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La-oxides as Tracers for PuO_2 to Simulate Contaminated Aerosol Behavior

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ABSTRACT

An analytical and experimental study was performed on the use of lanthanide oxides (La-oxides) as surrogates for plutonium oxides (PuO_2) during simulated buried waste retrieval. This study determined how well the La-oxides move compared to PuO_2 in aerosolized soils during retrieval scenarios. As part of the analytical study, physical properties of La-oxides and PuO_2, such as molecular diameter, diffusivity, density, and molecular weight are compared. In addition, an experimental study was performed in which Idaho National Engineering Laboratory (INEL) soil, INEL soil with lanthanides, and INEL soil with plutonium were aerosolized and collected in filters. Comparison of particle size distribution parameters from this experimental study show similarity between INEL soil, INEL soil with lanthanides, and INEL soil with plutonium.

This work was sponsored by the Office of Technology Development, Buried Waste Integrated Demonstration.

SUMMARY

Results of an analytical and experimental study are presented on the use of lanthanide oxides (La-oxides) as a surrogate for plutonium oxides (PuO_2) during simulated buried transuranic waste retrieval. Buried transuranic waste retrieval will generate aerosolized soils that are contaminated with PuO_2 and americium oxides and controlling the spread of these soils is considered mandatory to an efficient operation. Before performing actual transuranic waste retrieval, "cold" retrieval of simulated waste pits will be demonstrated to establish proof of concept. Since actual radioactive surrogates such as uranium or thorium are disallowed for the retrieval studies at the Idaho National Engineering Laboratory (INEL), other benign surrogates must be employed to evaluate the spread of contaminants. The lanthanides, or rare earths, offer an attractive choice from an analytical standpoint because there are low, naturally occurring levels (a few parts per million) in the INEL soils. Additionally, lanthanides are easily analyzed at the parts per million level by either neutron activation analysis or Inductively Coupled Mass Spectroscopy. Finally, the lanthanides are nonsoluble in water and, therefore, not likely to leach out of a waste/soil seam before the retrieval studies.

A full-scale cold test pit was constructed at the INEL simulating an actual pit in the Radioactive Waste Management Complex (RWMC), where over 56,000 m^3 of transuranic waste is commingled with 170,000 to 225,000 m^3 of soil. In this cold, simulated waste pit, drums and boxes are filled with simulated waste, and each container has a flour form (approximately 3-μm sized particles) of the rare earth tracer at 10 times background. The cold test pit will be used in full-scale, remote retrieval demonstrations in the mid-1990s to support the decision for final disposition of the buried transuranic waste at the INEL.

The chemistry of plutonium is complex, and the evaluation provided here considers only the physical and mechanical movement of plutonium and rare earth tracers with aerosols created by retrieval operations. The effects of size and concentration, gas suspension properties, particle motion, and particle agglomeration were examined. The analytical study showed that the rare earth tracers should move like the PuO_2 with the soils as long as the particle size of the contaminant is similar.[1,a] Particle size effects were found to be the most significant factor and more important then the density of the PuO_2 or La-oxide contaminant.

Experiments were conducted at the Lovelace Inhalation and Toxicology Research Institute (ITRI) in a specially designed aerosol generator and sampling chamber. La-oxides including dysprosium oxide (Dy_2O_3), ytterbium oxide (Yb_2O_3), terbium oxide (Tb_4O_7), and neodymium oxide (Nd_2O_3) were first mixed with INEL soil and then aerosolized to compare particle size distribution. The mix ratios were 300 g of the INEL soil labeled with 16.5 mg each of the Dy_2O_3, Yb_2O_3, and Tb_4O_7 and 150 mg of Nd_2O_3. Additional experiments were conducted using clean INEL soil and INEL soil combined with poly-dispersed PuO_2 particles. A compressed air venturi system was used to aerosolize each soil/oxide mixture. Particle size information was obtained from cascade impactor and multistage cyclone data, while particle concentration data were assessed from filter measurements. The impactor samples involving the rare earth tracers were sent to the University of Illinois for neutron activation analysis.

