STUDIES OF PLUTONIUM AEROSOL RESUSPENSION AT THE TIME OF THE MARALINGA CLEANUP

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JOSEPH H. SHINN

INTRODUCTION

At the former nuclear test site at Maralinga, South Australia, soil cleanup began in October 1996 with the objective to remove the potential for residual plutonium (Pu) exposures to the public. In this case the cleanup was to restore access to the closed test site. The proposed long-term land use was primarily to be a hunting area for Pitjantjatjara (Aboriginal) people, but also presumably to be available to the public who might have an interest in the history of the site. The long-term management objective for the site was to allow casual use, but to prohibit habitation.

The goal of this study is to provide an evaluation of the Maralinga soil cleanup in terms of potential long-term public inhalation exposures to particulate Pu, and in terms of a contribution to planning and conducting any such soil Pu-cleanup. Such cleanups might be carried out for example, on the Nevada Test Site in the United States. For Pu that has been deposited on the soil by atmospheric sources of finely divided particles, the dominant exposure pathway to humans is by inhalation. Other exposure pathways are less important because the Pu particles become oxidized into a nearly insoluble form, do not easily enter into the food chain, nor are they significantly transferred through the intestine to the bloodstream should Pu become ingested.

The purpose of this report is to provide results of the Pu resuspension measurements made before, during, and after the Pu cleanup at Maralinga, to compare these against similar measurements made elsewhere, and to interpret the results as they relate to potential long-term public exposures. (Exposures to Pu in dust plumes produced by mechanical disturbance during cleanup are considered short-term, unlikely to be significant for purposes of this report, and are not included).

A considerable amount of research had been conducted at Maralinga by the Australian Radiation Laboratory, now the Australian Radiation Protection and Nuclear Safety Agency (ARPANSA), prior to the cleanup (Johnston et al, 1992, Williams 1993, Johnston *et al* 1993, Burns *et al* 1994, Burns *et al* 1995). ARPANSA staff made major contributions to delineate the areas with Pu in the soil, to determine the degree of secondary soil contamination by fission products from nuclear testing, to measure Pu resuspension by wind erosion of the undisturbed soil, and to prepare assessments of the human health risk from residual soil Pu.

In addition, ARPANSA supported the Maralinga cleanup to assure compliance with criteria set by an independent technical advisory committee. During the cleanup ARPANSA monitored the residual Pu in the soil and certified that the cleanup was complete according to the criteria. It was not the reduction in potential inhalation exposure that usually was the main driver of the cleanup, but the requirement to also remove individual hot particles and

fragments. It is the residual microscopic particles of Pu in the soil, however, that have the potential for long-term human exposure.

The resuspension of respirable-size Pu particles has been studied with specialized equipment at the Nevada Test Site (Gilbert *et al* 1988a, Gilbert *et al* 1988b, Shinn *et al* 1989, and Shinn 1992), and at Bikini and Enewetak in the Marshall Islands (Shinn *et al* 1997). These efforts were in large part contributed by the Health and Ecological Assessment Division, University of California, Lawrence Livermore National Laboratory (LLNL). The study reported here is a collaboration between ARPANSA and LLNL, and was jointly supported by the United States Department of Energy, and the Commonwealth of Australia Department of Primary Industry and Energy.

METHODS

Previous experience has shown that there are site-specific variables which render uncertain the ability to predict Pu inhalation exposure, and that these variables should change as a direct result of mechanical disturbance, even after the short-term effects of dust plumes no longer persist. The definitions that follow are discussed in more detail in National Council on Radiation Protection and Measurements Report NCRP 129, 1999. The most common expression of Pu resuspension is a coefficient known as the "resuspension factor", S_f. It is defined as the ratio of *C*, the concentration of Pu in air (Bq m^{-3}), to *D*, the surface deposition of Pu (Bq m^{-2}):

$$S_f = C / D \hspace{4cm} [1]$$

The concentration, *C*, is dependent upon *A*, the activity in top soil (Bq g^{-1}), upon M, the *TSP*, or the amount of suspended dust expressed as total suspended particulates (g m^{-3}), and upon *Ef*, the enhancement factor, which is defined as the ratio of the activity in the suspended aerosol to the activity in the top soil (regardless of the relative difference in fundamental particle sizes of the two media):

$$C = E_f A M \hspace{4cm} [2]$$

While the activity in the soil, *A*, should be reduced by the cleanup, the equilibrium *TSP*, and the enhancement of activity in the suspended dust might increase. Furthermore, the size distribution of *TSP*, or the size distribution of Pu activity on the *TSP*, could be changed to the degree that the potential inhalation and lung retention of particles would change as a result of cleanup. The lack of experience with these effects of cleanup activities required that detailed measurements be undertaken.

In addition, a consideration in the long-term potential inhalation exposure is how stable the soil surfaces might be after cleanup. The vertical flux due to wind erosion of soil-borne respirable Pu particles (Bq m^{-2} s^{-1}), is constant with height, *z*, in the surface layer of the atmosphere and is given by

$$Q = K \{dC/dz\} \hspace{4cm} [3]$$

where the flux, Q, is proportional to the vertical gradient of the concentration, dC/dz, and the turbulent eddy diffusivity, K. If, for example, the vertical gradient of concentration increased drastically after cleanup it would be an indication of an unstable surface. The dependency of K on surface conditions and wind speed is also important:

$$K = u_* \, k \, z \, / \, \phi \qquad [4]$$

Here ϕ is a function of atmospheric stability, and k is the Karman constant (equal to 0.4). The friction velocity u_* is proportional to the wind speed (u_2, taken at the reference height of 2 m), and the square root of a "drag" coefficient c_d:

$$u_* = \{ c_d \}^{1/2} \, u_2 \qquad [5]$$

Thus, for example, a drag coefficient made "smoother" by the soil cleanup may decrease the turbulent eddy diffusivity and also increase the effective stability of the soil surface by causing reduced dust flux due to wind erosion.

We have investigated these potential effects through a measurement program at Maralinga as follows.

1. Measurement of the Particle Size of Suspended Dust and Suspended Pu

An optical particle counter (Climet, model 208) has been modified for field use through the addition of an isokinetic sampling inlet fixed on a wind vane to keep the inlet faced directly into the wind. Also, a log-linear amplifier and a 128-channel, multi-channel, analyzer were used to count and aerodynamically size individual particles entering the inlet during one-minute time periods. In order to approximate isokinetic sampling, three different size inlets were used and changed for the appropriate ranges of wind speed during the one-minute sampling periods. The optical particle counter was effective over the range from 0.5 to 11 μm, and was calibrated with latex spheres at 0.5 μm, 1.1 μm, 3.3 μm, and 7 μm aerodynamic diameters. In addition to the optical particle counter, a small laser-nephelometer (AQ-10) was used to continuously monitor dust, and responded linearly to *TSP* with a DC output generated by photometrically-measured bulk-scattering of particles drawn into the optical chamber. The chamber on the AQ-10 could not be kept optically clean, however, and gradually the scattering baseline with clean air drifted with time. This baseline was determined by numerical processing of the digitally-recorded signal, and the effects of this drift removed.

Cascade impactors (Graseby-Anderson, model 65-000) were operated to collect particles for size determination of suspended mass and Pu. These were five-stage, jet-plate type, collecting on Whatman 41 cellulose fiber media. The cascade impactors (CI) had a flow rate of 34 m^3 h^{-1} and were deployed in groups of two or four. Mass-determination was done by weighing; preceded by conditioning the fiber media at constant temperature and humidity before and after the samples were collected. Pu determination was done by radiochemical analysis of the Pu isotopes and americium (Am) present (discussed in part 2 below).

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2. Measurement of the Concentration of Pu in Air

Particles were collected by two additional methods that have increased the detectability for low levels of Pu particles in air. The common, "high volume" (HV) air samplers (General Metal Works) collecting on rectangular Whatman 41 (8×10) cellulose fiber filters were operated at 85 m^3 h^{-1}, usually in pairs. These units had no flow controllers but were monitored once or twice daily and the flow rates recorded and readjusted. There was no particle separation stage on these units (e.g. PM-10), and they were used with the tilt-roof access commonly used in *TSP* measurements. This type of HV air sampler has a long history of use for Pu measurements in air and served as a "reference method". The cellulose fiber filter media was preferred for dissolution or ashing prior to Pu determination, and Whatman 41 has a high collection efficiency for solid particles when the flow rates are large enough to sustain a face velocity over 0.25 m s^{-1}. Mass was determined for *TSP* in the same manner as for cascade impactors. Miniature HV systems were also utilized to measure the vertical gradient of Pu and *TSP*. Four of these units were run from a single HV motor and they were placed in a vertical array (VAS) at 0.25, 0.5, 1, and 2 m heights. Flow rates were periodically checked with a calibrated venturi unit.

"Ultra high volume" (UHV) air samplers were used to assure detectability of extremely low levels of Pu in air. Two of these air samplers were designed and constructed by LLNL, to collect particles at a rate from 800--1300 m^3 h^{-1} onto a one-m^2, cellulose fiber filter cut from Whatman 41 rolls. The UHV blowers were high-pressure centrifugal fans driven by 5-hp electric motors. The filter beds were stainless steel cylindrical screens with a vertical axis (effective screen height 0.62 m by 1.74 m circumference with an 0.12-m inefficient overlap) and three bands to hold the filters in place. Flow rates were monitored through manometers on pitot tubes mounted into the 0.2-m diameter pipe sections used to deflect fan exhaust. Calibration of this flow monitor was performed against a 1700 m^3 h^{-1} Merriam laminar flow element (LFE) on the loaded intake of the UHV. Calibration equations of LFE versus the flow monitors for filters under load were linear, but discharges at the exhaust were increased over intake flow due to small air leaks into the fan shroud, so the equations had a nonzero offset and a non-unity slope. The UHV filters were ashed, and ash weights were corrected by subtracting ash weights from blank filters to determine residual *TSP*. The UHV filters had sufficient particles collected in 48 h so that gamma spectroscopy of the compressed filters could be used to quantify the weak gamma peak of ^{241}Am at 60 keV. These measurements of ^{241}Am enabled rapid and economical estimation of Pu in air via the known Pu/Am ratio (discussed below), but also allowed the radiochemists to be prepared for the particular level of Pu that might be expected in subsequent analysis of UHV and other filters. After gamma spectroscopy, the UHV filters were analyzed by radiochemistry. At the conclusion of the rehabilitation at Maralinga, the UHV units were used to measure extremely low Pu concentrations in air, which demanded the use of accelerator mass spectroscopy methods (McAninch et al, 1999) to detect Pu on these special filters.

The Pu/Am ratio as observed is defined as the ratio of ^{239,240}Pu to ^{241}Am, and had been earlier determined in the soil by ARPANSA (Burns *et al*, 1994). The technical advisory group considered these Pu/Am ratios when the cleanup criteria were adopted. Because of the ease of measurement of ^{241}Am in the soil *in situ*, the Maralinga cleanup criteria were written to use that measurement, exclusively, to certify that cleanup had been complete. ARPANSA used both a mobile unit for surface gamma spectroscopy with a high purity germanium diode detector on a telescoping boom, and a small truck with four sodium iodide detectors mounted

on the bumper for ^{241}Am (60 keV) measurement. Both of these vehicles had geographical positioning systems for accurate mapping.

In addition, all air samples collected by us were analyzed for ^{239,240}Pu, ^{238}Pu, and ^{241}Am by radiochemistry, and the Pu/Am ratio (^{239,240}Pu/^{241}Am) determined. Thus measurements of ^{241}Am either on air samples or on bare soil by gamma spectroscopy could be used to estimate Pu via the Pu/Am ratio.

