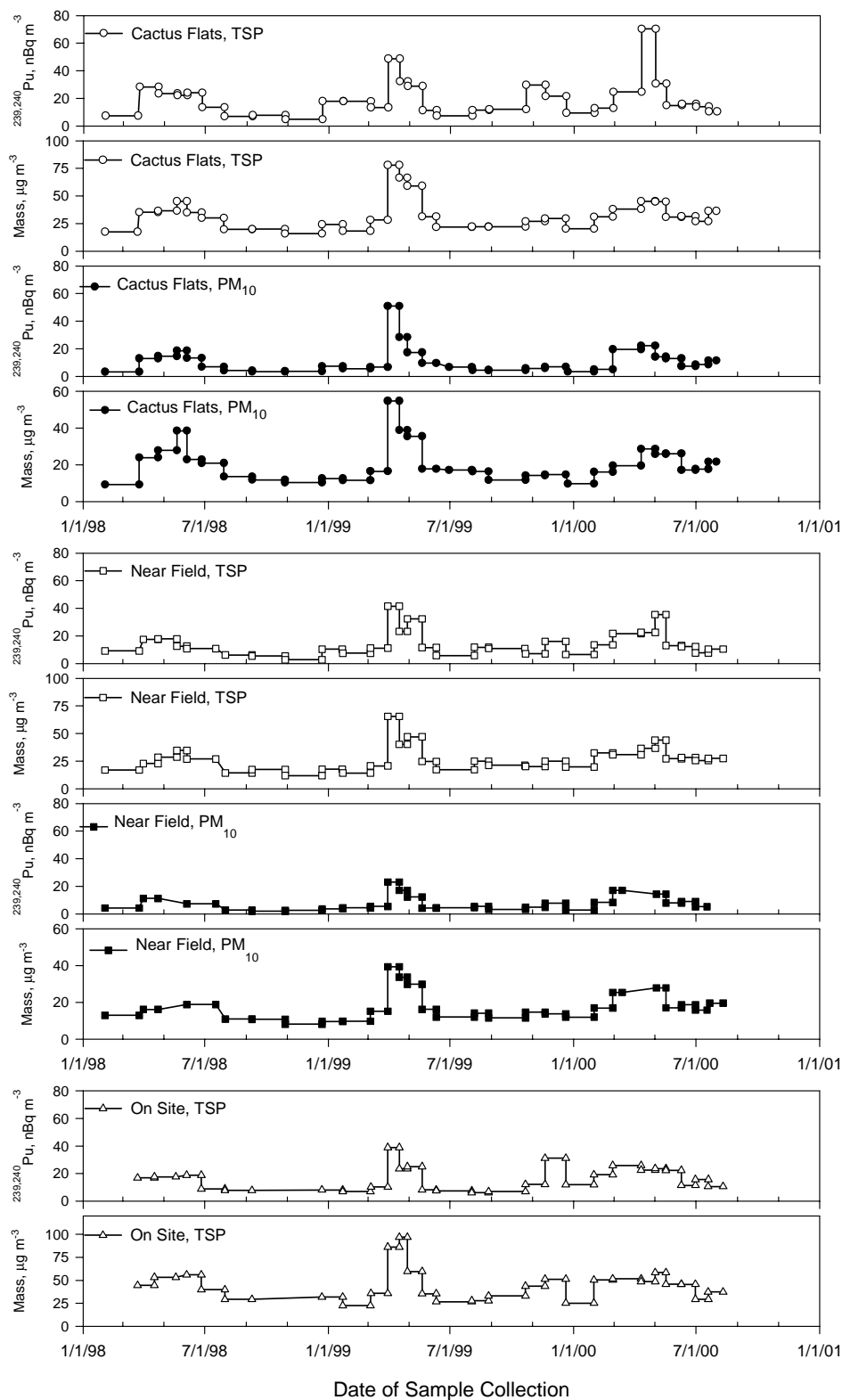


Figure 12. Mass and $^{239,240}\text{Pu}$ Activity Concentrations in Aerosol Samples Collected from February 1998 to August 2000.

