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# TECHNICAL REPORT

# Radioactivity Enhancement Factors of Maralinga Soils

**Stephen Long and Liesel Green** 

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by

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# **Executive Summary**

Under Memorandum of Understanding Number 001872 between ARPANSA and the Department of Resources Energy and Tourism (signed in 2011), ARPANSA was tasked to re-assess the potential doses arising from possible exposure to the residual radioactive contamination at Maralinga. In assessing the inhalation pathway, some knowledge of the size distribution and activity concentration of radioactive contamination in the soil is required.

This study is a re-assessment of the radioactivity enhancement factors for Maralinga soil. The enhancement factor is defined as the ratio of the activity concentration of the inhalable size fraction of dust arising from contaminated soil to that of the bulk surface soil. Field and laboratory measurements of seven soil samples are described and the results are compared to the data used in previous dose assessments.

This study also includes a re-assessment of the activity ratio of americium-241 to that of plutonium-239, knowledge of which is also required in assessing the doses.

Overall, this work shows no indication that the values for the enhancement factor or the activity ratio used for the previous dose assessments are questionable or have changed over the past two decades. Therefore, the current dose assessment may continue to use an enhancement factor of 6.2 and a Pu-239:Am-241 activity ratio of 7.2.

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## 1. Introduction

Between 1953 and 1963, the United Kingdom conducted a programme of nuclear weapons development trials at Maralinga in South Australia. Together with nine major trials involving nuclear explosions, several hundred smaller scale experiments were conducted which dispersed radioactive materials over large areas. Much of this radioactive contamination was removed during the Maralinga Rehabilitation Project which was completed in 2001. Nonetheless, large areas remain which are contaminated with low levels of radioactive material.

The inhalation pathway presents the most significant potential health hazard arising from this residual contamination. Previous studies (Williams 1990; Williams et al. 2002) have assessed the potential dose arising from inhaling contaminated dust at Maralinga. This study is intended to provide input to a new assessment of the potential inhalation dose due to the residual contamination at Maralinga.

By far the most significant radionuclide in the inhalation dose assessment is plutonium-239. Unfortunately, this radionuclide cannot be directly measured in the large area over which it is dispersed. However, plutonium-241 was a 'contaminant' in the plutonium used for the minor trials. This radionuclide decays with a half-life of approximately 14 years to americium-241. Americium-241 can be easily measured in the field over large areas and is used as a surrogate for the measurement of Pu-239. In order to estimate the activity concentrations of Pu-239 at Taranaki, an estimate of the activity ratio between Pu-239 and Am-241 must be evaluated.

The previous studies (Williams 1990; Williams et al. 2002) have shown that the activity distribution with particle size is quite different from that of mass distribution. That is, the activity concentration of dust containing only small diameter particles is considerably greater than that of the bulk surface soil. These previous studies have estimated that the americium-241 activity concentration in the respirable (< 7  $\mu$ m diameter) fraction of the soil is a factor of 7 greater than that of the bulk soil.

## 2. Measurements

#### 2.1 Sampling Locations

Between September 29 and October 14 2010, four ARPANSA staff members collected soil samples from six sites in the North plume and one site in the Northeast plume at the Taranaki test site at Maralinga (see Table 1 and Figure 1).

Site	Coordinate	s (AMG84)	Distance from	Am-241 Activity Concentration (kBq/m²)	
Number	Easting	Northing	Taranaki SRB (km)		
1	750151	6692337	1	55	
2	750170	6692855	1.5	~ 35	
3	750135	6693358	2	26	
4	750121	6694548	3	20	
5	750089	6695355	4	7	
7	750133	6698934	7	7	
9	751685	6691700	1	5	

 Table 1: Details of the soil sampling sites at Taranaki.

Notes: The fourth column indicates the distance from the Soil Removal Boundary.

The last column indicates the bulk activity concentration measured by in situ gamma-ray spectrometry.

Sites 6 and 8 were not sampled due to time constraints.