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a. B. Y. H. Liu, personal communication, September 1993.

In addition to the plutonium data from the ITRI experiments plutonium aerosol data were taken from two test series of the INEL electrostatic curtain experiments.[2,3] The first series were the electrostatic curtain experiments that used a mixture of clean INEL soil and poly-dispersed PuO_2. This soil mixture was prepared by ITRI and had a radioactivity level of less than 2 nCi/g. The second set of electrostatic curtain test data was taken using a blend of Rocky Flats Plant contaminated soil and INEL clean soil with an activity level of less than 2 nCi/g.

The results of this analytical and experimental study show that the rare earth tracers can act as a plutonium surrogate during buried waste retrieval studies. This information will be used to assess how well the contamination control aspects of the retrieval techniques worked so that recommendations can be offered about final deposition of buried transuranic waste.

If the lanthanide labeling of soil in the cold test pit was as thoroughly mixed as the samples for these tests, aerosol data derived from lanthanide quantification will be a useful surrogate for PuO_2. However, if moisture content, inadequate mixing, or other factors produce seed surrogate-oxide particles that have large different size characteristics than the actual PuO_2 waste form, one would not expect the soil/La-oxide aerosol to accurately simulate the aerosolized soil/PuO_2 mixtures. This information will enable the INEL to evaluate various dust suppression techniques and operational procedures without the added expense of working with PuO_2-contaminated soils during the demonstration phase of buried waste retrieval.

This work was sponsored by the Office of Technology Development, Buried Waste Integrated Demonstration.

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La-oxides as Tracers for PuO_2 to Simulate Contaminated Aerosol Behavior

1. INTRODUCTION

Results of an analytical and experimental study are presented on the use of lanthanide oxides (La-oxides) as a surrogate for plutonium oxides (PuO_2) during simulated buried transuranic waste retrieval. Buried transuranic waste retrieval will generate aerosolized soils that are contaminated with PuO_2 and americium oxides, and controlling the spread of these soils is considered mandatory to an efficient operation. Before performing actual transuranic waste retrieval, "cold" retrieval of simulated waste pits will be demonstrated to establish proof of concept. Since actual radioactive surrogates such as uranium or thorium are disallowed for the retrieval studies at the Idaho National Engineering Laboratory (INEL), other benign surrogates must be employed to evaluate the spread of contaminants. The lanthanides, or rare earths, offer an attractive choice from an analytical standpoint because there are low, naturally occurring levels (a few parts per million) in the INEL soils. The chemistry of plutonium is complex, and the evaluation provided here considers only the physical and mechanical movement of plutonium and rare earth tracers with aerosols created by retrieval operations.

This work was sponsored by the Office of Technology Development, Buried Waste Integrated Demonstration.

1.2 Background

A full-scale cold test pit was constructed at the INEL simulating an actual pit in the Radioactive Waste Management Complex (RWMC), where over 56,000 m^3 of transuranic waste is commingled with 170,000 to 225,000 m^3 of soil. In this cold, simulated waste pit, drums and boxes are filled with simulated waste, and each container has a flour form (approximately 3-μm sized particles) of the rare earth tracer at 10 times background. The cold test pit will be used in full-scale, remote retrieval demonstrations in the mid-1990s to support the decision for final disposition of the buried transuranic waste at the INEL.

In preparation for the Cold Test Pit Demonstration, INEL contracted with the Inhalation Toxicology Research Institute (ITRI) to conduct a series of smaller-scale tests in a well-controlled environment. These tests used the La-oxide surrogates in INEL soil to determine the aerosol particle size distributions and concentration of the various lanthanides within these aerosol size distributions. The amount of lanthanide content was determined by instrumental neutron activation analysis (INAA) at the Department of Nuclear Engineering at the University of Illinois. PuO_2 aerosol tests were also run in the same small-scale environment at ITRI with the radiochemistry being performed at the INEL. The testing and measurement techniques including applicable aerosol theory and mass data, were received from ITRI in a draft report.[a] A summary of the lanthanide INAA results was

a. G. J. Newton, A. W. Cronenberg, and D. Yazzie, *PuO_2 Aerosol Characterization for Buried Waste Retrieval Efforts: Use of Rare Earth Simulates*, 1992.

