

CHAPTER 1

Ambient Aerosol Studies for the WIPP-EM

INTRODUCTION

The CEMRC ambient aerosol monitoring studies focus on both man-made and naturally-occurring radionuclides, but special emphasis is given to the members of the actinide series that are major components of the wastes emplaced at the WIPP. The main objective for the aerosol studies presented here, and for the WIPP Environmental Monitoring (WIPP-EM) Program in general, has been to determine whether the nuclear waste handling and storage operations at the WIPP have released radionuclides into the environment around the WIPP. The aerosol program also has included investigations of several non-radioactive, inorganic chemical species because the data for those substances have been found to be useful for interpreting the results of the actinide studies. Summaries of the WIPP-EM aerosol studies have been included in prior Annual Reports from the Center starting in 1997, and two papers specifically based on the WIPP-EM aerosol research have been published in peer-reviewed journals (Arimoto et al. 2002 and 2006).

One element of particular interest for the WIPP-EM is plutonium (Pu, element 94), which has been dispersed throughout the global environment mainly by nuclear weapons tests. When quantified by alpha spectrometry ^{239}Pu typically is determined together with ^{240}Pu , because isotopes are difficult to separate chemically, and they are represented as $^{239,240}\text{Pu}$ (^{239}Pu half-life, $t_{1/2} = 24,110$ yr and ^{240}Pu $t_{1/2} = 6563$ yr). ^{239}Pu and ^{240}Pu also have similar alpha particle energies, about 5.25 MeV.

Another actinide of interest is ^{241}Am ($t_{1/2} = 432$ yr), which is not directly produced in significant quantities during the detonation of thermonuclear weapons but rather is a daughter of bomb-produced ^{241}Pu ($t_{1/2} = 14.3$ yr).

An important finding of the earlier studies was that the activity of Pu and the concentration of Al in aerosols were correlated and this was driven by the resuspension of dust particles contaminated with radioactive fallout from past nuclear weapons tests. Similar results were found for ^{241}Am and Al. Related studies of soils collected on and near the WIPP site have shown that correlations exist among Al and both naturally-occurring and bomb-derived radionuclides including $^{239,240}\text{Pu}$ (Kirchner et al., 2002).

Here we briefly review the methods used for the ambient aerosol studies and then summarize some recent results, highlighting the continuing efforts to evaluate potential releases from the WIPP. In addition to the environmental aerosol studies, aerosol particles also have been and continue to be collected using a fixed air sampler (FAS) in the WIPP exhaust shaft. Results of the FAS studies are presented in the following chapter.

METHODS

The sampling design for the ambient aerosol studies has changed over the course of the project, and detailed information regarding the sampling design has been presented in prior CEMRC reports starting in 1998. Samples for the aerosol/radionuclide studies have been

collected using high-volume samplers (“hivols,” flow rate $\sim 1.13 \text{ m}^3 \text{ min}^{-1}$) since the WIPP-EM program began in 1996. Three long-term aerosol sampling stations have been established; these are On Site, Near Field and Cactus Flats, and each supports a hivol sampler for collecting total suspended particulate (TSP) matter (see Figure 2-1). The Near Field and Cactus Flats stations also supported a second hivol sampler for a time, and those were used for studies of PM_{10} , particulate matter less than $10 \text{ }\mu\text{m}$ aerodynamic equivalent diameter. A fourth set of samples was collected at Hobbs over a period of approximately a year and a half, but the sampling there was discontinued in April 2002.

Until the end of March 2002, both low-volume samplers (“lovols,” 10 L min^{-1}) and Graseby-Anderson dichotomous samplers (dichots) were used for collection of aerosols for the studies of non-radioactive, inorganic constituents, specifically trace elements and selected water soluble ions. The WIPP-EM underwent major restructuring in FY 2002, and afterwards sampling for the non-radiological aerosol analytes was done using dichots exclusively. In November 2004, the collection of aerosols by dichots was discontinued.

In brief, the sampling strategy for the aerosol/radionuclide studies has been to collect as much particulate material as reasonably practical so as to maximize the chances of detecting the radionuclides of interest. Individual samples typically have been collected over periods of 3 to 5 weeks depending on the rate at which the sample filters become loaded. For these studies, high-volume samples were collected on $20 \times 25 \text{ cm}$ Gelman A/E™ glass fiber filters. Gravimetric measurements of the glass fiber filters were made to determine the mass of

aerosol material that accumulated over the sampling interval.

The high-volume samples were analyzed for selected radionuclides, including ^{238}Pu , $^{239,240}\text{Pu}$ and ^{241}Am following 4 hr of heating in a muffle furnace at 500°C , which drives off organics; dissolution of the material on the filters using strong acids (HF , HCl and HClO_4); and multiple precipitation, co-precipitation, and ion-exchange and/or extraction chromatography steps. The nuclides of interest were precipitated with LaF_3 , deposited onto filters, mounted on planchettes, and counted using an Oxford Oasis alpha spectroscopy system.

The radionuclide data are reported in the following two ways. First, the *activity concentration* is calculated as the nuclide’s activity per unit volume of air sampled (Bq m^{-3}). Second, *activity density* is calculated as the nuclide’s activity per unit aerosol mass collected (Bq g^{-1}).