In both the ARPANSA and LLNL laboratories, gamma spectroscopy for ^{241}Am was applied to air filters compressed into standard geometry containers that had been routinely used, and these samples were compared against reference ^{241}Am standards of the same geometry. Six of the Maralinga air samples were split and interchanged between ARPANSA and LLNL to compare ^{241}Am gamma measurement quality.

Pu radiochemistry was performed the same on all air samples (UHV, HV, cascade impactors, and vertical array samplers). After weights of collected mass were determined, and estimates made of the Pu levels based on soil activity levels and Pu/Am ratio, the samples were submitted for analysis.

3. Analytical Methods

For transuranic isotopes, ^{239,240}Pu and ^{241}Am, special chemical methods were required. On filter media, the isotopes were recovered by acid total-dissolution, ion-exchange separation, and electrodeposition, and measured by alpha spectrometry using internal chemical yield tracers of ^{242}Pu and ^{243}Am. Details of the protocol are provided in Wong *et al*, 1995. Quality assurance was carried out through adherence to established protocols, (Kehl *et al*, 1995) and by use of quality control procedures (blank filters, control filters-carried to the site and back without use, and standards). Analytical accuracy has been maintained among these methods for the analysis of Pu (from atmospheric fallout) in shallow or deep marine sediments or in soil and sediment samples collected from the Pacific Test Sites at Bikini and Enewetak Atolls (close-in fallout). The method has also been used in the analysis of Pu in NIST Radioactivity Standard Reference Materials (Rocky Flats Soil and Columbia River Sediment) and in IAEA interlaboratory comparison samples. The alpha chemistry precision of the air filters was better than 0.005 Bq/ sample for ^{239,240}Pu and ^{241}Am, and the minimum detectability was 0.005 Bq/ sample.

4. Measurement of Pu Emission Rate

Measurements of the vertical gradients of *TSP* and *C* were accomplished by a "vertical array" of air samplers at four heights (they varied only slightly from the heights 0.25 m, 0.5 m, 1 m, and 2 m). There were two sets of four each, inverted filter holders with Whatman 41 filters 0.09-m diameter, and a flow rate of 10 to 20 m^3 h^{-1}. These were deployed either at separate locations or as duplicates, and air flow-rates were monitored once or twice daily by means of a calibrated venturi device. These smaller filters were handled in the same manner as HV or cascade impactor filters. In order to facilitate calculation of the gradients, a nondimensional form of the gradient was used (Anspaugh *et al*, 1975):

$$p = \{z/c\} \, dC/dz \qquad\qquad [6]$$

From past experience, the value of p tended to remain constant for a given experiment, and was obtained by least-squares, linear fit to the log C, log z transformed variables. Values of p have been obtained as a measure of soil surface stability at many other sites where resuspension of Pu has been observed, and was determined for *TSP* in the same manner. To calculate the flux, Q, the mean value of C (or *TSP*) was used at the height of 1.2 m, and the diffusivity, K, was determined at 2 m height and the value adjusted for height.

To obtain values of the diffusivity, K, an automatic meteorological station (Campbell Scientific Inc, model CR-10) was installed, and meteorological data were recorded in ten-minute averages for the duration of the air-sampling period. The primary method of determining the diffusivity, K, was by measuring the sensible heat flux and then dividing the measured eddy correlation by the vertical temperature gradient over the same period. Sensible heat flux was measured by the eddy correlation of vertical turbulence, *w'*, and temperature fluctuation, *T'*, using a vertical, one-dimensional, sonic anemometer (Campbell Scientific Inc, model CA-27) with a fast response thermocouple,. This is a direct method, and assumes only that the diffusivity for Pu and *TSP* is the same as that of diffusivity for heat:

$$K = - <w'T'> / \{dT/dz\} \qquad [7]$$

The automatic meteorological station recorded the wind speed, wind direction, and as a quality check, recorded sensors that measure the surface energy budget by the Bowen ratio technique, and determine the sensible heat flux indirectly. This same station also recorded the *TSP* monitoring data from the AQ-10. Ten-minute summaries of all signals were recorded and downloaded daily into a laptop computer in the field.

5. The Experimental Strategy

A measurement strategy for the resuspension measurements at Maralinga was to compare two sites at a time. (A map of Maralinga as well as the detailed layout of cleaned plots is shown in Appendix 1.) Each site was chosen to be representative of a particular condition. In November 1996, a control site was established at a location known as FW, on the west deposition plume at Taranaki. This site was unique in that it had a non-hazardous but measurable amount of Pu in the soil. It had been the location of earlier resuspension measurements by ARPANSA, and it had never been disturbed by the ploughing operations in the attempts at soil stabilization by the British during Operation Brumby in 1967. During the period in November, 1996, a cleaned plot (Lot 37) in the northeast plume at Taranaki, representing a location that had been cleaned but not yet stabilized, was also studied. This site was also near the FNE location where resuspension measurements were made by ARPANSA prior to the cleanup. In March 1997, a cleaned plot (Lot 19) in the north plume at Taranaki was investigated, representing another recently cleaned but not yet stabilized location. This was also near the FN site where ARPANSA had measured resuspension prior to the cleanup. At the same time in March 1997, a site was studied to the north just outside of Lot 18, the north end of the cleaned north plume at Taranaki, where soil had blown out of Lot 18 during a dust storm in November 1996. This site represented a possible recontamination area, with unstabilized, unconsolidated, recent deposits. Then, in May, 1997, the UHVs were moved to a situation similar to the latter case. This was a simpler study (only UHVs) and compared the northernmost, cleaned lot in the northwest plume at Taranaki (Lot 1) to a site beyond the north end of the same plot. Here again, soil had blown out of Lot 1 during a dust

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storm, and could possibly represent a recontamination area with unstabilized and unconsolidated deposits.

In October 1997, the Taranaki surface cleanup had been completed, so that resuspension studies moved to the TM-100 cleanup location, where the type of contaminating-Pu dispersal was known to be different from that at Taranaki. A site was chosen in the middle of a cleaned area representative of the cleaned north plume (Lot 12) at TM-100. A second site was chosen at TM on the west side of Lot 12 on an uncleaned area with a non-hazardous, but measurable amount of Pu. This uncleaned area represented an undisturbed control site for TM-100 and was surveyed for [241]Am by ARPANSA.

Rehabilitation of cleaned areas were investigated during April and May 2000, when the UHV units were operated consecutively at Taranaki, TM-100, and Wewak sites to determine the Pu concentrations in air as a measure of rehabilitation success. As a control, 6 HV units were operated simultaneously and continuously at Maralinga Village, near the building containing the whole body counter. Another control was the GARD air sampling system operated at ARPANSA in Melbourne during the same time period.

RESULTS

1. Aerosol Particle Sizes

Particle size studies that were conducted with the optical particle counter (on Lot 19 and the recontaminated site north of Lot 18 in Taranaki North plume) showed that there was considerable detail in the *TSP* distribution with particle size (aerodynamic diameter). In the particle diameter range of 0.5 to 10 μm it was possible to detect at least 2 minor modes and one major mode; for example, see Fig 1. But the dominant mode was near 3.9 μm with a minor mode between 5 and 6 μm and a minor mode between 8 μm and 10 μm. Interpretation of minor-mode data and how they vary was undertaken in detail by Bertoldo 1998, See Appendix 2.
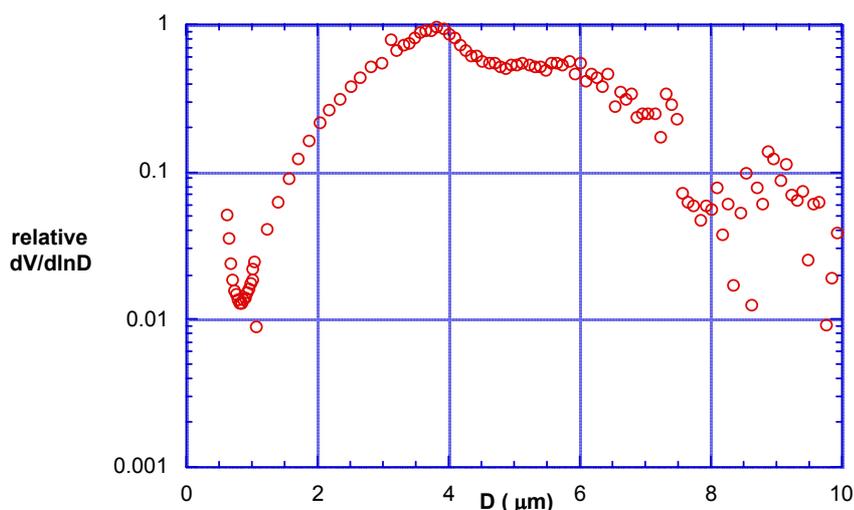

Fig 1. Typical suspended particle-size distribution, Lot 19

The cascade impactor data could not distinguish the minor modes at these sites, but they showed nevertheless that the mass median aerodynamic diameter of *TSP* (MMAD) was 3.0 μm on Lot 19 and 3.2 μm on Lot 18. The MMAD on the historically unploughed and uncleaned FW control site was 4.0 μm while the activity median aerodynamic diameter for Pu (AMAD) was 4.7 μm. All of these cascade impactor derived median values tend to agree with the dominant mode found by the optical particle counter. See Table 1. The particle sizes fall for the majority in the middle of the respirable size range, 1–10 μm.

TABLE 1. OBSERVED CHARACTERISTICS OF AIRBORNE PLUTONIUM

Site	Mean Air Activity (Bq/g)	AMAD* (μm)	MMAD** (μm)	Easting	Northing	Comments
Taranaki West	1	4.7	4			FW site, undisturbed
Taranaki North #19	2.2	2.8	3	750207	6691078	Cleaned site
Taranaki North #18	2.3	6.9	3.2	750145	6691313	Uncleaned, recontaminated
Taranaki Northeast	2.3	3.7	4.6	750920	6690870	Cleaned site
TM-100	0.0167	2.9	1	768060	6679903	Undisturbed site

* Activity Median Aerodynamic Diameter from high volume cascade impactors.
** Mass Median Aerodynamic Diameter from high volume cascade impactors.

2. Comparison of Pu/Am, Resuspension Factors, and Enhancement

We found good agreement between the Pu/Am ratios observed by LLNL during the 1996–1997 investigations and those recommended by Williams, 1993, for Taranaki sites in the IAEA dose assessment report. These measurements were a little different, in that the Pu/Am ratio from LLNL alpha spectroscopy was ^{239,240}Pu to ^{241}Am while Burns *et al*, 1994, were able to separate ^{239}Pu and ^{240}Pu, however, the results should be fairly comparable. The ratio of ^{239}Pu to ^{241}Am from Williams, 1993, is the Pu/Am reference value shown in Table 2. The observed standard deviations from the many samples in 1996-1997 are shown (sd in Table 2).

We found a much lower Pu/Am ratio for TM-100, however, and a very low enhancement factor (last row in Table 2). It could well be that our observations were made on a site that was not characteristic of the whole TM-100. In any case the dose assessment would have an added safety factor here.

TABLE 2. DOSE ASSESSMENT Pu/Am RATIOS AND ENHANCEMENT:
COMPARISON TO OBSERVED VALUES

Site	Reference* Pu/Am	Observed** Pu/Am	Reference* Enhancement	Observed Enhancement E_f
Taranaki West	8	6 (sd 2.5)	6	2
Taranaki North	8	10 (sd 3.7)	6	2
Taranaki Northeast	9	6.4 (sd 0.7)	6	6 (during storm)
TM-100	20	3 (sd 1.7)	4	0.0003

* G.A. Williams, IAEA-TECDOC-755, 1993

**Reference Pu is Pu-239, but Observed Pu includes both Pu-239 and Pu-240. Pu-239 was the dominant isotope in this mix, typically greater than 80%. Am is Am-241, a daughter isotope of Pu. Variability is shown by standard deviation, sd.