Figure 1 also shows the approximate location of three sites (historic sites) from which samples were obtained for the previous studies (Williams 1990). It should be noted that the samples were gathered from these historic sites before the commencement of the Maralinga Rehabilitation Project (MARTAC 2002) in 1996 and, therefore, before soil was removed from the site.





#### 2.2 Sampling Methodology

A custom made sampler consisting of a spacer, guide and pan shovel was used to sample the top 1 centimetre of soil (see Figure 2). It is assumed that the top centimetre contains approximately 80% of the activity of the soil coil column and that it is only soil to this depth that contributes to respirable dust. At each site, repeated scrapings were taken until approximately 25 kilograms of soil had been sampled in an approximately 2 metre transect. Care was taken not to disturb the soil area before it was sampled and any large plant material was avoided by moving the transect left or right of the original line. Smaller plant material and debris was removed either by hand at the time of collection or when the initial sample was sieved to less than 1000  $\mu$ m.

Figure 2: The equipment used to collect the soil samples.



The entire 25 kilogram sample was further sieved on site to yield 1000-500  $\mu$ m, 500-250  $\mu$ m, and 250-180  $\mu$ m sized fractions using standard test sieves 200 mm in diameter. Approximately 500 gram sub-samples were manually sieved batch-wise with the sample from the receiving pan retained for further size fractionation. Masses of the total sample and of the sieved fractions were recorded for each sub-sample and later combined in order to allow the calculation of an overall mass contribution for each fraction.

100 gram sub-samples of all sized fractions were taken for later gamma-spectrometry measurement in the laboratory. A further 2-3 kilogram sample of the less than 180  $\mu m$  fraction was retained for further particle sizing in the laboratory.

In the laboratory the less than 180  $\mu$ m fraction was further sized using an Endicott Test Sieve Shaker model EFL2 MK3 into 180-150  $\mu$ m, 150-90  $\mu$ m, 90-75  $\mu$ m, 75-45  $\mu$ m and <45  $\mu$ m fractions. Approximately 150 gram sub-samples were sieved with test sieves stacked and shaken for a set time interval. The mass of each sieved fraction and the mass of each sub-sample was recorded and the calculated results were again combined in calculating the mass contribution for each fraction. At least 10 replicate fractionations were made before the sized fractions were sub-sampled for gamma-spectrometry analysis.

A Gilsonic Autosiever GA-6 was used to further separate the less than 45  $\mu$ m fraction into 45-35  $\mu$ m, 35-20  $\mu$ m and less than 20  $\mu$ m fractions. Again the masses of each sieved fraction were recorded along with the mass of the sub-sample (approximately 5 grams was used) in order to calculate the mass contribution of each fraction. Attempts to separate the < 20  $\mu$ m fraction into smaller size fractions failed because the clay content of the soil precluded consistent fractionation, as the particles tended to agglomerate.

#### 2.3 Gamma-Spectrometry

A sub-sample of approximately 16 g (12–20 g, depending on sample density) from each fraction was weighed into a standard, plastic, 50 mm diameter by 5 mm deep container. These sub-samples were measured using High-Purity Germanium gamma-spectrometers, with a relative efficiency of approximately 35%, to ascertain the activity concentration of Am-241. The detectors were calibrated against a traceable standard containing Am-241 in the same-shaped container. A correction for self-absorption in the soil was calculated with LabSOCS (Bronson 2003). The activity concentration of each sub-sample was taken to be representative of the entire fraction.

#### 2.4 Radiochemical Analysis

Of the  $< 20 \,\mu\text{m}$  fraction from each site, 0.1 g was taken to determine the activity ratio between plutonium and americium. Each sub-sample was digested overnight in a mixture of concentrated nitric and hydrochloric acids to leach the radioactive elements from the soil. The resultant solution was filtered prior to adding known amounts of plutonium-242 and americium-243, as radiochemical tracers, and precipitating the radionuclides by addition of ferric chloride and ammonia. After decanting the supernatant, the precipitate was re-dissolved in 9 M nitric acid prior to adding 1 g of sodium nitrate. The solution was then heated until all brown nitrous oxide fumes disappear.