received in a letter report.[b] In addition, data from two INEL aerosol tests from the electrostatic curtain test series using plutonium-contaminated aerosols are included in this report for comparison with the ITRI small-scale tests. The first electrostatic curtain test used Rocky Flats Plant plutonium-contaminated soil mixed with INEL clean soil, and the second test used the same ITRI mixed PuO_2/INEL soil as that used in the ITRI small-scale tests.[1,2]

b. G. J. Newton, M. D. Hoover, A. W. Cronenberg, G. G. Loomis, and S. H. Landsberger, *Use of Lanthanide Oxides as Surrogates for Plutonium in Simulated Waste Retrieval*, Annual Report, AR93INEL, October 25, 1993.

2. OVERVIEW OF ITRI AEROSOL EXPERIMENTS

Experiments were conducted at the ITRI in a specially designed aerosol generator and sampling chamber. La-oxides including dysprosium oxide (Dy_2O_3), ytterbium oxide (Yb_2O_3), terbium oxide (Tb_4O_7), and neodymium oxide (Nd_2O_3) were first mixed with INEL soil and then aerosolized to compare particle size distribution. The mix ratios were 300 g of the INEL soil labeled with 16.5 mg each of the Dy_2O_3, Yb_2O_3, and Tb_4O_7 and 150 mg of Nd_2O_3. The plutonium mixture contained less than 2 nCi/g of PuO_2. A compressed air venturi system was used to aerosolize each soil/oxide mixture. Particle size information was obtained from cascade impactor and multistage cyclone data, while particle concentration data were assessed from filter measurements. The impactor samples involving the rare earth tracers were sent to the University of Illinois for neutron activation analysis.

The experimental apparatus consisted of an aerosol generator, sampling chamber, sample filters, cascade impactor, and cyclone samplers as shown in Figure 1. An on-line, optically-based, mass concentration device monitored aerosol concentration during the test. A cyclone sampler placed at the input removed larger particles (greater than 9 μm).

Separate test runs (test times of about 30 minutes) were conducted with these seed mixtures fed into an ITRI aerosol-venturi generator to obtain a theoretical aerosol mass concentration of 0.2 to 2 g/m^3. The resultant aerosol was delivered to a cylindrical catch container instrumented for aerosol characterization.

Primary test data were obtained with an eight stage cascade impactor using Apiezon L, grease-coated, stainless steel plates (particle size range was 14 to 0.5 μm). Other instruments were used for experiment control and particle morphology.

The fraction of total mass in each aerodynamic size range was determined from gravimetric measurements. The amount of La-oxides collected on the filters was obtained from neutron activation analysis, and the plutonium content was determined by radiochemistry.

Five test runs were made with clean INEL soil to obtain an average for baseline data. Three runs were made for each of the lanthanide/soil mixtures and PuO_2/soil mixture. For the INAA and PuO_2 analysis, the mass from each of the cascade impactor stages for the three test runs was combined to get an average value.