The resuspension factors obtained during the period 1996-1997, combining all HV, UHV, and CI data offer useful comparisons to data from Johnston *et al* (1993)—see Table 3. For FW site at Taranaki West, which was the undisturbed control plot for Taranaki, we obtain nearly the same results. But for sites that were cleaned in the Taranaki North and Northeast plumes the resuspension factor increased by an order of magnitude as a result of disturbance and removal of topsoil, as expected. During a dust storm in November 1996, the resuspension factor was temporarily increased, most likely a result from advected Pu aerosols from beyond the plot boundary. At TM-100 the cleaned site had a resuspension factor increased by a factor of more than 2 compared to the undisturbed control plot. The mean mass loading of the atmosphere, *TSP,* had similar values observed in 1996-1997 to those reported by Johnston *et al*, 1993, which had a range 14-41 µg/m^3. From comparison to the reference values of resuspension factor and mean *TSP* that were recommended in the IAEA dose report (see the first row of Table 3), it is clear that the observations show how conservative the assumption was for dose assessment purposes.

TABLE 3. RESUSPENSION FACTOR AND TSP FROM DOSE ASSESSMENT: COMPARISON TO OBSERVED VALUES

Site	Resuspension Factor (1/m) 1993***	Resuspension Factor (1/m) 1996-1997	Mean TSP (µg/m^3) 1996-1997	TSP Variability**	Comment
Reference Values*	6.00×10^{-08}		1000	(not applicable)	Conservative estimate
Taranaki West	3.3×10^{-10}	3.0×10^{-10}	14	GM 13, GSD 1.6	Undisturbed control
Taranaki North, #19	6.0×10^{-11}	4.0×10^{-09}	28	GM 23, GSD 1.9	Cleaned site
Taranaki North, #18	6.0×10^{-11}	1.3×10^{-09}	47	GM 36, GSD 2.1	Undisturbed, recontaminated
Taranaki Northeast	1.2×10^{-10}	1.4×10^{-09}	14	GM 13, GSD 1.6	cleaned site
Taranaki Northeast, (storm)	1.2×10^{-10}	5.9×10^{-07}	967	GM 903, GSD 1.5	24-hour dust storm
TM-100 undisturbed	UNKNOWN	2.8×10^{-11}	21	SD 9	Undisturbed control
TM-100 cleaned site	UNKNOWN	6.6×10^{-11}	15	SD 3	Cleaned site

 * G. A. Williams, IAEA-TECDOC-755, 1993
**Variability is shown by standard deviation, SD, or geometric standard deviation, GSD, of a log normal distribution. GM is the geometric mean. This is combined data of all HV, UHV, and CI.
*** Johnston *et al*, *J. Environ. Radioactiv.*20,1993, 117-131.

3. Variability and Uncertainty in Observed Variables

Pu activity in the air and the resulting air concentrations vary in time and space. A complete discussion of variability of the terms in the equations for inhalation exposure (Eqns 1 and 2 in

Methods section) is given in the report NCRP 129, 1999. During any particular observation period, the LLNL sampler systems showed variation as well. It is not possible to separate sources of variability since observational problems combine with environmental factors to produce the total variability. A summary of the variability observed during 1996-1997 is presented in Table 4. In most cases, the concentrations of Pu in air, and *TSP* in air approximately follow a log normal distribution. The results are that the mean is greater than the median, and deviation is expressed as the geometric standard deviation, or GSD. The properties of a log normal distribution are that the cumulative 84th percentile is a multiple of the median times the GSD, and the cumulative 16th percentile is given by the median divided by the GSD.

TABLE 4. OBSERVED VARIABILITY OF PLUTONIUM CONCENTRATIONS IN AIR

Site	Mean ($\mu Bq/m^3$)	Median ($\mu Bq/m^3$)	Variability*	Comments
Taranaki West	22	13	GSD 3.5	Undisturbed control site
Taranaki North #19	51	52	GSD 2.7	Cleaned site
Taranaki North #18	77	56	GSD 2.5	Uncleaned, recontaminated
Taranaki Northeast (normal)	7	7	SD 3.3	Cleaned site
Taranaki Northeast (storm)	3212	2418	GSD 2.3	Cleaned site, 24-h storm
TM-100	0.35	0.31	GSD 1.62	Undisturbed control site

* Variability is shown by standard deviation, SD, or geometric standard deviation, GSD, of a log normal distribution.

Occasionally, however, a normal distribution or some unknown distribution might apply. In these cases the variation is expressed as a standard deviation, SD.

At the undisturbed control plot on the Taranaki West plume, FW site, the concentration C of Pu, concentration M of *TSP*, and air activity, *Ef* A of Eq 2, were all log normally distributed, and are shown in Figs 2, 3, and 4.


$$y = 11.582 * e^{(1.249 \, norm(x))} \quad R^2 = 0.93902$$
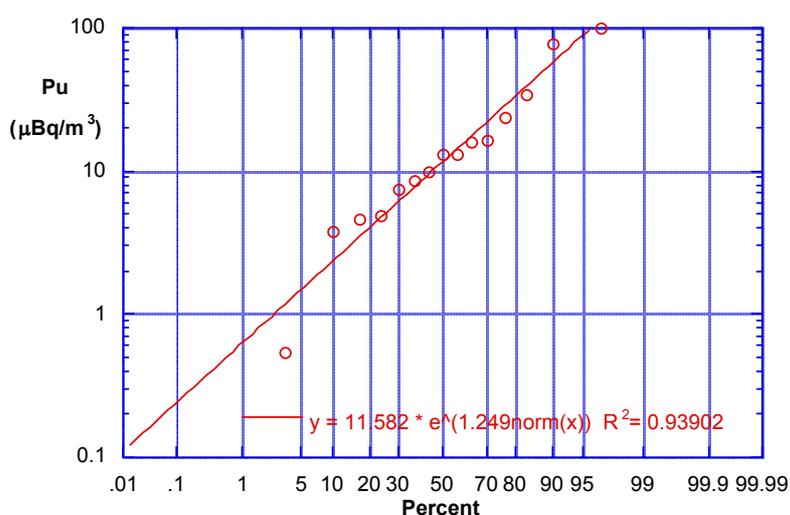
**Fig 2. Pu concentrations in air at FW control site**
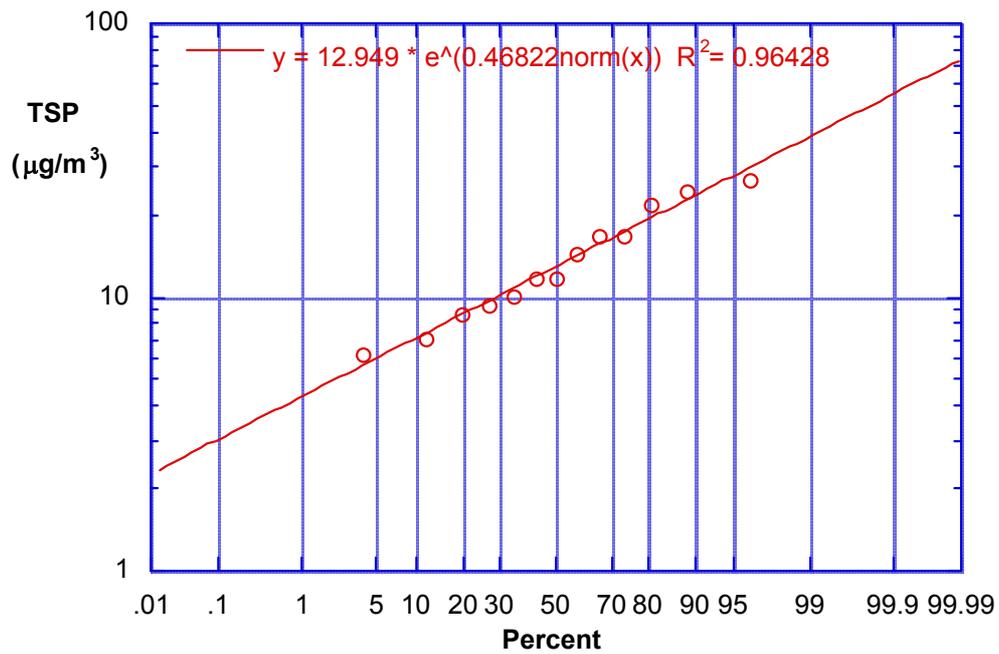
**Fig 3. TSP in air at FW control site**
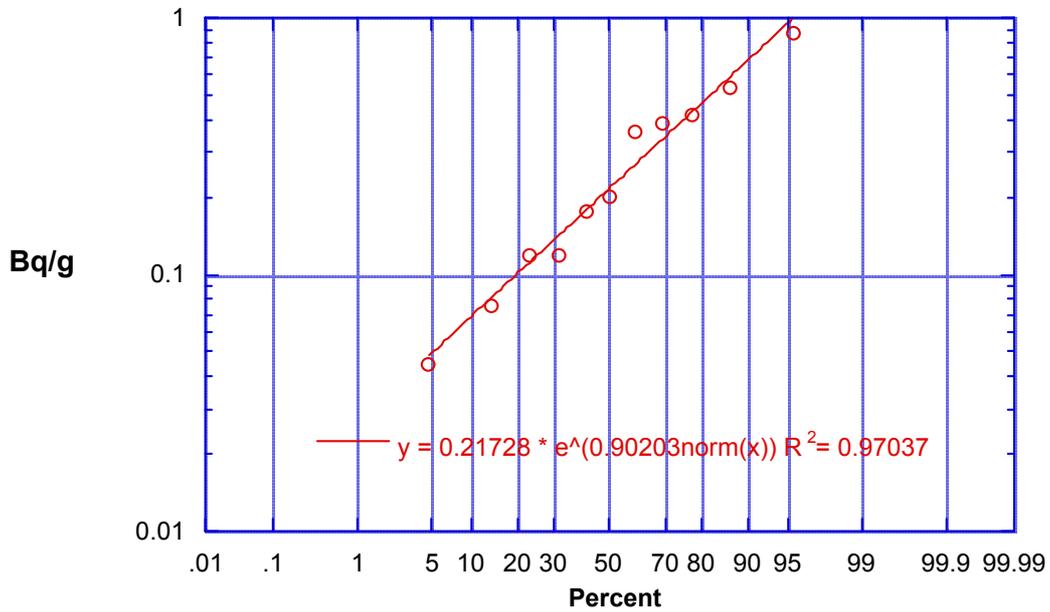
Fig 4. Pu activity in aerosols, FW control site