The solution was then added to an ion-exchange column containing Bio-Rad<sup>™</sup> Anion Exchange Resin (AG-1X8) washed with 8 M nitric acid to convert it to the nitrate form. The plutonium was retained on the resin, while the americium passes through. Any thorium in

the sample was eluted with a solution of 3 parts concentrated hydrochloric acid with 1 part de-ionised water and discarded. The plutonium was eluted with a mixture of ammonium iodide and hydrochloric acid.

The effluent containing americium was evaporated to near dryness and re-dissolved in a mixture 0.5 M aluminium nitrate in 2 M nitric acid. The resultant solution was then added to an ion-exchange column containing Eichrom  $TRU^{TM}$  resin washed in 2 M nitric acid. The column was then rinsed in 2 M, then 1 M nitric acid prior to eluting the americium with 0.025 M nitric acid.

The radionuclides in each of the solutions containing americium or plutonium were then electrodeposited onto stainless steel discs. The alpha-emission rates of the radionuclides were measured using Passivated Ion-implanted Planar Silicon detectors. The detectors have with an active area of 450 mm<sup>2</sup> and a detection efficiency of approximately 35% and are operated under partial vacuum to minimise ion recoil. The measured emission rate was converted to activity by comparison with the measured emission rate and known activity of the americium-243 or plutonium-242 tracer.

# 3. Results

### 3.1 Mass Distribution

The total mass in each fraction from each sampling site is detailed in Table 2, below.

Fraction Size	Mass of Fraction (g)								
(µm)	Site 1	Site 2	Site 3	Site 4	Site 5	Site 7	Site 9		
1000-500	6564	7324	5431	4401	1668	6818	6983		
500-250	10998	8102	983	10782	12817	16644	11863		
250-180	3113	2959	4082	4004	4979	1926	3168		
180-150	660	196	619	485	692	881	752		
150-125	987	445	1186	960	1478	1285	1319		
125-90	1553	874	1839	1483	2125	1589	1826		
90-75	835	541	1034	805	944	583	761		
75-45	1361	1012	1780	1381	1238	772	1075		
45-35	183	128	216	139	108	68	85		
35-20	163	121	254	158	125	61	85		
<20	60	76	152	57	103	44	60		

 Table 2: Mass of each fraction from each sampling site.
 Image: Comparison of the sampling site is a standard s

Note that sites 6 and 8 were not sampled due to lack of time. The relative uncertainty (k=1) for all values is less than 0.1%.

The fractional contribution by mass is shown in Figure 3. This figure indicates that the samples from Site 3 have a different mass distribution from that of the other locations.

*Figure 3:* The fractional contribution by mass of the size fractions from each sampling site.



The mass fraction passing through each of the sieve mesh sizes, for samples from each sampling site, are detailed in Table 3.

Mesh Size	Mass Fraction Passing Through Mesh									
(μm)	Site 1	Site 2	Site 3	Site 4	Site 5	Site 7	Site 9			
1000	100%	100%	100%	100%	100%	100%	100%			
500	75.21%	66.37%	69.09%	82.15%	93.65%	77.77%	75.04%			
250	33.67%	29.17%	63.50%	38.42%	44.88%	23.50%	32.64%			
180	21.91%	15.58%	40.28%	22.18%	25.93%	17.22%	21.31%			
150	19.42%	14.68%	36.75%	20.21%	23.29%	14.35%	18.63%			
125	15.69%	12.63%	30.01%	16.32%	17.67%	10.16%	13.91%			
90	9.83%	8.62%	19.54%	10.30%	9.58%	4.98%	7.38%			
75	6.67%	6.14%	13.66%	7.04%	5.99%	3.08%	4.66%			
45	1.53%	1.49%	3.53%	1.44%	1.28%	0.56%	0.82%			
35	0.84%	0.90%	2.31%	0.87%	0.87%	0.34%	0.52%			
20	0.23%	0.35%	0.86%	0.23%	0.39%	0.14%	0.21%			

Table 3: Mass fraction passing through each mesh size, for each sampling location.