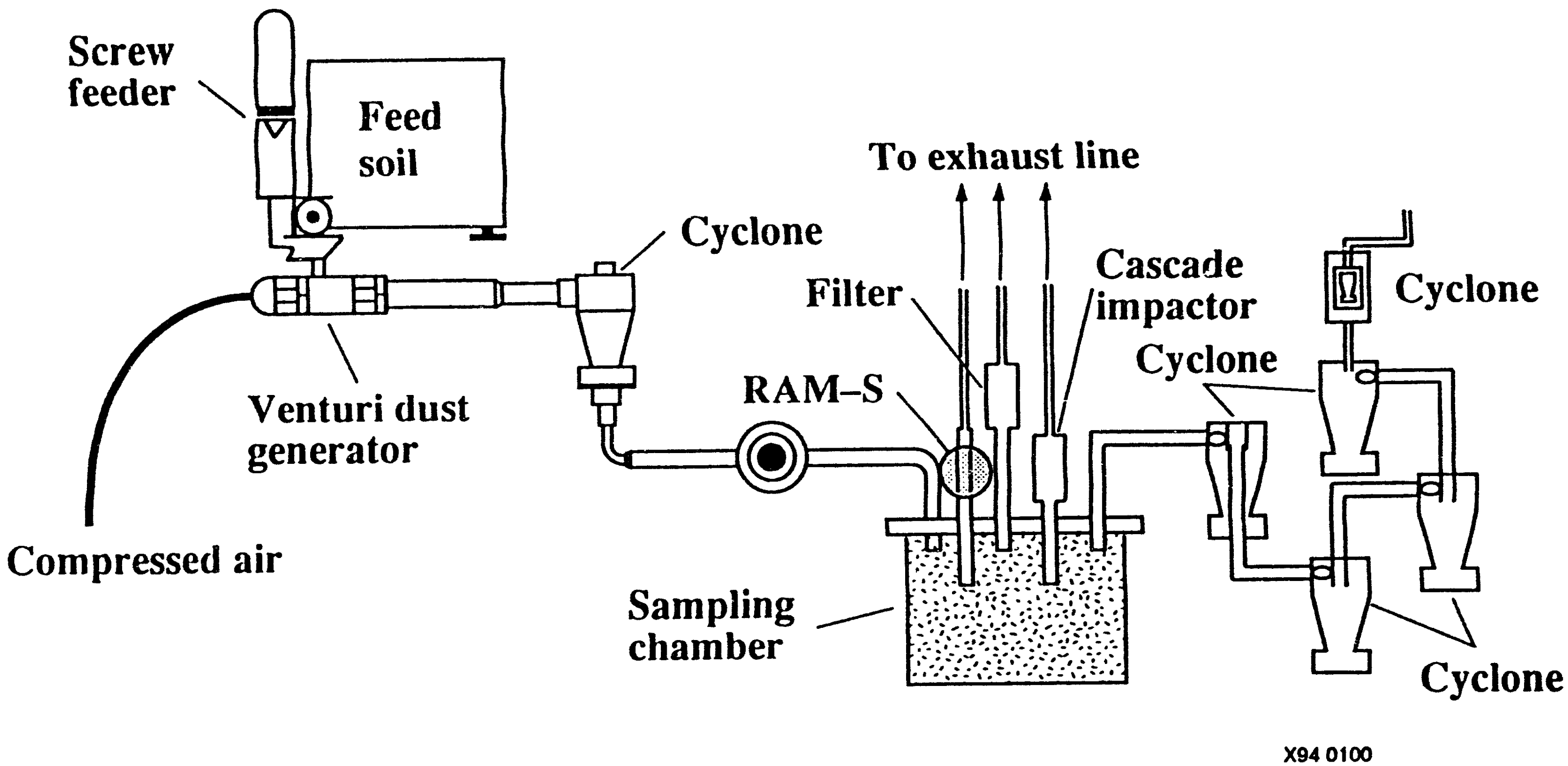


Figure 1. ITRI aerosol generator and measurement configuration.

3. ITRI TEST RESULTS

The results for each of the tests are reported as mass collected on each stage of the cascade impactor, as cumulative percent of mass (activity) greater than particle diameter (effective cut-off aerodynamic particle diameter for that stage), and in log-probability plots. The Log-probability plots are used in aerosol analysis and are a convenient technique to compare the clean, lanthanide, and PuO_2 soil cases. The x-axes are the log of the cumulative fraction (or percent) scale converted to a probability scale, and the y-axes are the log of the effective cut-off diameter for particles collected on a cascade impactor. The probability scale compresses the percent scale near the median (50% point) and expands the scale near the ends so that a cumulative plot of a lognormal distribution will yield a straight line. The median particle size can be read directly from any cumulative plot, and the slope of the straight line is related to the geometric standard deviation. A line with a steep slope is associated with a wide particle size distribution, and a shallow slope is associated with a narrow distribution.