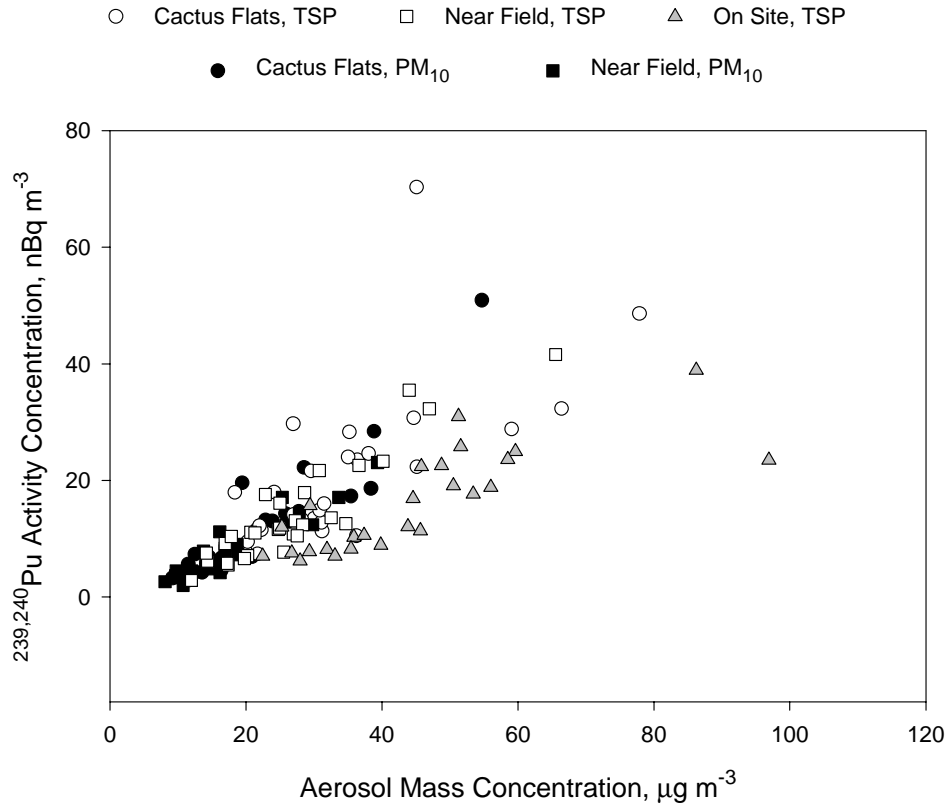


Figure 13. $^{239,240}\text{Pu}$ Activity Concentrations versus Aerosol Mass for High-Volume Samples.