Log normal distributions were observed even in the case of an unusual dust storm, such as occurred in November 1996 on the Taranaki Northeast plume after it had been disturbed in preparation for soil removal—(see distributions of C, M, and *Ef* A in Figs 5, 6, and 7).
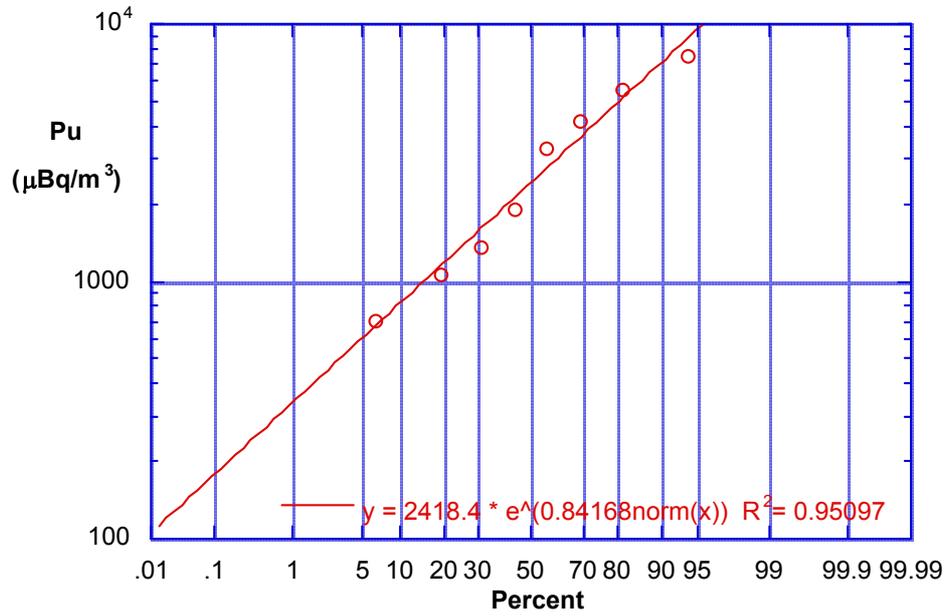
**Fig 5. Pu concentration in air, FNE site, Nov 96 storm.**
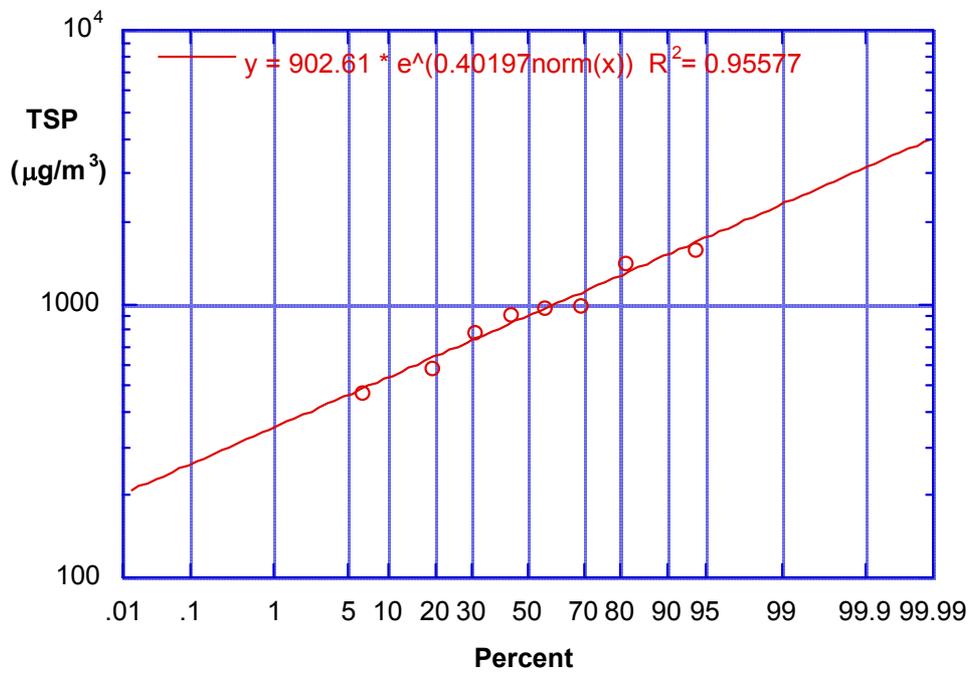
**Fig 6. TSP at FNE site, Nov 96 storm**
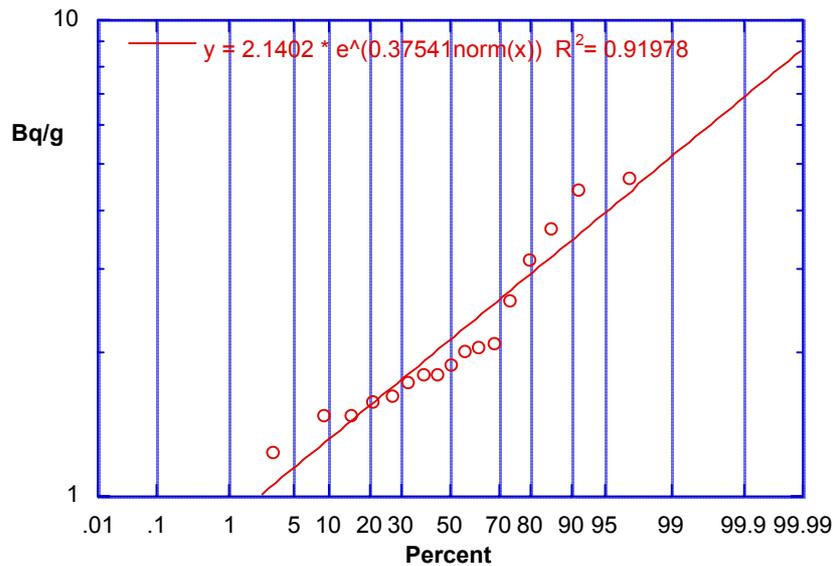
Fig 7. Pu activity in aerosols, FNE, Nov 1996, storm

The dust storm caused the mean values to be elevated, but the characteristics of the log normal distribution remained the same. For detailed risk assessments, the variability would be useful to determine the propagation of variability to the estimate of exposure from combined variables, and the uncertainty could be modeled in a Monte-Carlo sense. For bounding-case dose assessments, as in the case of Maralinga where conservative values are applied the combined variability is not always used.

Another source of variability is in the Pu/Am ratios. (In our observations, we did not calculate Pu concentrations through applications of the Pu/Am ratio, so this variability did not affect the data of Figs 2, 3, 4, 5, 6, and 7.) It is useful to examine the distributions of Pu/Am because it varies with the original, contaminating event. Fig 8 shows that when multiple weapons tests caused the event we are likely to find a log normal Pu/Am distribution. But when only a single weapons test occurred, a normal Pu/Am distribution obtains—(see Fig 9).

In the case of the North plume, Lots 18 and 19, where several weapons tests contributed to the contamination, there were nevertheless log normal distributions of the concentration C of Pu, concentration M of *TSP*, and aerosol activity, *Ef* A.

Figs 10, 11, and 12 show the distribution of measurements of C, M, and *Ef* A in suspended aerosols north of the uncleaned north border of the north plume (Lot 18), where recontamination of undisturbed soil occurred.

13
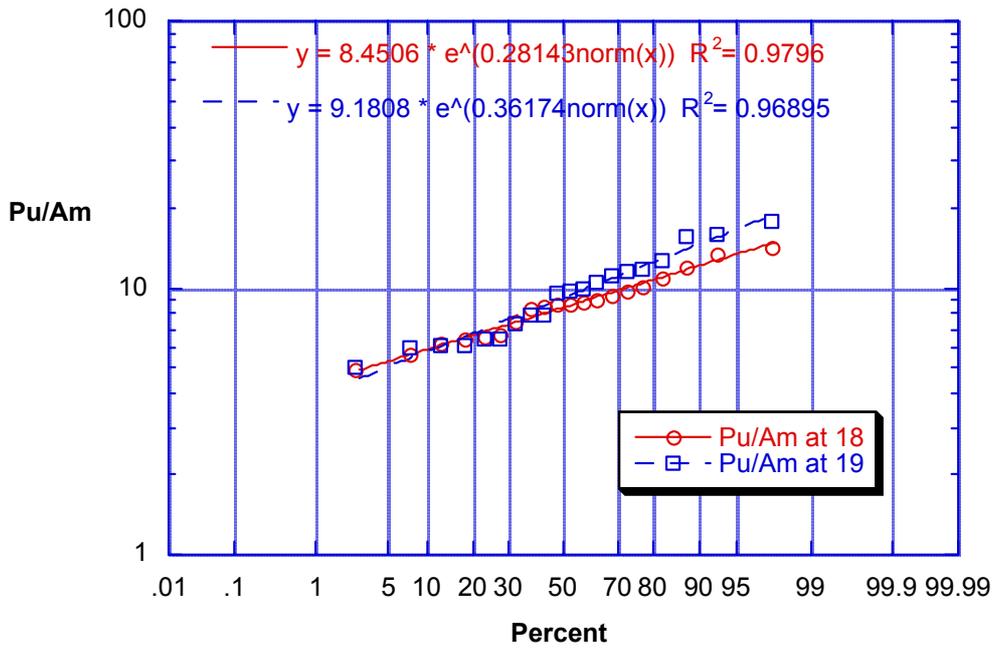
Fig 8. Ratio ^{239,240} Pu / ^{241} Am North plume, Lots 18 & 19
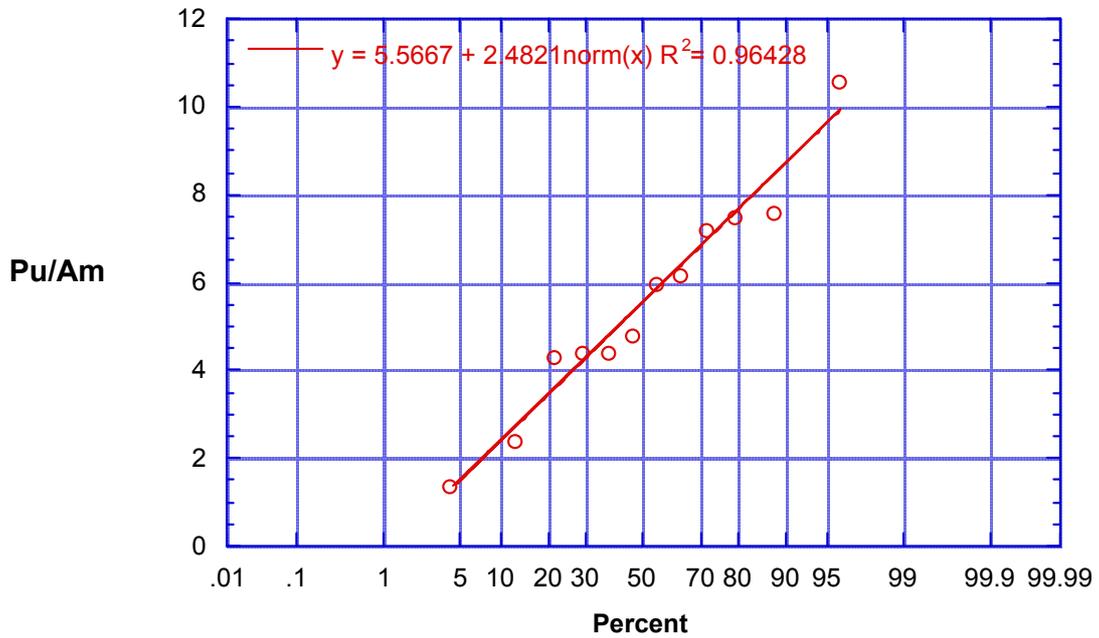