The data in Table 3 are shown graphically in Figure 4.

*Figure 4:* The fraction passing through each mesh size, at each sampling location.



Again, Figure 4 indicates that the mass distribution of the samples from Site 3 is different from those obtained at the other locations.

Figure 5 shows the weighted relative mass distribution. This figure shows that the difference between the samples from Site 3 and those from other sites is a relative lack of mass at 400  $\mu$ m, with more mass centred on 250  $\mu$ m. The relative mass distribution of particles less than 180  $\mu$ m diameter are very similar. The data in Table 3 and Figure 5 show

that the mass distribution is approximately bi-modal, with approximately 90% of particles, by mass, having diameters greater than 150  $\mu m$ . Figure 5 shows that the soil has at least 2 modes, centred on 350  $\mu m$  and 80  $\mu m$ . However, the distributions at particle sizes less than 150  $\mu m$  is complex and modes at particle sizes less than 20  $\mu m$  cannot be discounted.





The relative mass is weighted by the logarithm of average particle size passing through the larger sieve but retained by the smaller sieve.

#### 3.2 Activity Distribution

The total Am-241 activity contained in each fraction is detailed in Table 4.

Fraction Size	Am-241 Activity of Fraction (Bq)									
(µm)	Site 1	Site 2	Site 3	Site 4	Site 5	Site 7	Site 9			
1000-500	696	571	815	823	190	37	247			
500-250	1694	1248	224	1563	1179	23	819			
250-180	1220	1122	2396	420	697	15	93			
180-150	55	23	150	24	55	5	9			
150-125	204	68	530	57	136	14	35			
125-90	283	187	848	165	270	15	72			
90-75	639	112	985	193	434	7	76			
75-45	3034	848	3850	367	1473	323	63			
45-35	330	249	706	103	611	41	12			
35-20	685	298	1633	331	850	15	30			
<20	386	388	1884	172	923	35	90			

 Table 4: Am-241 activity in each fraction from each site.

*The uncertainty (k=1) for all values is 11%.* 

On several occasions, a new sub-sample had to be prepared and measured because the activity was not observed to be homogeneously distributed in the sample container. That is, if the container was flipped, so that the top of the container, rather than the bottom, was now placed on the detector end-cap, a quite different value of the Am-241 activity was measured. This inhomogeneity was attributed to the presence of a few active particles within the subsample. The total Am-241 activity in an individual sub-sample was approximately 10 Bq, while the Am-241 activity of a 20  $\mu$ m diameter plutonium oxide particle is calculated to be approximately 16 Bq. Therefore, the presence of a single plutonium oxide particle can significantly affect the measured activity. However, the presence of such a particle can be inferred by a second measurement with the container inverted. This is because it is probable that the particle will be closer to one side than the other, producing significantly different values due to the different detection efficiencies of the two geometries. Only those sub-samples showing no inhomogeneity (that is the activities from each side were measured to be within 1 standard deviation of one another) are reported here.

The fractional contribution by activity is shown in Figure 6.



Figure 6: Fractional contribution by activity of each size fraction at each site.

Table 5 details the activity fraction passing through each mesh size, at each sampling location.

Mesh Size	Activity Fraction Passing Through Mesh									
(µm)	Site 1	Site 2	Site 3	Site 4	Site 5	Site 7	Site 9			
1000	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%			
500	92.5%	88.8%	94.2%	80.5%	97.2%	93.1%	84.1%			
250	74.1%	64.4%	92.6%	43.4%	79.9%	88.7%	31.1%			
180	60.9%	42.5%	75.5%	33.5%	69.7%	85.8%	25.1%			
150	60.3%	42.0%	74.4%	32.9%	68.9%	84.8%	24.5%			
125	58.1%	40.7%	70.7%	31.6%	66.9%	82.2%	22.3%			
90	55.0%	37.1%	64.6%	27.7%	62.9%	79.3%	17.6%			
75	48.1%	34.9%	57.6%	23.1%	56.6%	78.0%	12.7%			
45	15.2%	18.3%	30.1%	14.4%	35.0%	17.2%	8.6%			
35	11.6%	13.4%	25.1%	11.9%	26.0%	9.5%	7.8%			
20	4.2%	7.6%	13.4%	4.1%	13.5%	6.6%	5.8%			

 Table 5: Activity fraction passing through each mesh size at each sampling location.