RESULTS AND DISCUSSION

Summary Statistics

Summary data reported for high volume aerosol samples (hi-vols) are presented in Table 1-1. ^{238}Pu was infrequently detected, with activity concentrations slightly above minimum detectable levels in only six of the 208 samples. $^{239,240}\text{Pu}$ was above detection limits in 205 of the 208 samples. As in prior years, the $^{239,240}\text{Pu}$ activity concentrations showed a strong annual cycle with activities greatest in the spring (Figure 1.1).

During most years studied, the peak $^{239,240}\text{Pu}$ activities generally occur in the March to June timeframe, which is when strong and gusty winds in the area frequently give rise to blowing dust. Some samples taken at Cactus Flats in 1999 and

2000, and at On Site in 2004, exhibited slightly higher $^{239,240}\text{Pu}$ activity concentrations (Figure 1.1) than surrounding data points. The points correspond with higher activity densities as well (Figure 1.2). However, insufficient auxiliary data is available for attributing a cause to this result.

Methods for determining the activity of ^{241}Am were developed by the CEMRC radiochemistry group over a period of years, and the available ambient aerosol data for this nuclide are presented here. The activity concentrations of ^{241}Am (Figure 1.3) in the high-volume samples closely tracked those of $^{239,240}\text{Pu}$ as shown in Figure 1.1. Most notably, strong springtime peaks in ^{241}Am activity concentrations were evident in the samples from 2001 through 2002, and 2004 through 2005. Data from 2003 do not exhibit these springtime peaks. A time series plot for ^{241}Am activity density is presented in Figure 1.4.

In contrast to the actinide data, the aerosol mass loadings at On Site were generally the highest of the three stations with comparable data sets (Table 1-1 and Figures 1.6, 1.7, and 1.8). A timeseries plot (Figure 1.5) shows that the aerosol mass loadings at all stations tend to track one another remarkably well, but that during several extended periods, most noticeably January 1999 to July 2000 and July 2001 to January 2002, the mass loadings at On Site were consistently higher than at the other sites.

As a consequence of the similar $^{239,240}\text{Pu}$ activity concentrations at all stations and the higher mass loadings at On Site, the activity densities at On Site tended to be lower than at Cactus Flats or Near Field (Table 1-1 and Figure 1.6). The combination of $^{239,240}\text{Pu}$ and gravimetric data thus suggest that activities at the

WIPP may in fact generate detectable levels of aerosol particles, but those particles actually contain less $^{239,240}\text{Pu}$ than typical ambient aerosols. These are most probably particles from construction dusts or salt from the underground operations.

NEW DIRECTIONS FOR THE AEROSOL PROGRAM

Operational aspects of the ambient aerosol component of the WIPP EM have changed since the 2003 Annual Report. Whatman 41 sampling began on 1/4/07. 8" X 10" filters are being used on Hi-Q Hi-Vol HVP-3800AFC samplers. These samplers are located at sites 107 and 108 and are directly across from the Hi-Vol glass fiber sampler. The samplers are set at 20 SCFM and are changed approximately every 2 weeks and in conjunction with the glass fiber filters. No gravimetric data is collected from the Whatman 41 filters. It is anticipated that these filters may be used to more directly compare trace and major elemental concentrations to actinide and mass concentrations collected at the same locations. A summary of the latest ambient aerosol sampling program is given in Table 1-2.

SUMMARY STATEMENTS

The results presented here demonstrate that actinide concentrations have not changed significantly since the WIPP began receiving waste. Ambient aerosol samples continue to be collected on a regular basis and will be analyzed and the data reported as time permits.

Table 1-1: Summary Statistics for Aerosol Mass Loadings and Actinide Activities in High-Volume Aerosol Samples

Station		Cactus Flats	Near Field	On Site
Type of Sample		TSP	TSP	TSP
Number of Samples		70	69	69
Aerosol Mass, micrograms per cubic meter	^a N	70	69	69
	Mean	1.37	1.33	1.65
	StdDev	0.57	0.50	0.59
²⁴¹ Am Activity Concentration, Bq m ⁻³	N	33	30	35
	Mean	5.4E-09	4.2E-09	4.4E-09
	StdDev	3.2E-09	2.0E-09	2.2E-09
²⁴¹ Am Activity Density, Bq g ⁻¹	N	32	29	35
	Mean	1.9E-04	1.6E-04	1.4E-04
	StdDev	4.5E-05	5.7E-05	5.2E-05
²³⁸ Pu Activity Concentration, Bq m ⁻³	N	4	1	4
	Mean	3.1E-09	1.5E-09	2.8E-09
	StdDev	2.8E-09		2.0E-09
²³⁸ Pu Activity Density, Bq g ⁻¹	N	4	1	4
	Mean	9.0E-05	3.4E-05	7.1E-05
	StdDev	4.8E-05		2.8E-05
^{239,240} Pu Activity Concentration, Bq m ⁻³	N	70	68	67
	Mean	1.6E-08	1.2E-08	1.3E-08
	StdDev	1.2E-08	7.7E-09	8.2E-09
^{239,240} Pu Activity Density, Bq g ⁻¹	N	69	68	68
	Mean	5.3E-04	4.5E-04	4.0E+00
	StdDev	2.1E-04	1.4E-04	2.1E-04

^aN stands for number of samples with masses or activities above detection limits.