Fig 9. Ratio ^{239,240} Pu / ^{241} Am in Air, FW site, Taranaki

14
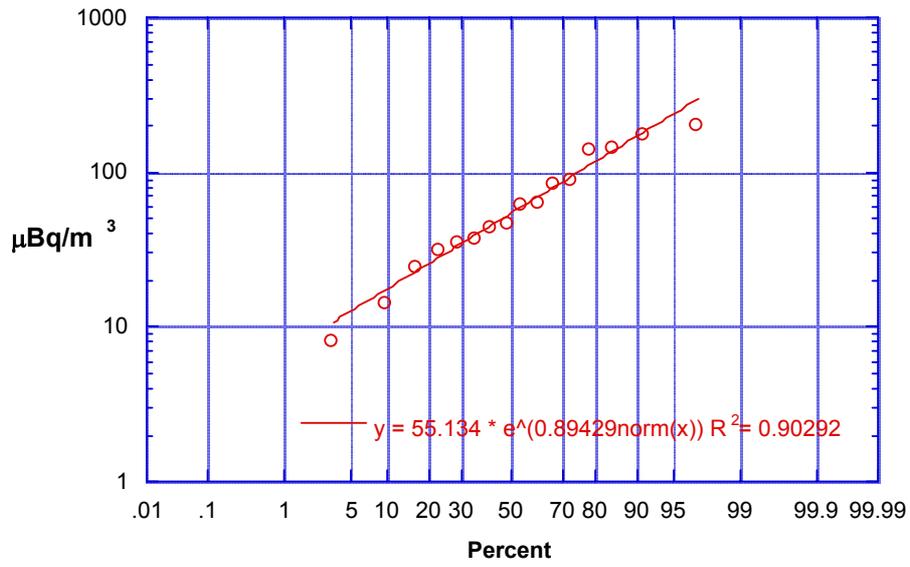
Fig 10. Concentrations of Pu in air, North plume Lot 18, Taranaki
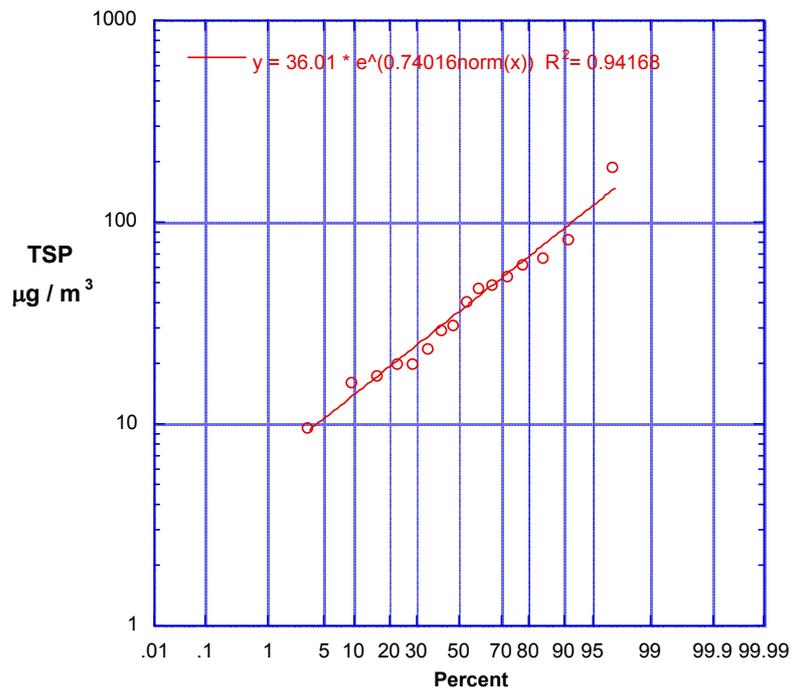
Fig 11. Concentrations of TSP in air, North plume Lot 18, Taranaki

15
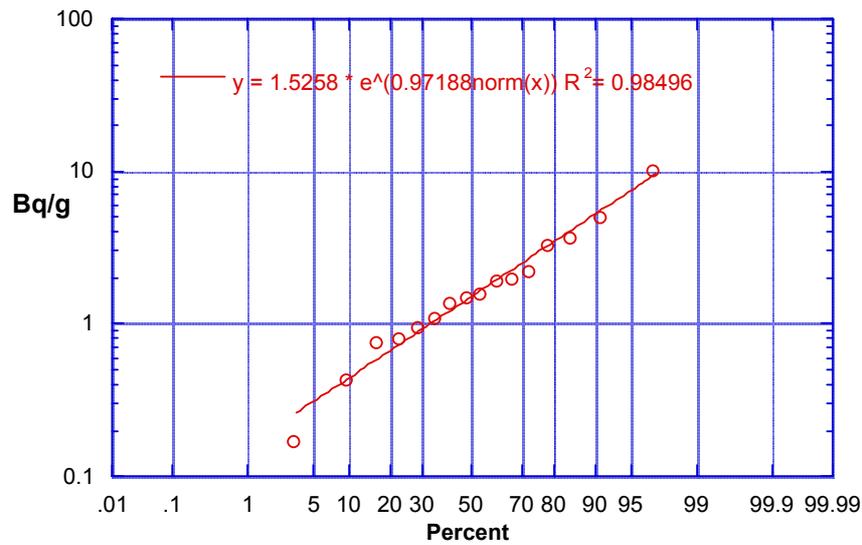
Fig 12. Pu activity, North plume Lot 18, Taranaki

The same kind of log normal behavior occurred in the cleaned Taranaki North plume, Lot 19. Results for C and M at this site are shown in Figs 13 and 14, while a notable exception is the distribution of air activity, Ef A, shown in Fig 15.
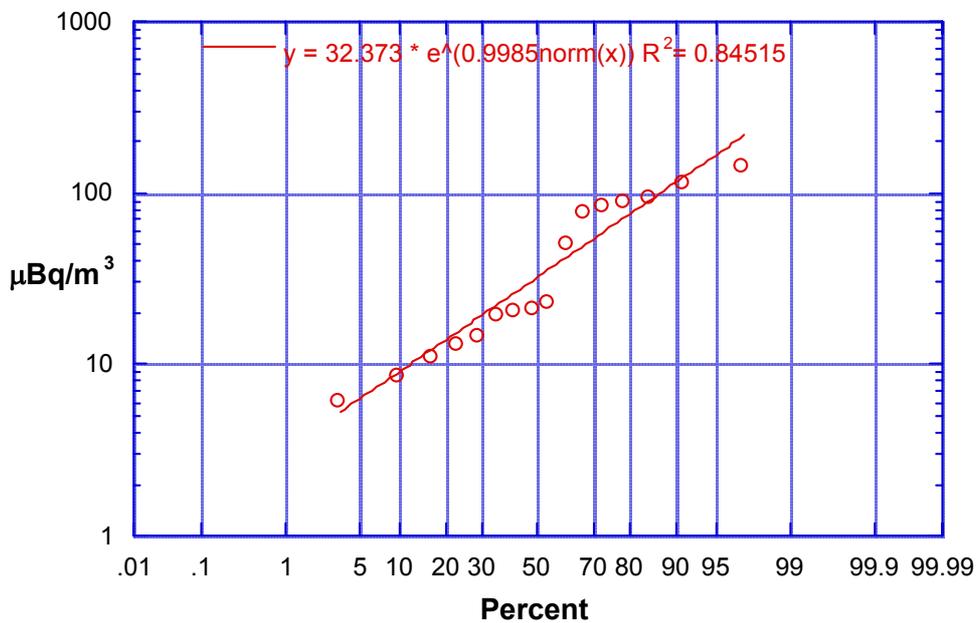

Fig 13. Pu concentration in air, North plume Lot 19, Taranaki

16
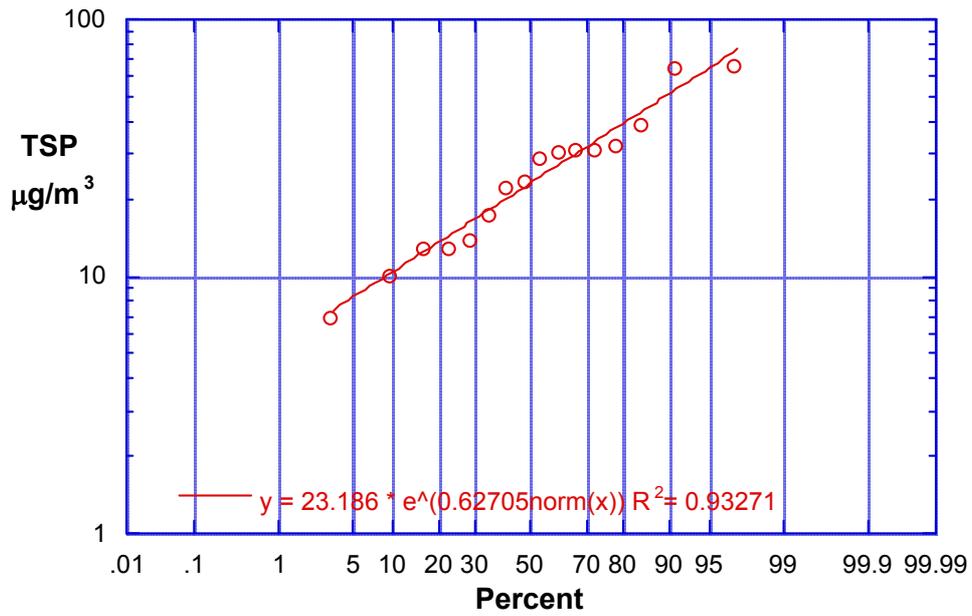
Fig 14. Concentration of TSP, North plume Lot 19, Taranaki

$y = 23.186 * e^{\wedge}(0.62705\,norm(x))\ R^2 = 0.93271$

|	Pu Activity, Bq.g
Minimum	0.36899999
Maximum	6.1700001
Sum	44.567
Points	20
Mean	2.22835
Median	1.022
RMS	2.9925577
Std Deviation	2.0493536
Variance	4.1998502
Std Error	0.4582494
Skewness	0.71279031
Kurtosis	-0.99760504
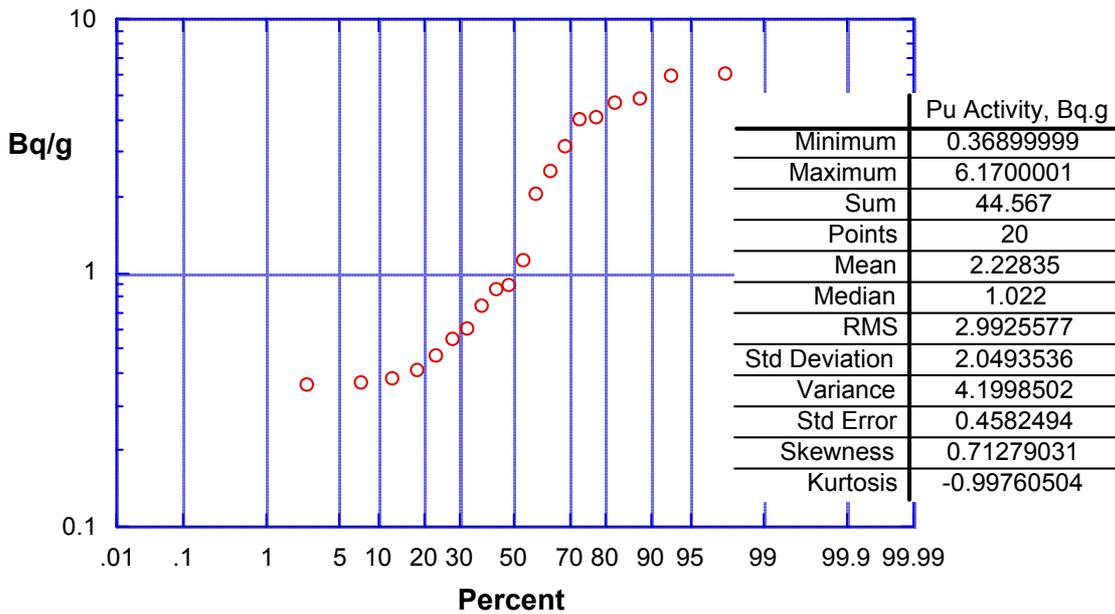
Fig 15. Pu Activity in Aerosols, North plume Lot 19, Taranaki
(an unusual distribution).