The percent of total mass in each size range for clean INEL soil and the four lanthanide/soil mixtures is given in Table 1. The size distribution for the lanthanide soil mixtures follow the clean soil size distribution well with about 35% of the mass for each type found at 1.4 μ. The percent of La-oxides as determined from the INAA along with the percent of clean soil collected on each cascade impactor stage is shown in Table 2. The Dy_2O_3 had 50% of mass in the range with the 2.2-μm cut-off diameter. The lanthanide particle size distributions show a larger variation in the INAA than for the lanthanide/soil mixture mass data because the lanthanides were only a small part of the lanthanide/soil mixture. The cumulative percent of lanthanide/soil and clean soil greater than the cut-off diameter (d) is given in Table 3, and La-oxides cumulative percent greater than d is presented in Table 4. The spread in Table 4 shows that the Dy_2O_3 had a slightly larger particle size than Tb_4O_7, and they were both slightly larger than Nb_2O_3 with Yb_2O_3 having the smallest particle size. The size distribution parameters for INEL soil labeled with various La-oxides are given in Table 5. The INAA results for each lanthanide and gravimetric data for each lanthanide/soil mixture are presented as log-probability plots in Figure 2. The flatter slope for the INAA data for Dy_2O_3 indicates that it had a smaller particle size distribution. A horizontal line would mean that the aerosol was monodispersed (all one size).

Table 1. Percent total mass in each particle size range for clean soil and four lanthanide mixtures.

ECAD[a] (μm)	INEL clean soil	Dy_2O_3/soil	Yb_2O_3/soil	Tb_4O_7/soil	Nd_2O_3/soil
0.3	4.26	8.31	3.8	4.01	4.84
0.8	21.08	22.15	19.59	20.7	17.26
1.4	35.27	33.95	35.19	35.11	34.21
2.2	28.34	26.2	31.64	29.01	31.2
3.5	9.42	8.38	9.84	10.26	11.13
5.7	1.15	0.72	0.93	0.9	1.14
9	0.31	0.11	0	0	0.22
14.1	0.18	0.21	0	0	0

a. Effective cut-off aerodynamic diameter.