Table 1-2: Aerosol Sampling Status for WIPP EM (January 2007)

Site	^a Sampler	Analyses	Frequency	Comments
Station A (Exhaust Shaft)	PM ₁₀ -Shrouded Probe	Mass, Gross Alpha and Beta Activities, Trace Elements, Gamma Emitters, Actinides	Daily	Monthly Composites
Station B (Post Filtration)		Gross Alpha and Beta Activities, Actinides	Weekly	Monthly Composites
Cactus Flats Near Field On Site	TSP-HI VOL Glass Fiber Filter	Mass & Radionuclides	^b Variable	Continuous
	^c TSP-HI VOL Whatman 41 Filter	Elemental	Variable	Continuous

^aSampler types are as follows: PM₁₀-Shrouded Probe = particles greater than 10 µm diameter (50% cut-size), TSP-HI VOL = high volume total suspended particles.

^bSamples are changed when the flow drops to 90% of original for the 2-stage pumps.

^cTSP-HI VOL Whatman 41 Filters are collected at Cactus Flats and Near Field.

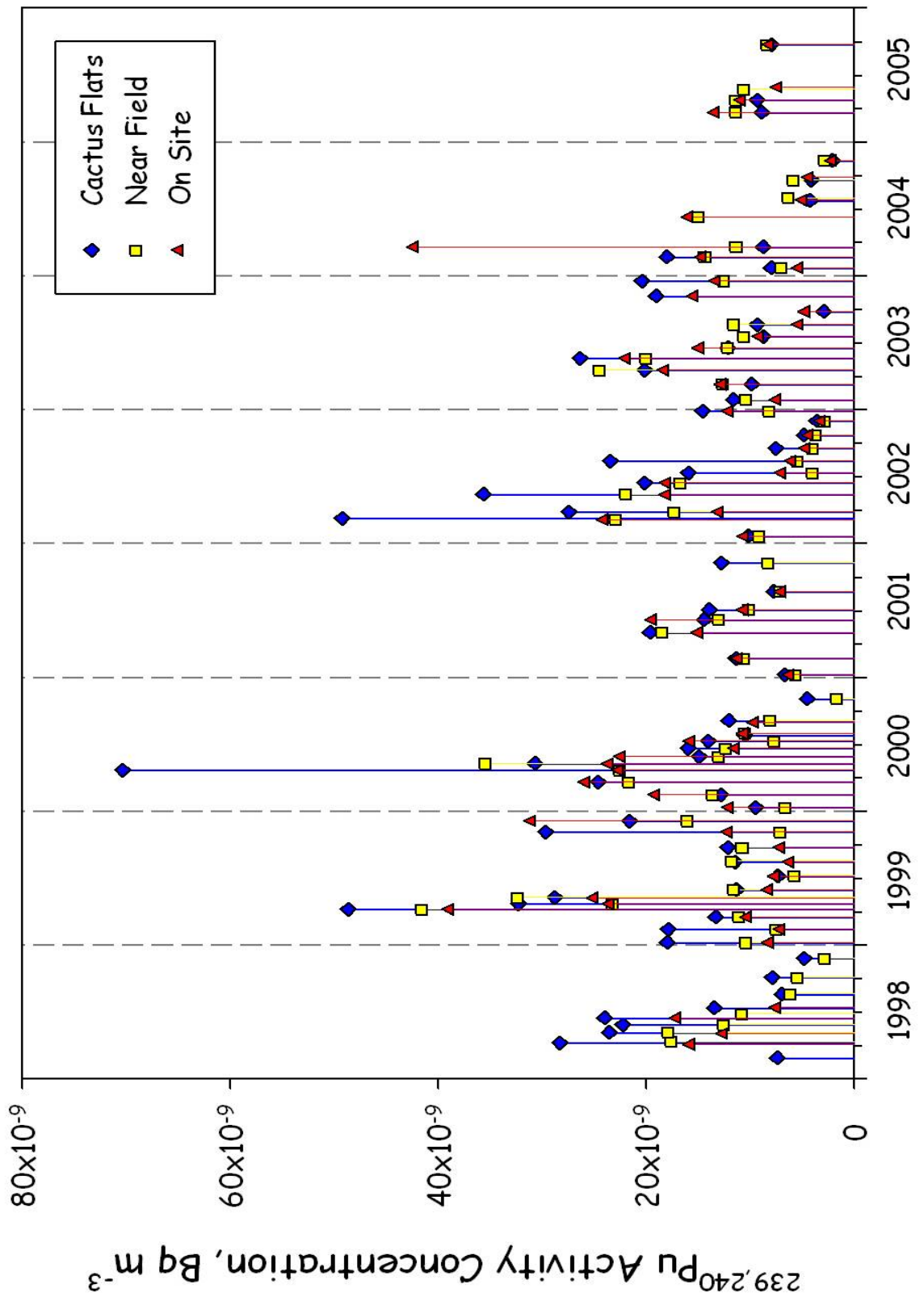


Figure 1.1: High Volume Ambient Aerosol $^{239,240}\text{Pu}$ Activity Concentration