Figure 7 shows the weighted relative activity distribution with particle size.

Figure 7: The weighted relative activity distribution at each sampling location.



The relative activity is weighted by the logarithm of average particle size passing through the larger sieve but retained by the smaller sieve.

Figure 7 shows that samples from sites 3 and 7 differ from the other locations in that both have far less activity associated with particles larger than 150  $\mu$ m. Except for site 9, the sites exhibit similar activity distributions below 150  $\mu$ m in that a large fraction of the activity is associated with particles between 30 and 90  $\mu$ m. However, a significant fraction is also associated with particles less than 20  $\mu$ m diameter. This figure also shows that the activity distribution is similar to the mass distribution in that both are bimodal above 35  $\mu$ m.

#### 3.3 Enhancement Factor

A comparison of the data in Tables 3 and 5 show that, in general, the fraction of activity passing through a particular mesh size is greater than the fraction of mass passing through the same mesh. In most cases, less than 2% of the mass is contained in the fraction < 45  $\mu$ m in diameter, but that same size fraction contains more than 15% of the Am-241 activity.

The large-scale mapping of radioactive contamination levels at Maralinga yield activity concentrations for the bulk soil. However, any inhalation dose will arise from particles with diameters less than a particular size, typically taken as 7  $\mu$ m. In order to estimate the activity concentration of small particles, we define the **enhancement factor**.

The enhancement factor is defined as the ratio of activity concentration of a size fraction of the soil to that of the bulk soil. That is,  $f = \left(\frac{a_i}{m_i}\right) / \left(\frac{\sum a_i}{\sum m_i}\right)$ , where *f* is the enhancement factor,  $a_i$  is the activity and  $m_i$  is the mass of a particular size fraction and the sum is over all fractions.

The enhancement factors for the size fractions into which the soil was classified for this work are detailed in Table 6 and shown in Figure 8.

Fraction	Enhancement Factor									
Size (µm)	Site 1	Site 2	Site 3	Site 4	Site 5	Site 7	Site 9			
1000-500	0.30	0.33	0.19	1.09	0.44	0.31	0.64			
500-250	0.44	0.66	0.29	0.85	0.35	0.08	1.25			
250-180	1.12	1.61	0.74	0.61	0.54	0.46	0.53			
180-150	0.24	0.49	0.30	0.29	0.31	0.33	0.22			
150-125	0.59	0.65	0.56	0.34	0.35	0.63	0.48			
125-90	0.52	0.91	0.58	0.65	0.49	0.56	0.72			
90-75	2.20	0.88	1.19	1.40	1.77	0.66	1.81			
75-45	6.40	3.57	2.71	1.55	4.59	24.15	1.07			
45-35	5.19	8.31	4.10	4.32	21.73	34.67	2.59			
35-20	12.05	10.52	8.07	12.27	26.17	14.68	6.51			
<20	18.37	21.85	15.58	17.59	34.68	46.23	27.14			

Table 6: Enhancement factors for the size fractions from each sampling location.Uncertainties (k=1) are approximately 12% for all values.





Figure 8 shows that for the fractions less than 90  $\mu$ m in diameter, the enhancement factor is, generally, greater than 1 and increases with decreasing particle size. Figure 9 shows the enhancement factor of the fraction passing through each mesh size, from soil taken from each sampling location. This figure shows that as the particle size decreases, the enhancement factor increases, at least down to particles with diameters less than 20  $\mu$ m. Also shown by the dotted line in figure 8 is a simple inverse relationship between particle size and enhancement factor, for particles less than 450  $\mu$ m in diameter.