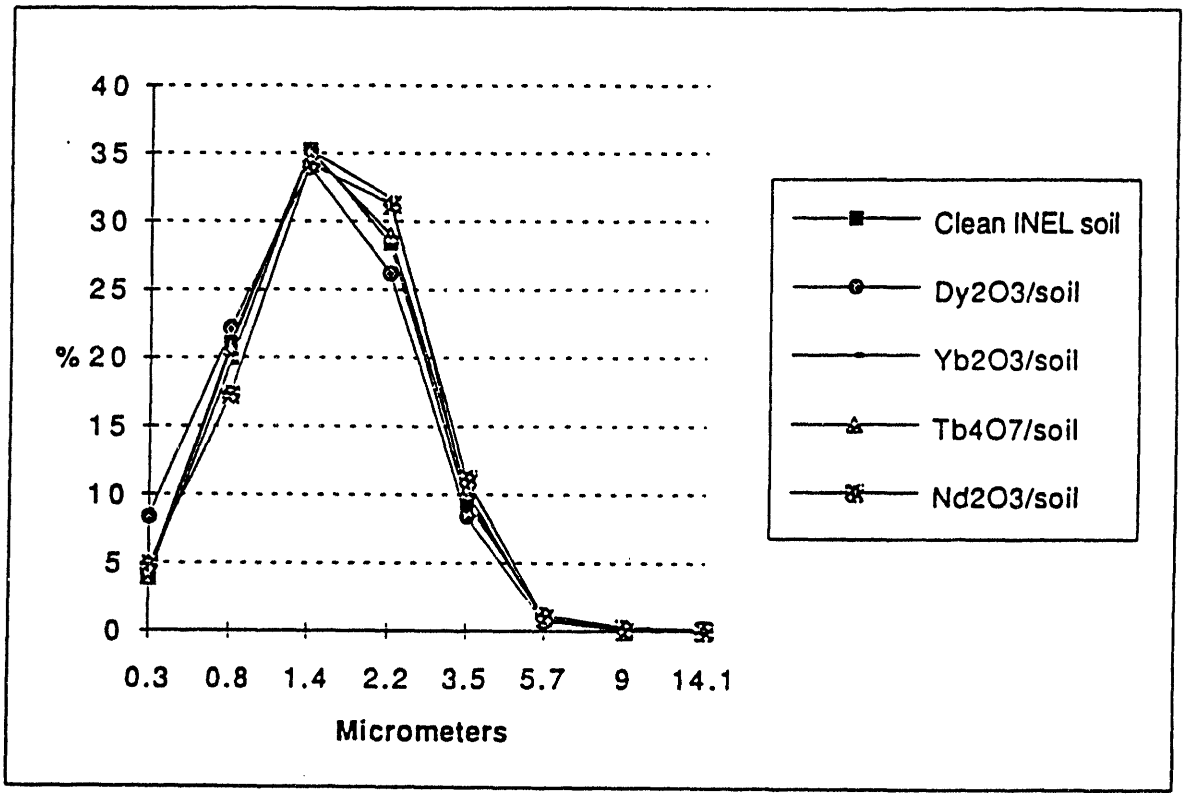


Table 2. Percent of La-oxides (INAA analysis) and clean soil collected on each cascade impactor stage.

ECAD[a] (μm)	Percent mass per stage				
	Yb_2O_3	Tb_4O_7	Dy_2O_3	Nb_2O_3	Clean soil
0.3	5.6	0.3	0	2.6	4.26
0.8	31.1	11	3.4	23	21.08
1.4	36.2	38.7	29.8	39.5	35.27
2.2	22.7	39.3	50.9	27.7	28.34
3.5	4.1	10.2	15.2	6.7	9.42
5.7	0.3	0.5	0.6	0.6	1.15
9	0	0	0	0	0.31
14.1	0	0	0	0	0.18

a. Effective cut-off aerodynamic diameter.