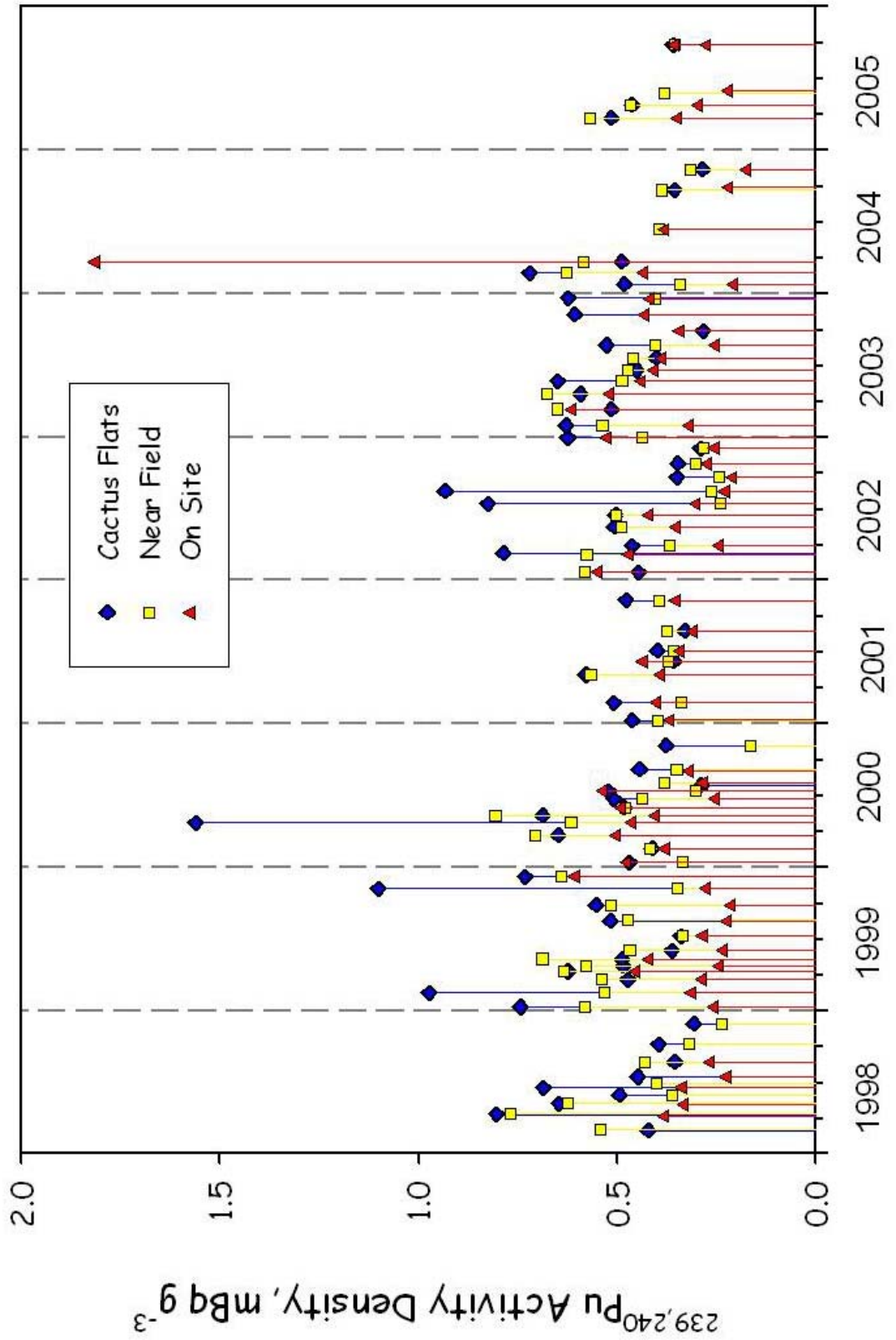


Figure 1.2: High Volume Ambient Aerosol $^{239,240}\text{Pu}$ Activity Density

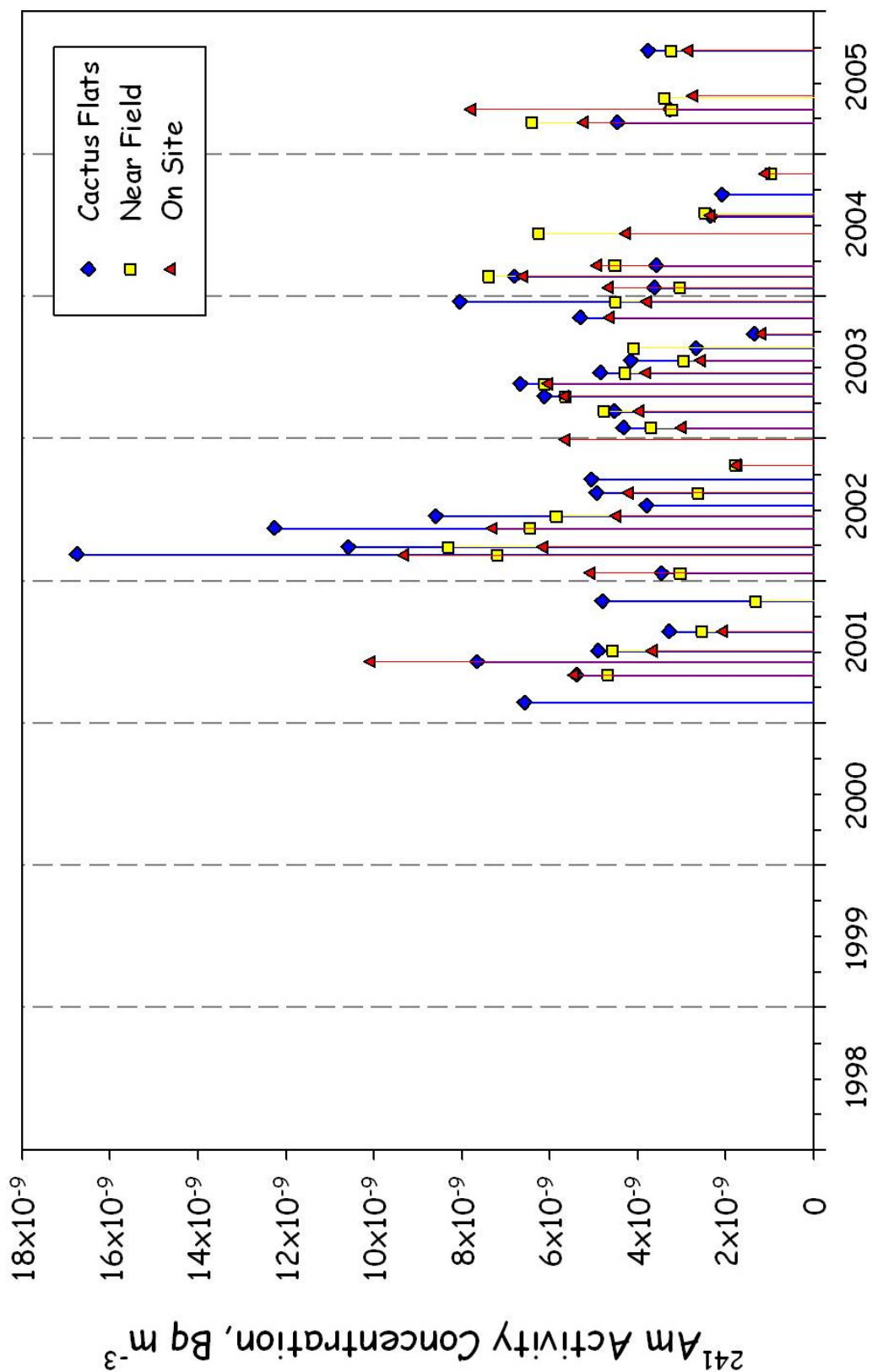