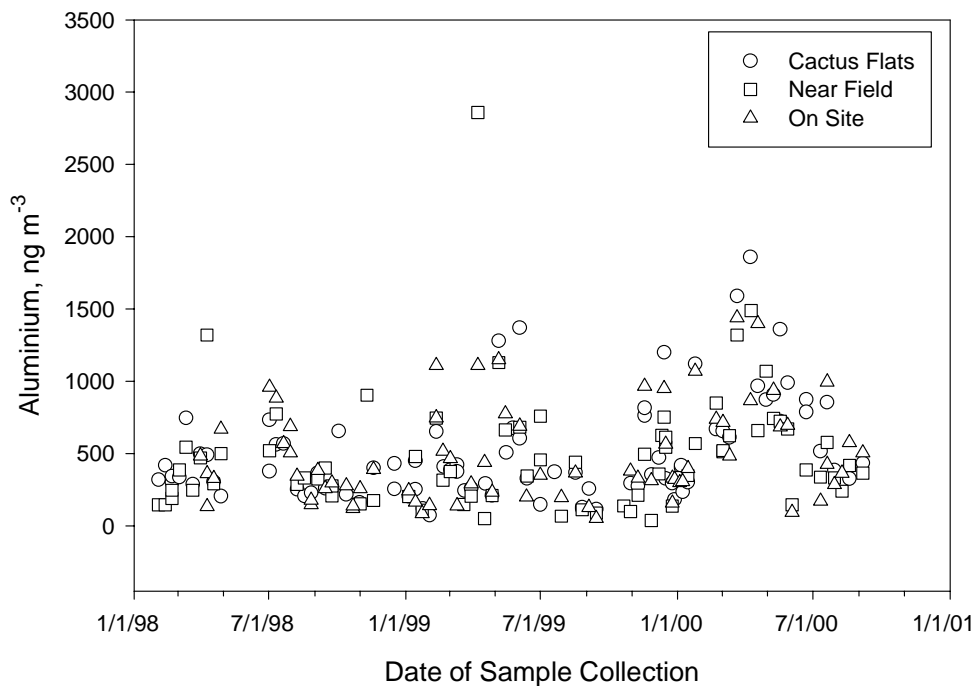


Figure 14. Temporal Variations in Aluminum Concentrations in Total Suspended Particle Samples Collected during February 1998 to July 2000.

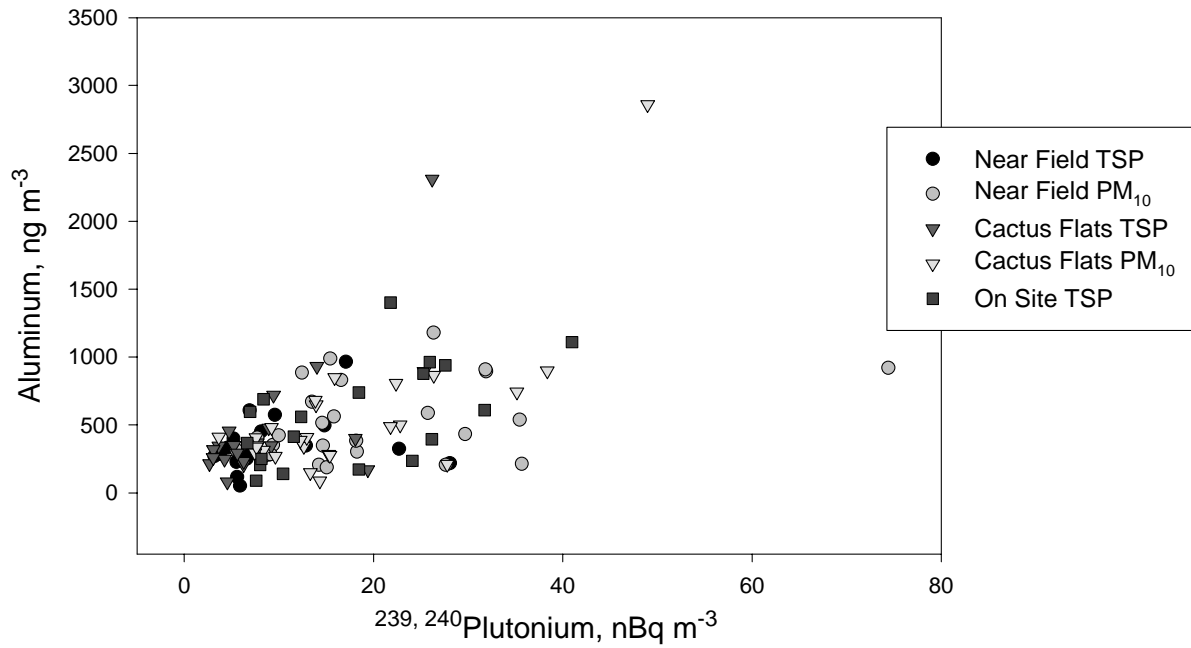


Figure 15. Relationship between $^{239,240}\text{Pu}$ vs. Aluminum Aerosol Samples from Cactus Flats, Near Field, and On Site Stations.

PM₁₀ stands for particles less than 10 μm aerodynamic diameter and TSP stands for total suspended particles.