A possible explanation of the strange distribution of Pu activity observed in the particulate aerosols in Lot 19 is that after the surface cleaning the locations of deposition patterns from the intermingled weapons tests became heterogeneous. If so, we observed contamination from

different weapons tests on separate samples and hence we have observed a confounded, composite distribution.

The data collected at TM-100 in October 1997 is somewhat at odds with what we expected to find. We observed much lower Pu/Am ratios than expected (Table 2) and much lower resuspension factors S_f than found at any of the Taranaki sites (Table 3). This difference was not due to sampling errors, as low values showed up in many observations of both C, Pu concentrations in air (Fig 16, where the deposition was about 4 kBq/m^2) and the Pu/Am ratios (Fig 17) for TM-100, Lot 12.
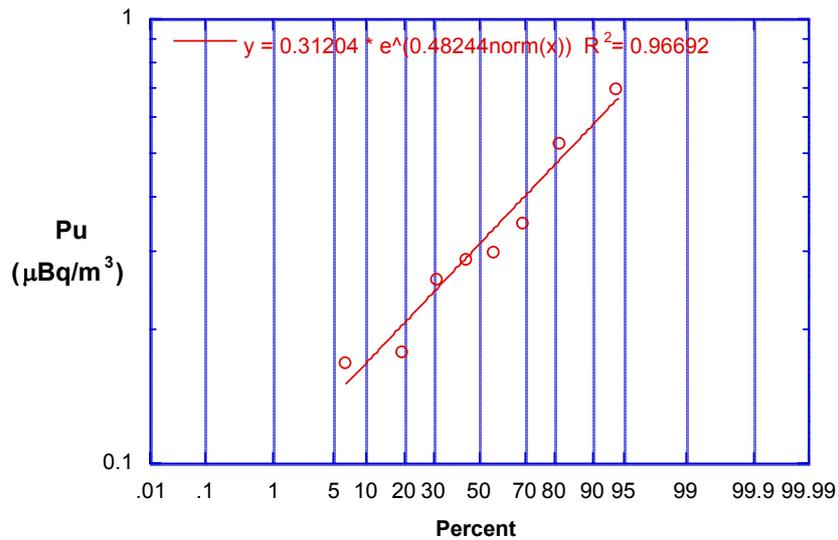

**Fig 16. Pu concentration in air, TM-100 Lot 12, undisturbed control.**
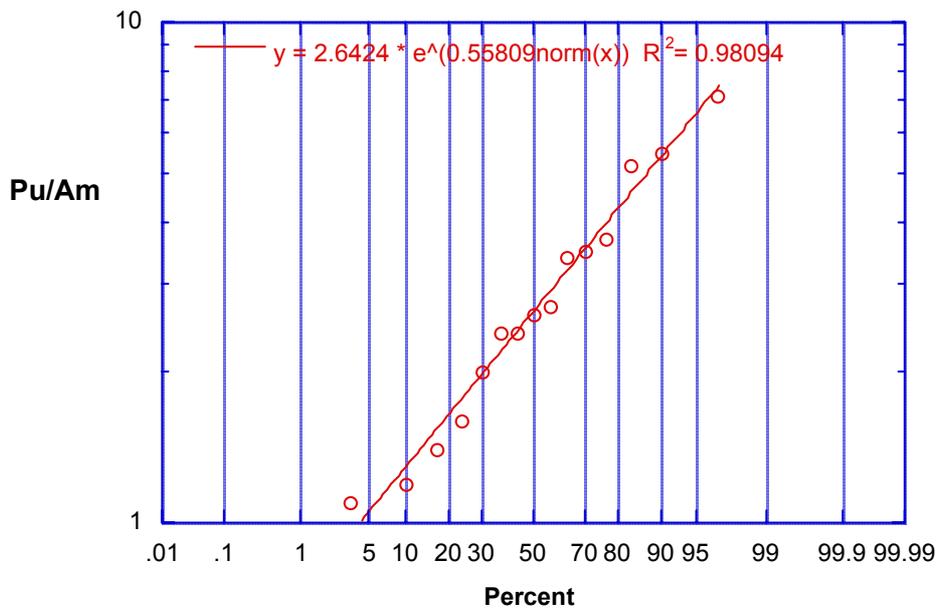
Fig 17. Ratio ^{239,240}Pu / ^{241}Am, TM-100 Lot 12.

4. Loss of Pu through continued resuspension due to wind erosion.

Since the variability has been well documented, one remaining question was the predictability of the surface stability. The estimation of resuspension rates S_f depended upon the vertical gradient of C and M measured using the vertical array system (VAS) described under Methods Part 2. The Eqn 6 vertical gradient parameter, p, observations are summarized in Table 5. The values are similar to those observed elsewhere.

TABLE 5. VALUES OF NON-DIMENSIONAL VERTICAL GRADIENT, p

Site	Observed p (TSP)	Observed p(Pu)	Comments*
Taranaki West	-0.23	-0.54	Undisturbed control site
Taranaki North, #19	-0.11	-0.28	Cleaned site
Taranaki North, #18	-0.18	-0.39	Uncleaned, recontaminated
Taranaki Northeast	-0.28	-0.40	Cleaned site (during storm)
TM-100	-0.20	-0.58	Undisturbed control site

*Values are negative indicating a decrease in concentration with height, and a loss from the surface. The average ratio of p(Pu)/p(*TSP*) = 2.3 with standard deviation 0.6

The mean vertical profiles of C and M are shown in Figs 18 and 19 with the slopes fitted by least squares regression given in the first row of Table 5.
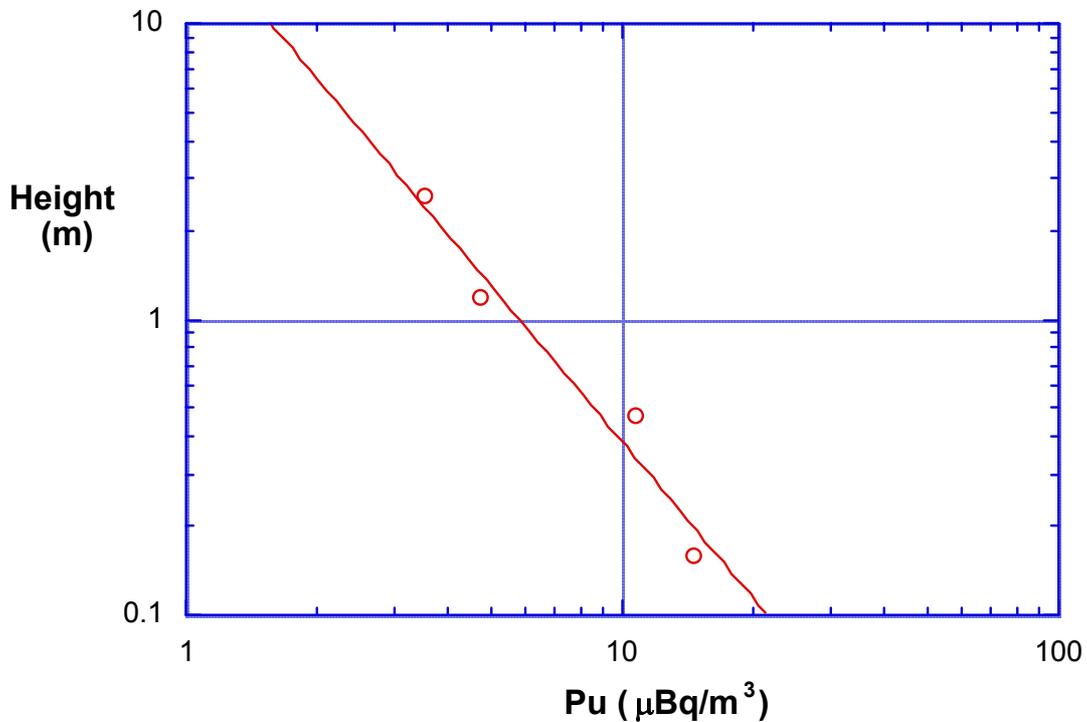
Fig 18. Vertical gradient of Pu observed at Taranki West, FW site.

The vertical profiles of C and M can be seen to be slightly different. This same characteristic is shown during the dust storm of November, 1996, at Taranaki Northeast, in Figs 20 and 21. The ratio of the slopes is on average about 2.3 (see Table 5).
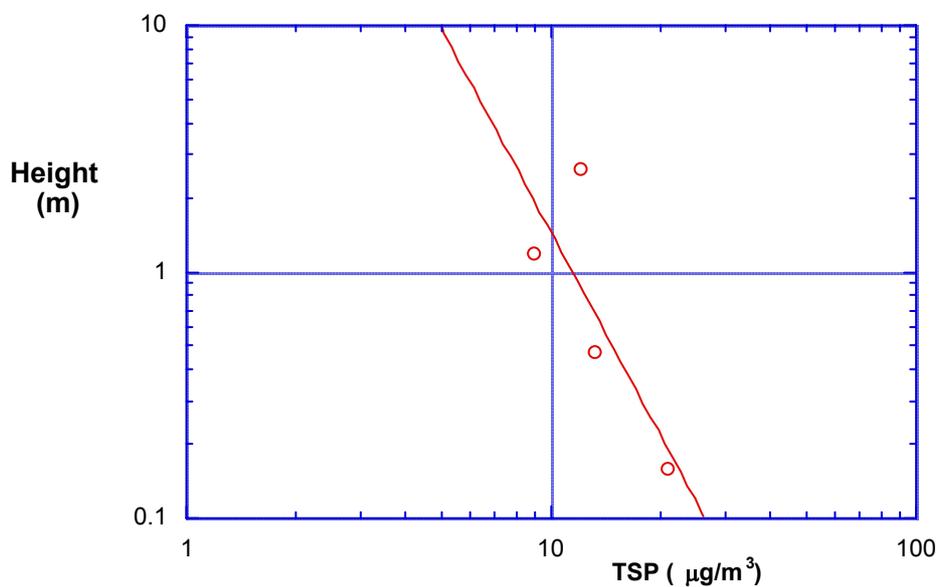
Fig 19. TSP profile at Taranaki West, FW site.
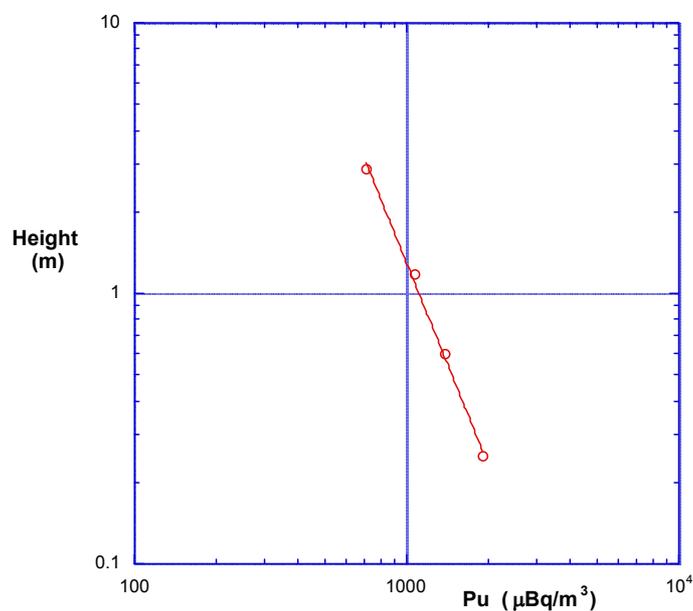
Fig 20. Vertical gradient of Pu at Taranaki Northeast during storm.
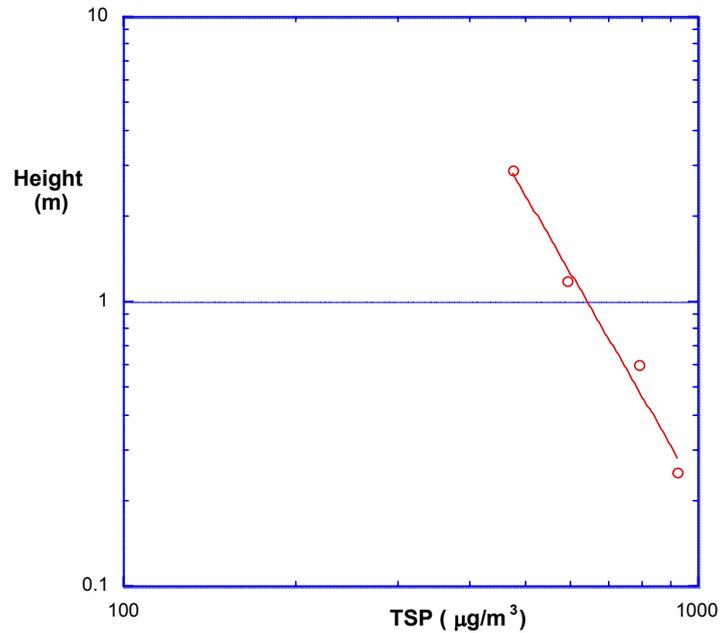
Fig 21. Vertical gradient of TSP at Taranaki Northeast during storm.