Figure 1.3: High Volume Ambient Aerosol ²⁴¹Am Activity Concentration

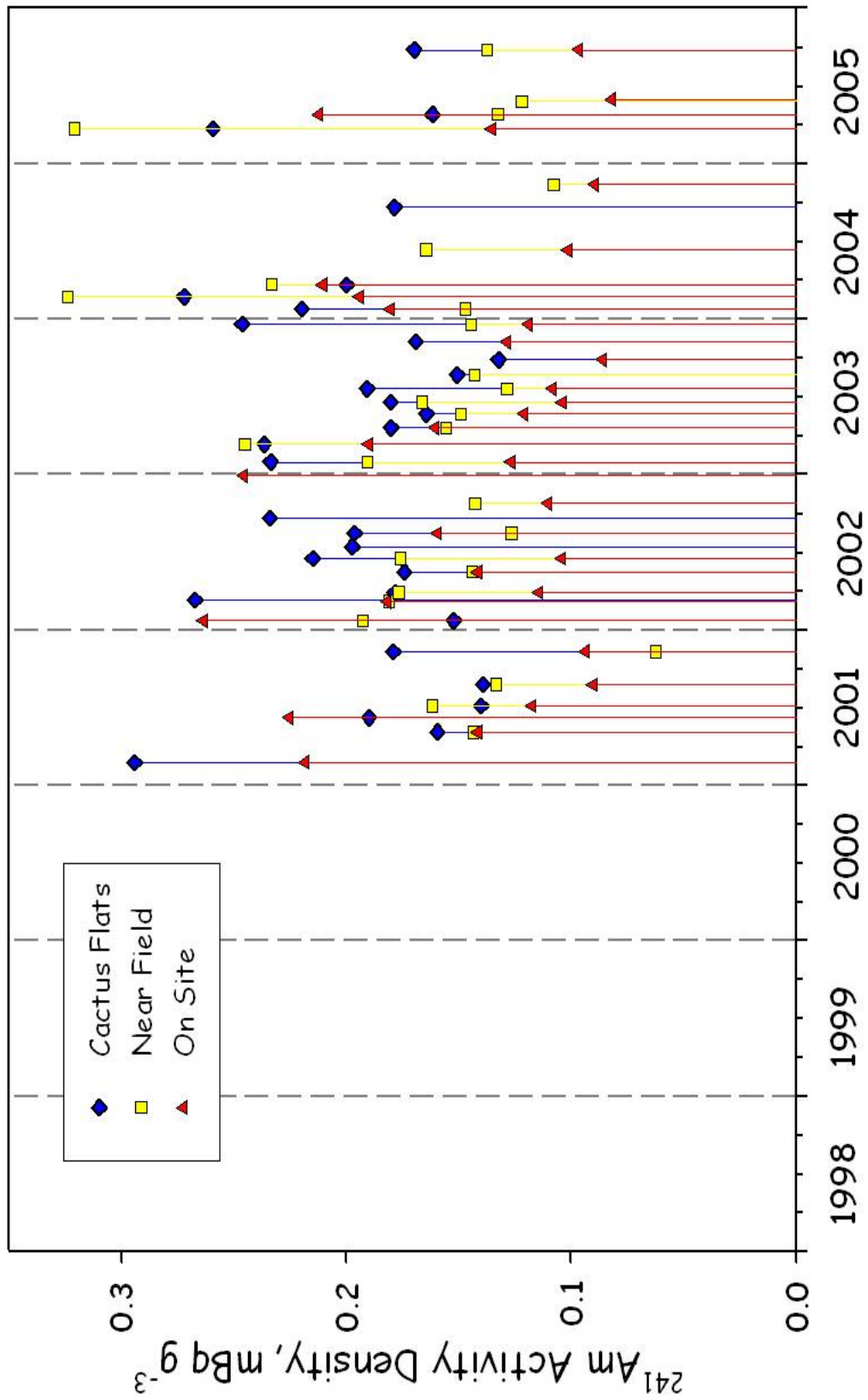


Figure 1.4: High Volume