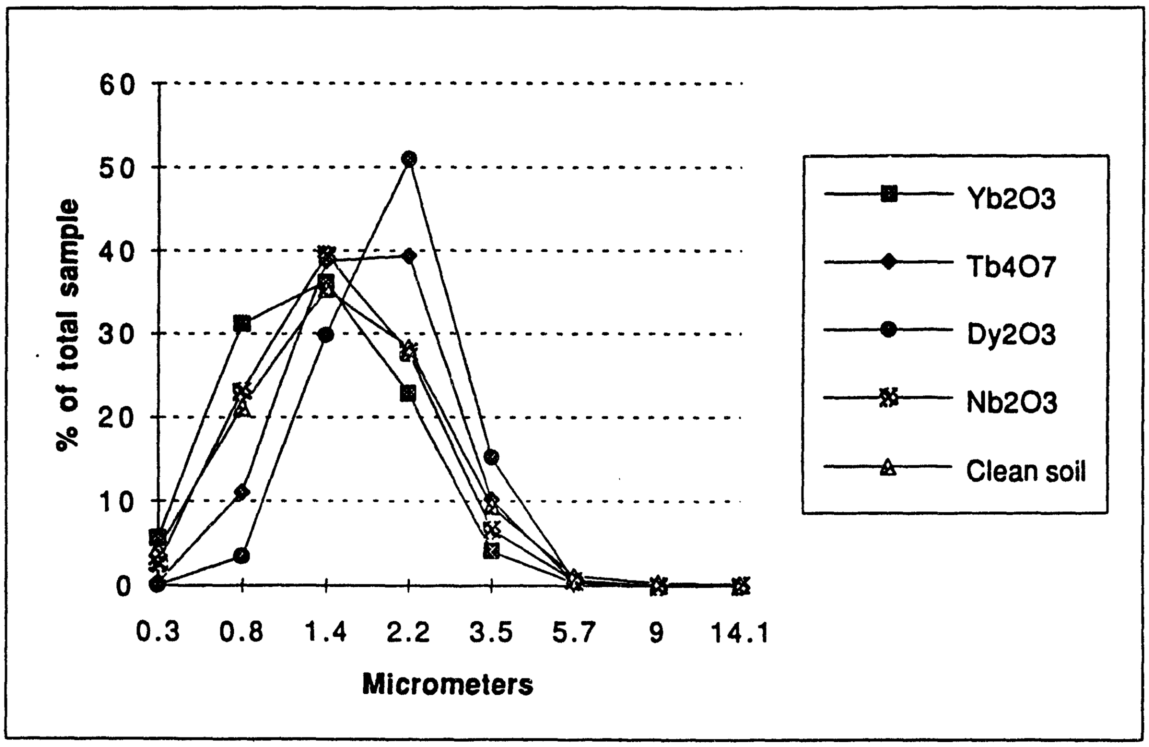


Table 3. Cumulative percent of soil and La-oxides/soil greater than particle diameter.

Micrometers	INEL clean soil	Dy_2O_3/soil	Yb_2O_3/soil	Tb_4O_7/soil	Nb_2O_3/soil
0.3	100.01	100.03	100	100	100
0.8	95.75	91.72	97.19	95.98	95.16
1.4	74.67	69.57	77.6	75.28	77.9
2.2	39.4	35.62	42.41	40.17	43.69
3.5	11.06	9.42	10.77	11.16	12.49
5.7	1.64	1.04	0.93	0.9	1.36
9	0.49	0.32	0	0	0.22
14	0.18	0.21	0	0	0

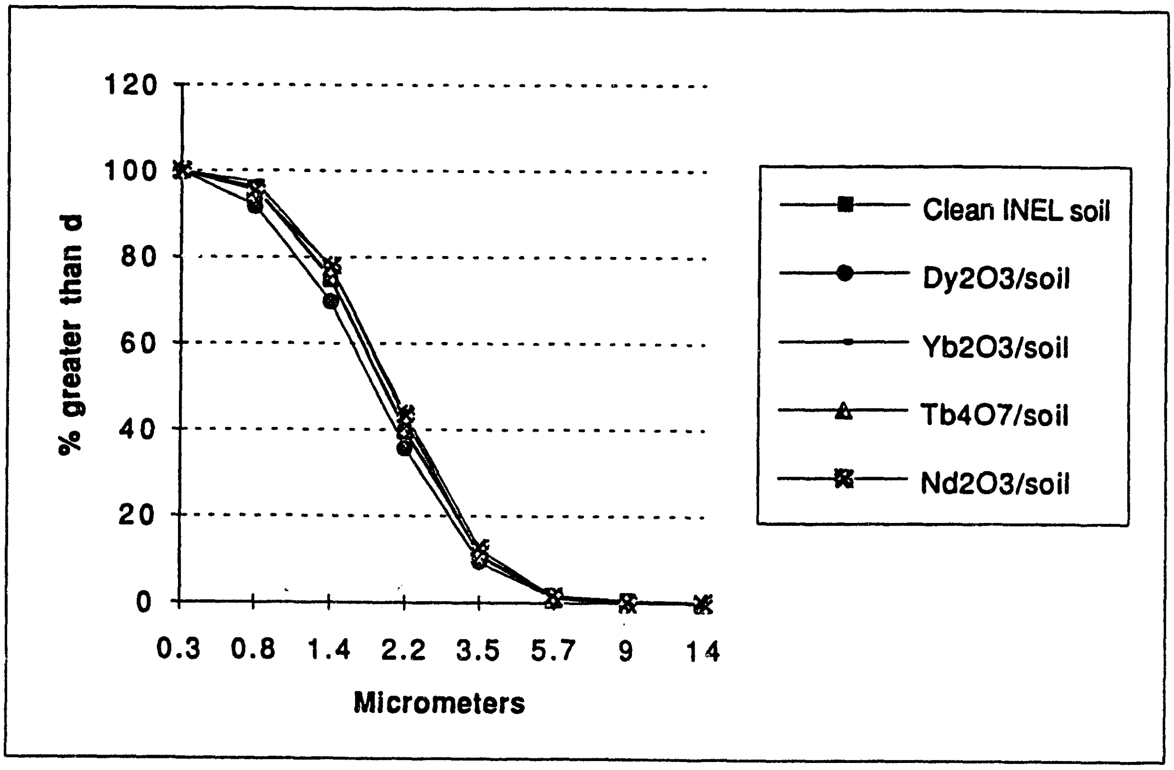


Table 4. Cumulative percent of La-oxides greater than particle diameter.