Figure 9: The enhancement factor of particles passing through each mesh size.



The dotted line shows a simple inverse relationship for comparison.

The simple inverse relationship shown in figure 9 would indicate that the enhancement factor for particles less than 7  $\mu$ m in diameter would be approximately 60 if the line were simply extrapolated. However, a previous study (Williams 1990) has shown that the enhancement factor for particles of respirable size is less than that for particles less than 45  $\mu$ m in diameter. The Williams study indicates that the relationship between enhancement factor and particle size is more complex at particle sizes smaller than those measured by this study.

This study could not reproducibly classify the soils into smaller particle sizes due to the composition of the soils and the limitations of the equipment. This study utilised sonic sieving whereby a vertically oscillating air column lifts particles, then carries them back against the mesh openings at 3,600 pulses per minute. The sonic siever also utilises a tapping action to help clear blinding of near-sized particles and assist in de-agglomerating samples with electrostatic, hygroscopic, or other adhesion problems. This equipment is designed to separate particles down to 5  $\mu$ m diameter. Unfortunately, the high clay content of the samples resulted in agglomeration when sieving below 20  $\mu$ m, resulting in inconsistent fractionation.

The Williams study used a micro-particle classifier wherein smaller, lighter particles are separated from larger, heavier particles using a vertically directed stream of air. The size of the fractionated particles was then determined by optical methods.

While the method employed by Williams was able to efficiently separate sufficient material for radiometric analysis, the complexity of the apparatus meant that it could not be decontaminated. The method employed in this study utilised equipment that could be easily decontaminated.

#### 3.4 Plutonium to Americium Activity Ratios

After processing sub-samples of the <20  $\mu$ m fraction from each site to chemically separate the americium and plutonium from the soil, these elements were electroplated onto discs and measured by alpha-spectrometry to determine the activity concentration of Am-241 and Pu-239. In order to estimate the reproducibility of the leaching processes, two samples from each of site 2 and site 9 were processed. Also, each leached solution was separated into two aliquots, which were processed separately to evaluate the reproducibility of the chemical separation processes.

The results for the Am-241 activity concentrations are detailed in Table 7. Four of the samples had very poor chemical recovery of this element (<20%) and were, therefore, unable to be measured. There is good agreement between the two aliquots in 3 of the five cases where both aliquots could be measured. The significant differences between the aliquots from sites 1 and 3 may indicate the leaching process was not reproducible for these samples. The significant differences between the two samples from each of sites 2 and 9 indicate that either the leaching process is not reproducible or that the sample is inhomogeneous.

	Am-241 (mBq/g)									
Site #	Aliqu	iot 1	Aliq	uot 2	Average					
1	1840	±170	1330	±100	1590	±140				
2	1200	±110	1410	±130	1310	±120				
2	1760	±130	1900	±140	1830	±140				
3	13180	±780	7040	±420	10110	±630				
4	6140	±430								
5	10800	$\pm 740$	9880	±620	10340	±680				
7			181	±47						
9			146	±44						
9			216	±43						

Table 7: Americium-241 activity concentration of the < 20 μm soil fraction from each site.

Blank entries occur where no result was recorded due to poor chemical recovery. The uncertainties quoted have a coverage factor of 2.

The activity concentrations of Pu-239 found in the samples are detailed in Table 8. The consistency of results between the two aliquots indicates that the elemental separation processes are highly reproducible. The pairs of results from sites 2 and 9 are highly discrepant (Z-test scores greater than 5). This may indicate that the leaching process is not reproducible. However, it may also indicate that the samples contained high-activity particles, the number of which in any sub-sample is highly variable. Previous studies (Williams 1990) have discussed the problem of high-activity particles in samples from Taranaki.

Table 8: Plutonium-239 activity concentration of the < 20  $\mu$ m soil fraction from each site.