Ambient Aerosol ²⁴¹Am Activity Density

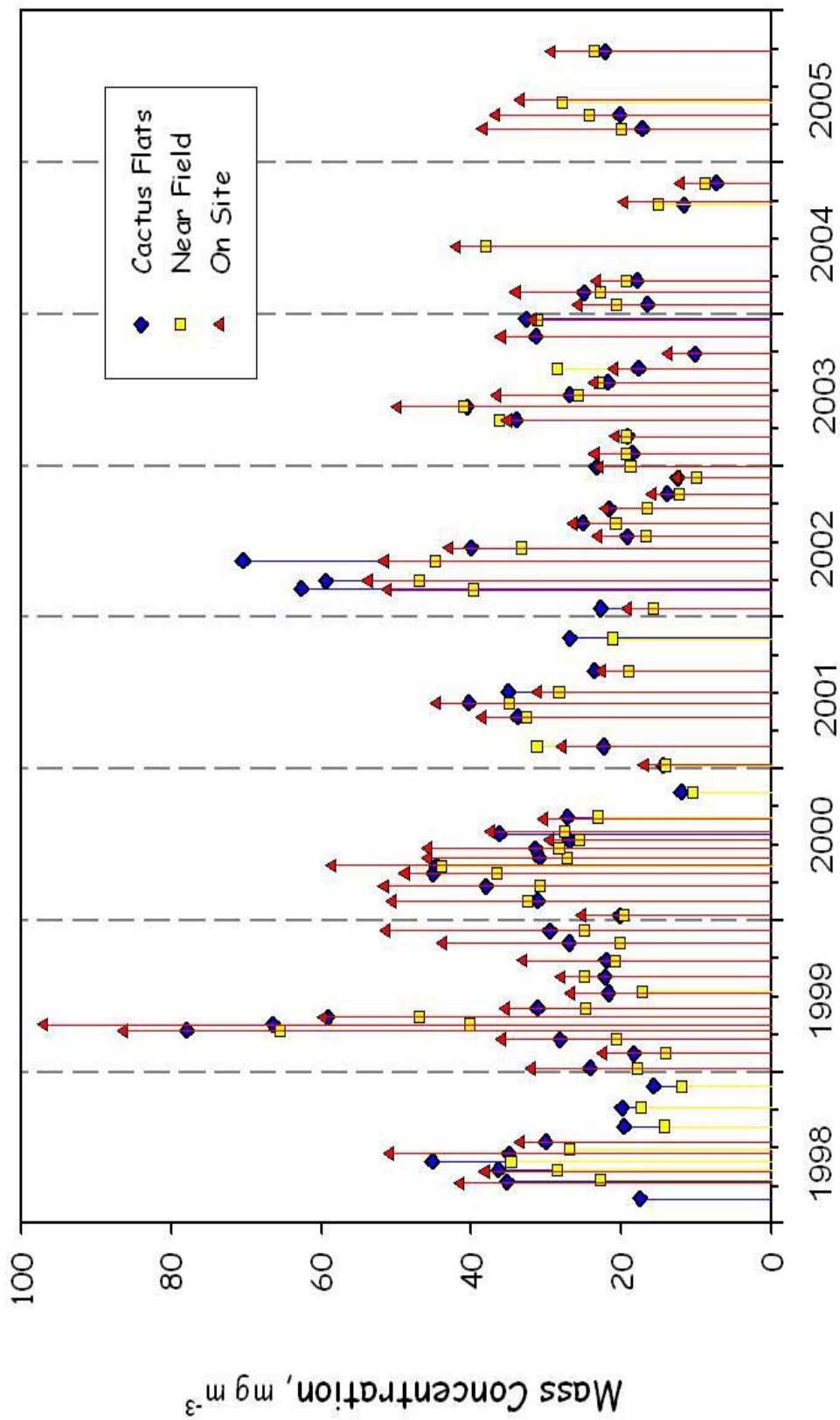


Figure 1.5: High Volume Ambient Aerosol Mass Concentrations