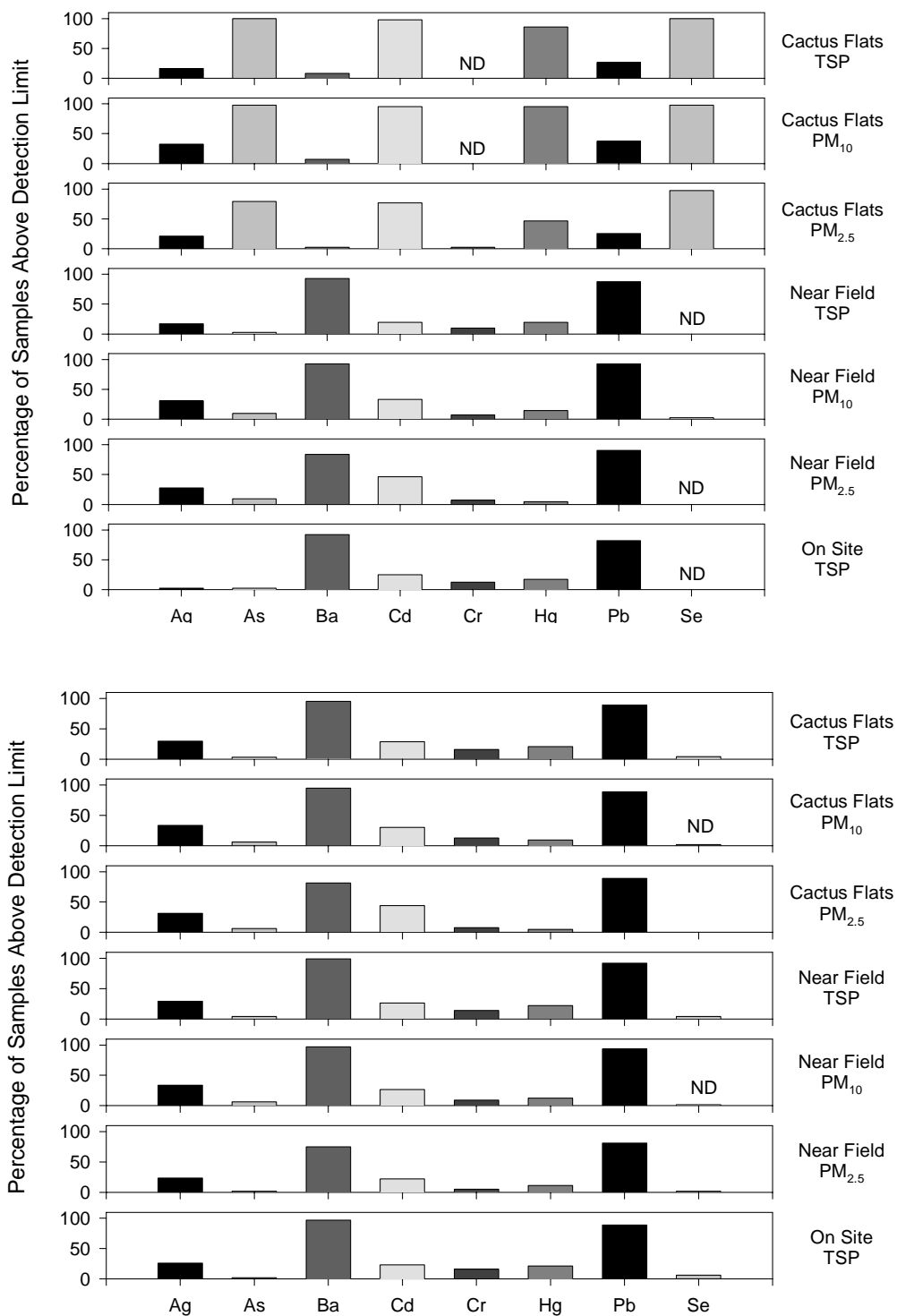


Figure 16. Percentages of Samples with Selected Elemental Concentrations above Detection Limits for Samples Collected from February 1998 to July 1999 (above) and February 1998 to July 2001 (below).

ND = not detected