Pu-239 (mBq/g)									
Site #	Aliq	uot 1	Aliq	uot 2	Average				
1	12950	±620	13470	$\pm 650$	13210	$\pm 640$			
2	8050	±360	7870	±500	7960	±440			
2	10800	±470	11480	±820	11140	±670			
3	76900	±3400	73800	±5120	75350	±4350			
4	33200	$\pm 2400$	34200	±2700	33700	±2550			
5	65600	±2900	66400	$\pm 4600$	66000	±3850			
7	1033	±85	1093	±94	1063	±90			
9	1310	±110	1210	±100	1260	±110			
9	1750	±110	1700	±110	1730	±110			

The uncertainties quoted have a coverage factor of 2.

The ratio of these activity concentrations is shown in the first column of Table 9. The last figure in this column is the mean of the ratios from each sample, together with the standard deviation. While there is some variability between the samples, the ratios have a

very narrow distribution. The americium-241 is produced by the decay of plutonium-241, which was a contaminant in the plutonium used for the minor trials. Assuming the plutonium was produced in 1958, the americium-241 activity has increased by approximately 20% since 1985 (see figure 10), when these ratios were derived in a previous study (Johnston et al. 1988). In order to compare results with this previous study, the right-hand column details the ratios that would have been found in 1985, assuming the americium-241 activity has increased by 20%.

	Pu-239 to Am-241 Ratio							
Site #	2010	Ratio	<b>Ratio Corrected to 1985</b>					
1	8.3	±0.8	10.0	±1.0				
2	6.1	±0.6	7.3	±0.8				
2	6.1	±0.6	7.3	±0.7				
3	7.5	±0.6	8.9	±0.8				
4	5.5	±0.5	6.6	±0.6				
5	6.4	±0.6	7.7	±0.7				
7	5.9	±1.2	7.0	±1.4				
9	8.6	±2.0	10.4	$\pm 2.4$				
9	8.0 ±1.2		9.6	±1.5				
Average	Average 6.9 ±1.2		<b>8</b> .3	±1.4				

Table 9: Plutonium-239 to americium-241 activity ratio.

The figures in the left column are those derived directly from the activity concentration measurements. The figures in the right column have been adjusted to account for the ingrowth of americium-241 since 1985.

The figures in bold italics are the mean and standard deviation of all of the results.

The uncertainties quoted have a coverage factor of 2.

*Figure 10:* The relative activity of plutonium-241 and americium-241 with time.



The two points indicate the activity of Am-241, relative to that of Pu-241, in 1985 and 2011. Note that the activity of plutonium-239 does not appreciably change over this period.

#### 3.5 Comparison with Historic Data

The principle purpose of this study is to determine whether the input data for the dose re-assessment has significantly changed in the two decades since the original assessment was made. While this study did not determine the enhancement factor for the respirable fraction of the samples collected, the data obtained for the coarser fractions can be compared to that contained in the previous study (Williams 1990). The Williams study details data from three locations in the north plume, labelled ZD600, FN and IN (see figure 1).

#### 3.5.1 Mass Distribution

Figure 11 shows the mass contribution of each of the size fractions for samples from both the current sampling sites and the historic sites from Williams' study. This figure indicates that, apart from site 3, the historic mass distributions are not dissimilar to those from this study.

In order to more easily compare the historic and current data, the contributions were averaged. The average for the current data excludes the results from site 3 because its distribution was significantly different from the other sampling sites (see figures 3 and 4). These averages are compared in figure 12.

Figure 12 shows that the current data is very consistent (Z-test scores less than 1.68) with that obtained more than two decades ago. In particular, the contributions of the two smallest fractions from each data set are nearly identical (7.4% and 1.2% compared with 7.3% and 1.2%).







*Figure 12: Comparison between historic and current average mass distributions.* 

The error bars indicate the standard deviation of the data used to calculate the averages.

#### 3.5.2 Enhancement Factors

The enhancement factor for each size fraction for each of the current and historic sampling sites are shown in figure 13. This figure shows that, apart from the data from site 7, the distributions with particle size are very similar.