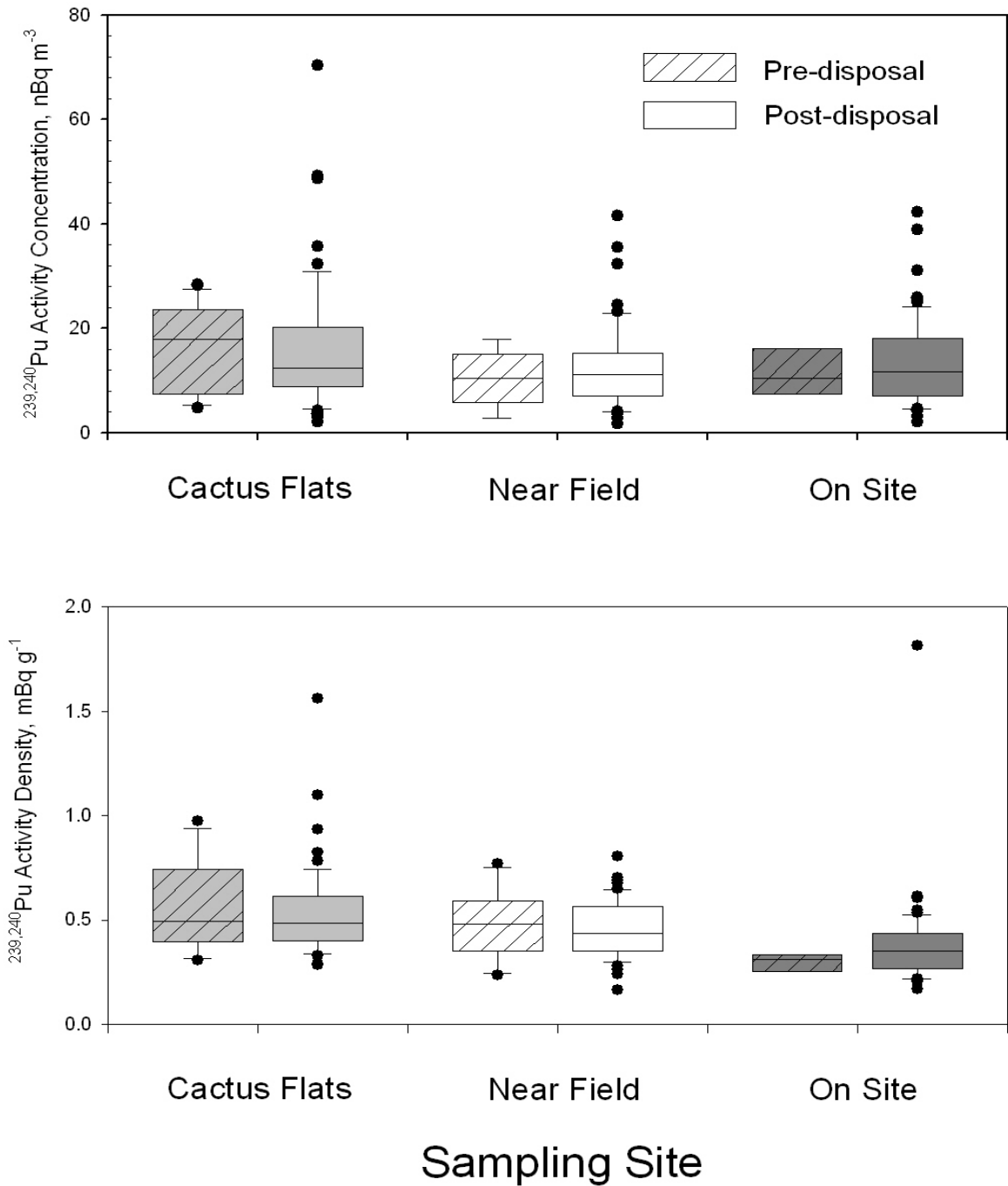


Figure 1.6: Whisker Plots Showing High Volume Ambient Aerosol $^{239,240}\text{Pu}$ Activity Concentrations and Densities