We have observed at Tonopah, Nevada, that the profiles are often shifted toward higher concentrations without a change in slope. Hence, p-values can be similar in stormy and non-stormy conditions. This is shown in Table 5 where p-values in row 4 are not much different than p-values in row 1. Likewise TM-100 with very low resuspension rates has similar p-values as the Taranaki sites; (see Fig 22, which is plotted with a graphical rotation to show both C and M).
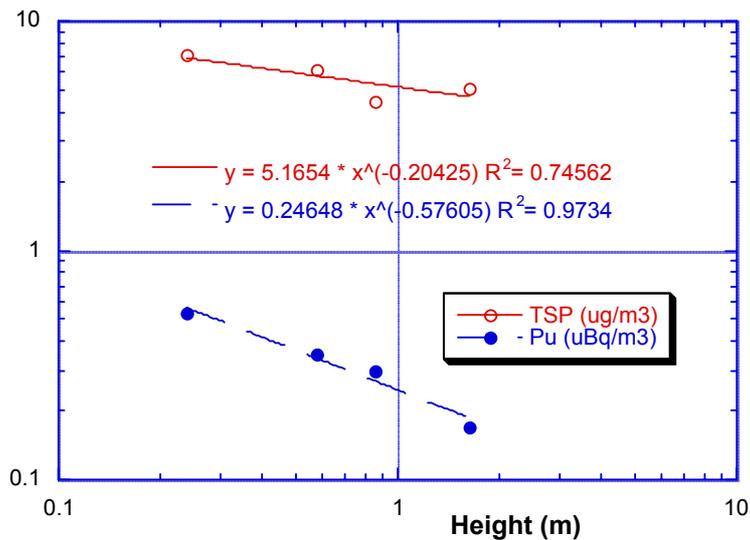
Fig 22. Vertical profiles of Pu and TSP at TM-100 Lot 12

Wind, p-values, and resuspension factors enter into the determination of the stability of Maralinga sites. The key parameters as defined by Eqns 3–7 and then observed at Maralinga are shown in Table 6. We calculated a resuspension rate for Taranaki control site, a cleaned site, and the TM-100 uncleaned site. The calculations show that if the resuspension rate remained constant, it would take a few thousand years to remove it by wind erosion in the worst case, which is the cleaned plot. On the other hand the resuspension rate at Taranaki North would be expected to eventually decrease to probably something like the Taranaki West undisturbed site. The revegetation rate will assist this if rehabilitation is successful.

TABLE 6. CALCULATED RESUSPENSION RATES

Site	Wind speed at 2 m (m/s)	Diffusivity K (m^2/s)	Vertical Gradient p(Pu)	Resuspension factor S_f (m^{-1})	Resuspension rate (sec^{-1})*	Time to lose one-half (yrs)**
Taranaki West, undisturbed	4.3	0.205	-.54	3×10^{-10}	2.77×10^{-11}	7900
Taranaki North, #19 cleaned lot	3.5	0.106	-.28	4×10^{-9}	1.0×10^{-10}	2200
TM-100, uncleaned	3.6	0.168	-.58	3×10^{-11}	2.3×10^{-12}	95500

* Resuspension rate = $K(p/z) S_f$ where height of Pu measurement z =1.2 m
**For the sake of visualization, the resuspension rate is converted to time for resuspension to remove one-half of the Pu in the soil providing that the rate stays constant over time.

The diffusivity, K, would be expected to decrease on a clean, smooth plot due to the removal of surface roughness, z_o. This was observed at Maralinga—(see Fig 23). The effect may be temporary, however, because restoration with vegetation and water erosion will combine to make the cleaned surface rough over time.
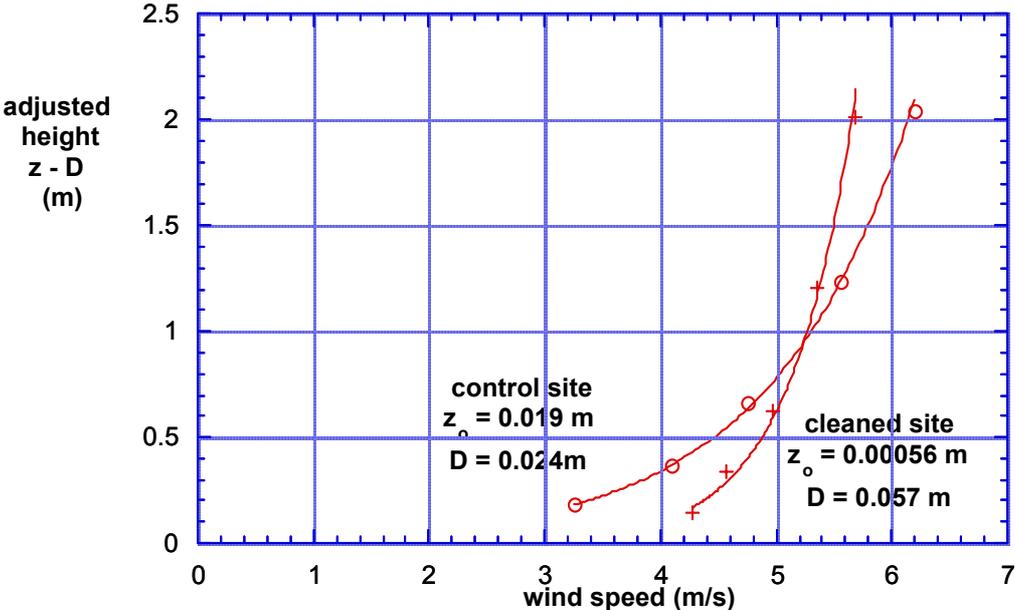

Fig 23. Wind speed profiles on cleaned and uncleaned plots.

EFFECTIVENESS OF REHABILITATION

After the soil surfaces had been cleaned and stabilized and following a few years of natural weathering, the rehabilitation effect was noticeable in the reduced Pu concentrations in air. In April-May 2000, the air flowing over the former deposition plumes at Taranaki picked up Pu concentrations near 0.1 μBq/m^3, and at TM-100 and Wewak, the Pu concentrations in air were on the order of 0.01 μBq/m^3. These compare to a value of 0.006 μBq/m^3 at Maralinga Village during the same period. See Table 7. (There are very few locations in the world monitoring airborne Pu, but we obtained a value of 0.0004 μBq/m^3 for a station in Melbourne, and a value of 0.002 μBq/m^3 for a representative northern hemisphere site in California.)

Rehabilitation has reduced Pu concentrations in air, C, by more than a factor of 100 at Taranaki by comparison to values in Table 4. Measurements by ARPANSA in the vicinity of the air samplers in April-May 2000 showed deposition **of** [241]Am, D, approximately 0.3 kBq/m^2 and with the aid of a Pu/Am ratio of 10 from Table 2, and Eq 1, we calculated an approximate resuspension factor S_f of 3 x10^{-11} m^{-1}. This shows that the resuspension factor has returned to the undisturbed state below that shown in Table 6 for Taranaki during cleanup.

TABLE 7. PU CONCENTRATIONS IN AIR AFTER MARALINGA REHABILITATION

Site	Sampled air Volume (m^3)	Observed Pu (μBq/m^3)	Dates In 2000
Taranaki FNE, Northeast	102000	0.050	April 27-29
Taranaki FN, North	149000	0.122	May 2-5
Taranaki FNW, Northwest	123000	0.126	April 30-May 2
TM-100, Northeast	110000	0.012	May 5-7
Wewak	101000	0.011	May 7-9
CONTROLS			
Maralinga Village	112000	0.006	April 27-May 8
California Rural Site*	---	0.002	Annual Median
ARPANSA, Melbourne**	316000	0.0004	March 31-May 15

*Annual Site Environmental Report, Lawrence Livermore National Laboratory.
**Uncertainty of Maralinga samples is less than 2% and uncertainty of Melbourne samples is 60% by the method of accelerator mass spectroscopy.

CONCLUSIONS

There are indications that in the long run the concentrations of Pu in the air will become equivalent to world-wide background; values near 0.1 μBq/m^3 were already evident at Taranaki in 2000. The resuspension factors in 1996-1997 and 2000 were low and within an order of magnitude, or below, the typical values found worldwide, 10^{-10} m^{-1}. Surface sealing by rain, and revegetation has continued to stabilize the soil.

We have observed the important variables and the uncertainty: size of the suspended aerosols, Pu concentrations in air, *TSP* concentrations in air, Pu activity on aerosols, resuspension factors, and wind effects. We have calculated potential resuspension rates. There apparently is nothing out of the ordinary with respect to these values in contributing to potential human exposure. Our measurements for the most part agree with those made by ARPANSA, and show that the dose assessment considerations were very conservative and protective of the public from a radiological perspective. Assumptions made about enhancements and dusty environments for dose assessment purposes are likely to be bounding case assumptions and even more extra protective of the Aboriginal people.

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APPENDIX 1.