The enhancement factors from each of the data sets, for each size fraction, were averaged to more easily compare the current data with the historic data. In this case, data from site 7 was excluded as an outlier in the current data (see figures 7 and 8). These averages are compared in figure 14. This figure shows that, except for the 150-90  $\mu$ m fraction, the historic and current enhancement factors are not discrepant (Z-test scores less than 1.68).

*Figure 14:* The average enhancement factor for each size fraction from each of the historic and current sampling sites.



The error bars indicate the standard deviation of the data used to calculate the averages.

#### 3.5.3 Plutonium to Americium Ratio

This study has found that the Pu-239 to Am-241 activity ratio varied between 5.5 and 8.6, with a mean of  $6.9\pm1.2$ , in 2010. In order to compare these results with those measured previously (Johnston et al. 1988), the results were corrected for the ingrowth of Am-241 over the intervening years to give a ratio of  $8.3\pm1.4$  in 1985.

This value is larger than the mean value found in the previous study of  $7.2\pm0.6$ . However, when the uncertainties are taken into account, the values are not discrepant (Z-test = 1.4). Furthermore, the range of corrected values found in this study, 6.6-10.4, are within the range found in the previous study of 6.0-11.0.

# 4. Conclusion

Soil samples from the top 1 cm were collected from 7 sites at various distances from the Taranaki soil removal boundary. These samples were sieved into various size fractions down to particles less than 20  $\mu$ m in diameter.

The mass distribution of these fractions was compared between sampling sites and found to exhibit similar behaviour, except for samples from one of the sites. It was found that the mass distributions were bimodal above 35  $\mu$ m diameter with the modes centred on approximately 80 and 350  $\mu$ m respectively. However, it was observed that the finest fractions exhibited a more complex distribution, indicating other modes at fractions that were not separated in this work.

The mass distributions in the current work were also compared to those obtained in an earlier study (Williams 1990). It was found that the average mass distribution from this work was very similar to the average of those from the earlier work, particularly at the smaller sizes.

The enhancement factors, defined as the ratio of the activity concentration of a particular fraction to that of the bulk soil, of the size fractions were also calculated and found to exhibit similar behaviour. It was found that the enhancement factors were far more variable and that the distributions of enhancement factors with particle size for samples from two sites were significantly different from the distributions from the other sites. It was also found that, for the fractions measured, the enhancement factor exhibited an approximately inverse relationship with particle size. The earlier work found that this relationship did not hold at particle sizes less than those measured for this work.

The enhancement factor distributions with particle size from this work were also compared to those obtained in the earlier work. It was found that the average distribution from this work and that from the earlier work were similar.

Finally, the activity ratio between Am-241 and Pu-239 measured in this work is consistent with that found in earlier studies.

Overall, this work shows no indication that the values underpinning the previous dose assessments are questionable or have changed over the past two decades. Therefore, the dose assessment may continue to use an enhancement factor of 6.2 and a Pu-239:Am-241 activity ratio of 7.2.

# 5. References

- Bronson FL 2003. Validation of the accuracy of the LabSOCS software for mathematical efficiency calibration of Ge detectors for typical laboratory samples, Journal of Radioanalytical and Nuclear Chemistry, Vol. 255 (1):1 137–141.
- Johnston PN, Burns PA, Cooper MB & Williams GA 1988. Isotopic Ratios of Actinides used in British Nuclear Trials at Maralinga and Emu, ARL/TR080. Commonwealth of Australia.
- MARTAC (Maralinga Rehabilitation Technical Advisory Committee) 2002. Rehabilitation of former nuclear test sites at Emu and Maralinga (Australia). Commonwealth of Australia: Department of Education, Science and Training.
- Williams GA 1990. Inhalation hazard assessment at Maralinga and Emu. Technical Report 87, ARL/TR087. Commonwealth of Australia.
- Williams GA, Martin LJ & Long SA 2002. Inhalation Dose Assessment for Remediated Land at Maralinga. Australian Radiation Protection and Nuclear Safety Agency, Melbourne. Attachment 6.2 of the MARTAC Report 2002.