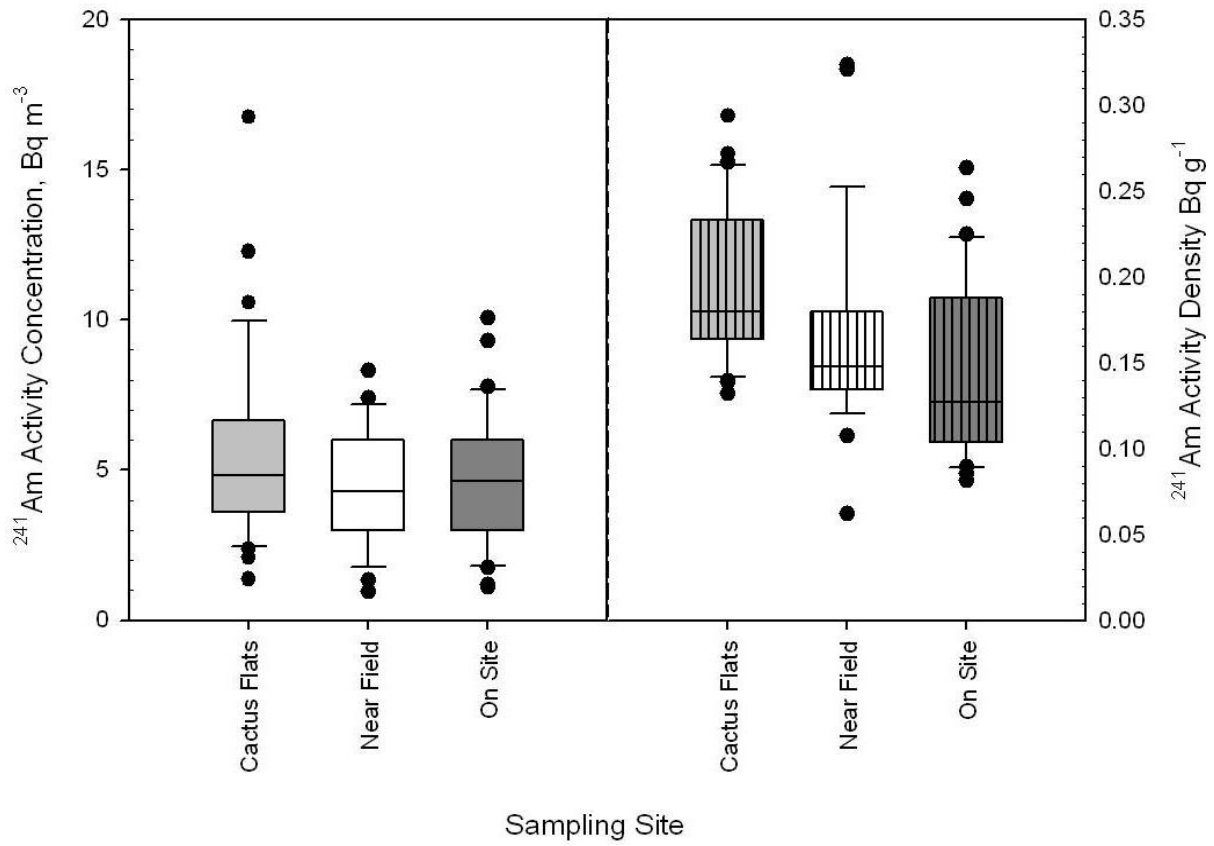


Figure 1.7: Whisker Plots Showing High Volume Ambient Aerosol ^{241}Am Activity Concentrations and Densities

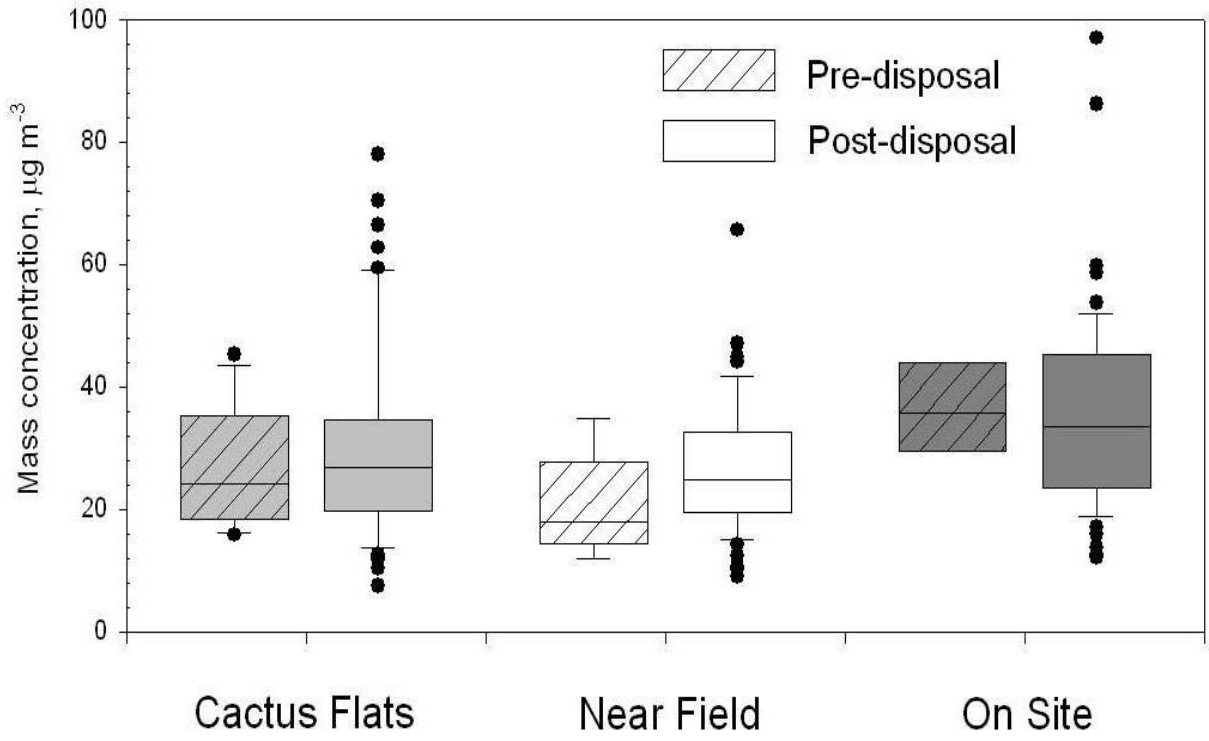


Figure 1.8: Whisker Plots Showing High Volume Ambient Aerosol Mass Concentrations