Maralinga Map and Cleanup Lot Maps. Taken from Maralinga Rehabilitation Technical Advisory Committee, *Rehabilitation of Former Nuclear Test Sites at Emu and Maralinga (Australia) 2003*, Commonwealth of Australia, Department of Education, Science and Training, 403pp.
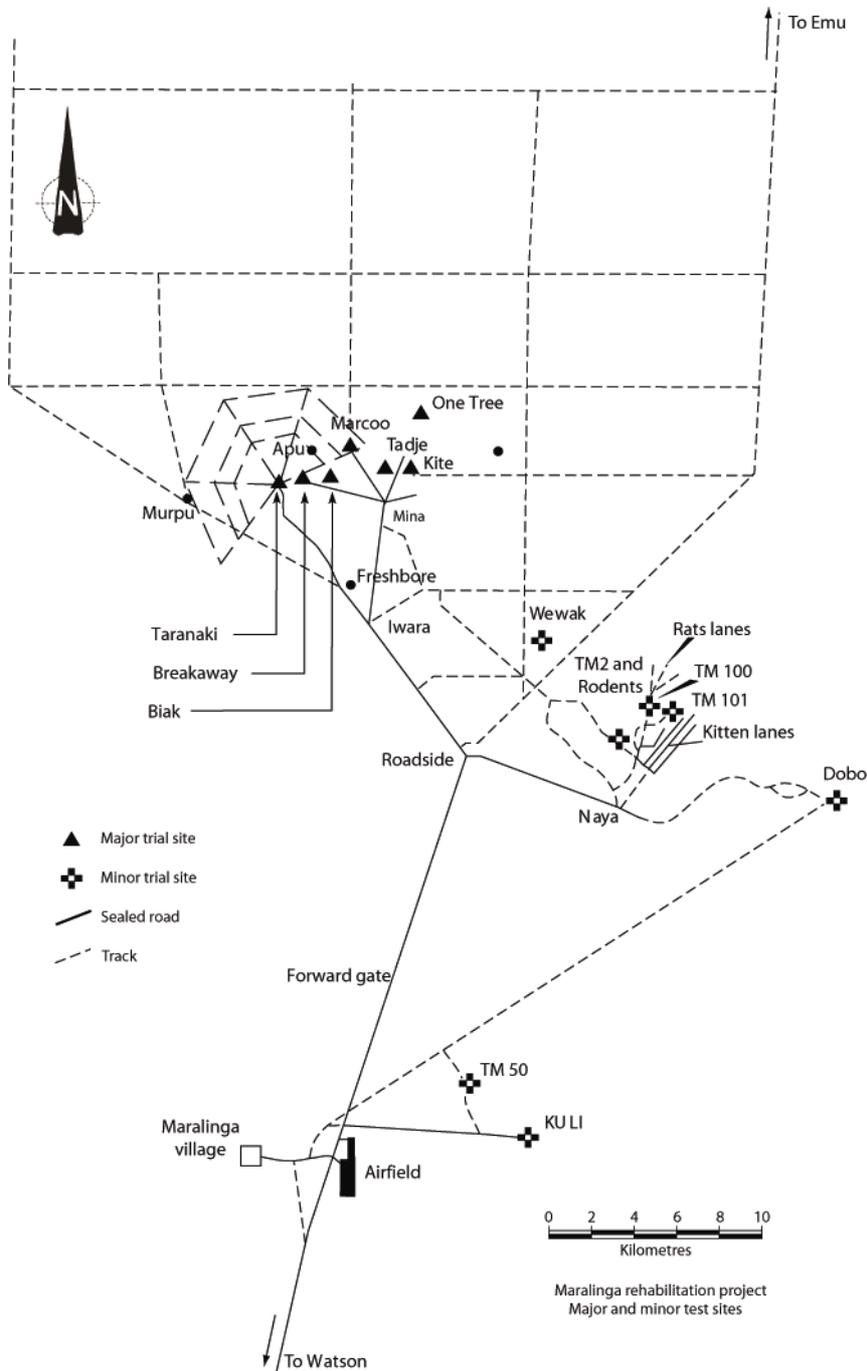

Map showing the locations of Taranaki, TM-100/101, Wewak and Maralinga Village.
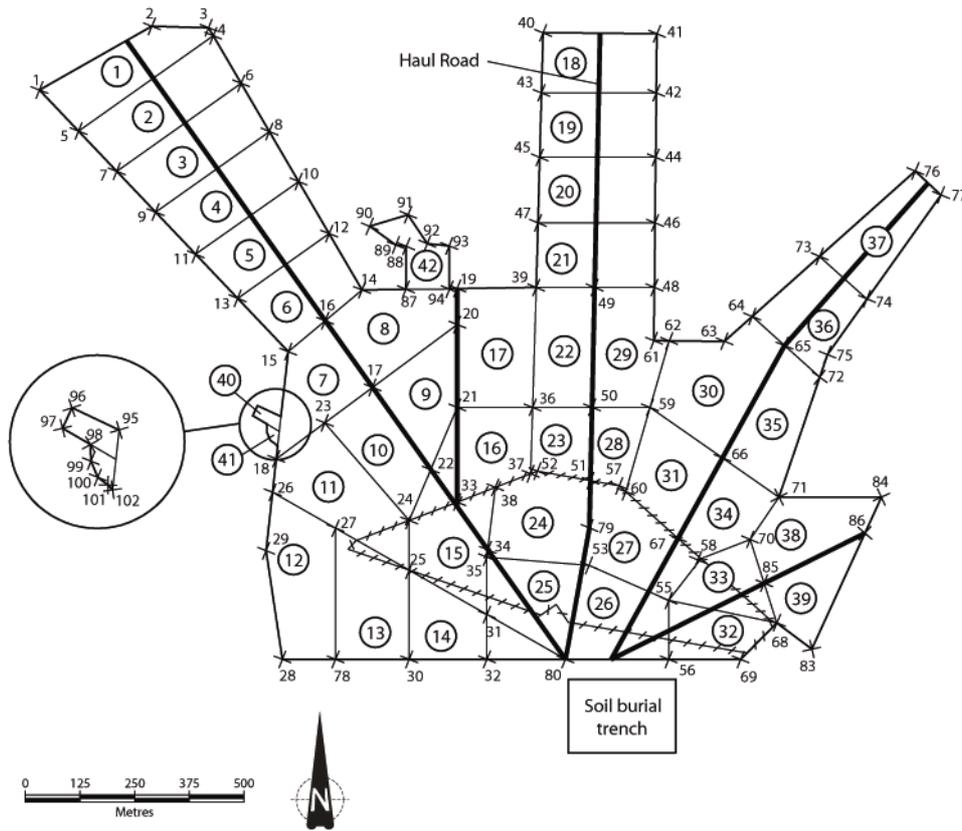
Taranaki cleanup lots.
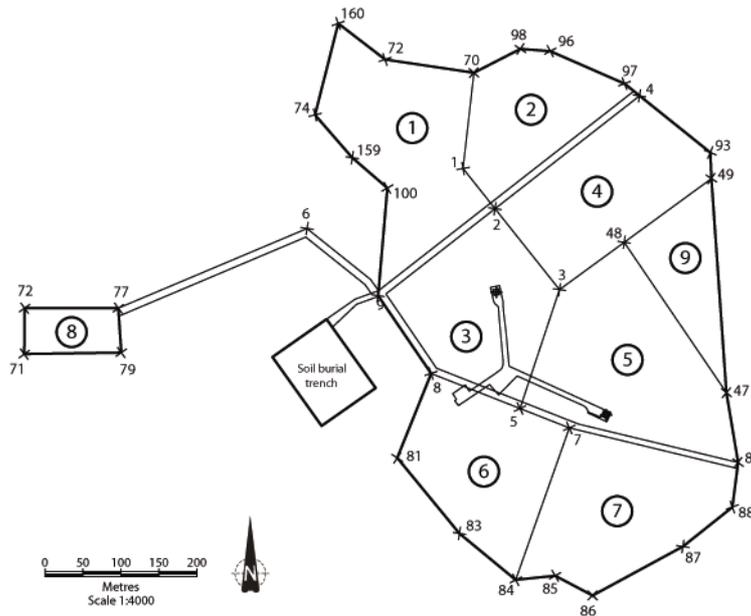
Wewak cleanup lots.
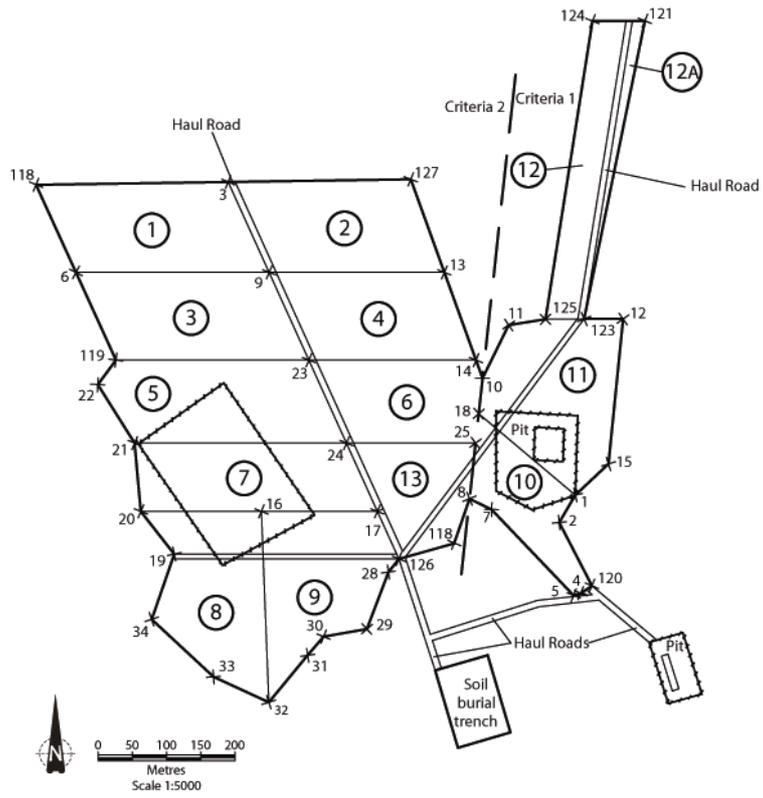
TM-100/101 cleanup lots.

APPENDIX 2.

Investigations by Bertoldo, 1999, using data collected with the isokinetic particle counting system included an attempt to fit the complex, observed size distributions by the sum of three, log normal distributions. The width of each distribution by GSD, the % area enclosed by the curve, and the mass median aerodynamic diameters (MMAD) of the distributions were calculated. These can be compared by wind speed during the same period in Table A. The data were collected during March 1997 in the Taranaki North plume, Lot 18 (undisturbed, recontaminated plot) and Lot 19 (cleaned plot).

It is difficult to draw major conclusions about the results except that it is clear that from time to time small contributions are made by minor modes of larger size particles, and these contributions tend to increase when the winds increase beyond 4 m/s. It is sufficient to recognize that the particle size distribution is dynamic, and it is probably the average size distribution that is more important for purposes of human dose assessment. The cascade impactor thus serves a useful purpose in collecting median aerodynamic diameter information of both the Pu aerosols and suspended mass averaged over time.

TABLE A. FITTED LOG NORMAL CURVES TO OBSERVED PARTICLE SIZE DISTRIBUTIONS, AFTER BERTOLDO, 1999

Wind Speed (m/s)	SITE & TRIAL	#1 GSD	#1 % area	#1 MMAD	#2 GSD	#2 % area	#2 MMAD	#3 GSD	#3 % area	#3 MMAD
1.73	18/4	1.57	63	5.9	1.13	24	3.8	1.13	13	6.0
2.78	18/5	1.62	75	7.0	1.10	12	3.8	1.13	13	6.0
3.52	18/6	1.55	77	6.2	1.10	11	3.8	1.14	12	5.9
3.52	18/7	1.60	63	6.6	1.14	17	3.9	1.16	20	6.0
4.39	18/8	1.60	47	7.1	1.15	40	4.9	1.10	13	6.8
4.35	18/9	1.50	63	6.9	1.10	22	4.7	1.13	15	6.5
2.79	19/1	1.55	69	6.6	1.12	18	4.2	1.14	13	6.0
3.15	19/2	1.57	74	6.7	1.10	18	4.1	1.12	8	5.9
4.95	19/3	1.57	65	6.8	1.13	20	4.0	1.14	15	6.0
6.27	19/4	1.62	40	7.2	1.23	33	4.8	1.15	27	6.2
5.77	19/5	1.46	40	5.9	1.30	57	6.5	1.02	3	8.9
7.36	19/6	1.56	36	7.9	1.47	59	8.9	1.02	5	8.9
6.82	19/7	1.55	88	10.3	1.03	3	7.1	1.03	9	8.8
3.88	19/8	1.52	55	6.4	1.15	33	4.6	1.11	12	6.5
3.32	19/9	1.52	60	6.4	1.13	27	4.5	1.13	13	6.8