Micrometers	Yb_2O_3	Tb_4O_7	Dy_2O_3	Nb_2O_3
0.3	100	100	99.9	100.1
0.8	94.4	99.7	99.9	97.5
1.4	63.3	88.7	96.5	74.5
2.2	27.1	50	66.7	35
3.5	4.4	10.7	15.8	7.3
5.7	0.3	0.5	0.6	0.6
9	0	0	0	0
14	0	0	0	0

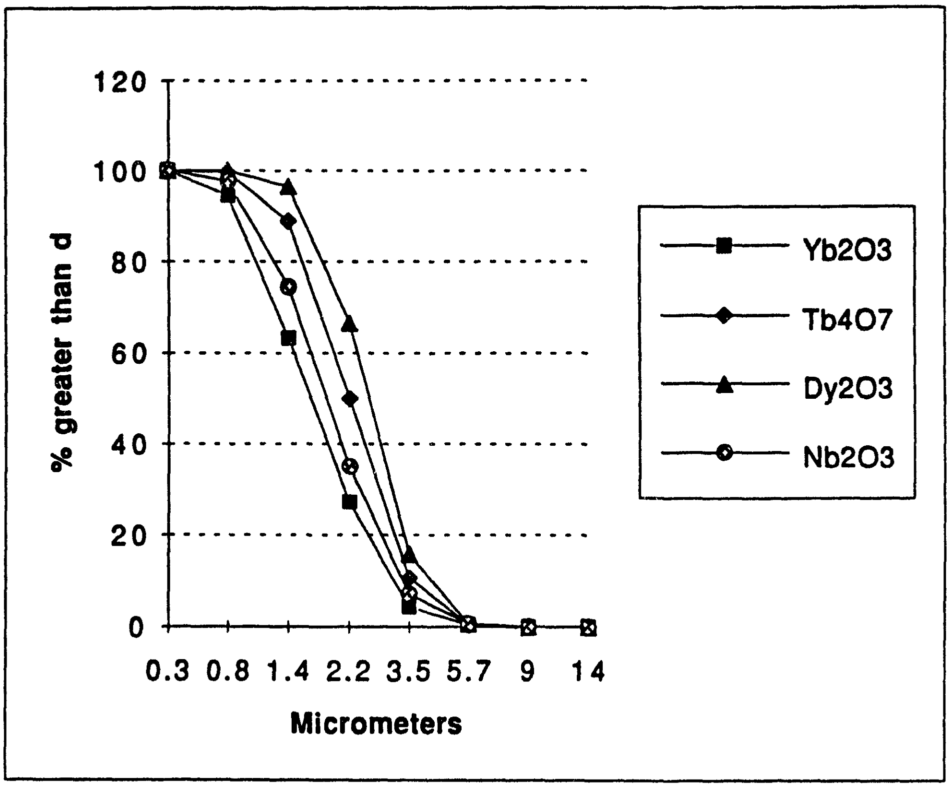


Table 5. Size distribution parameters for INEL soil labeled with various La-oxides.[a]

	Gravimetric		INAA	
	MMAD[b] (μm)	GSD[c] (σm)	MMAD[b] (μm)	GSD[c] (σg)
Yb_2O_3	1.95	1.53	1.63	1.56
Tb_4O_7	1.93	1.53	2.20	1.45
Dy_2O_3	1.84	1.84	2.53	1.38
Nb_2O_3	2.01	1.61	1.86	1.54
Mean ± S.D.	1.93 ± 0.07	1.63 ± 0.15	2.06 ± 0.39	1.48 ± 0.08

a. Aerodynamic size distribution parameters determined from cascade impactor samples by total airborne mass and Instrumental Neutron Activation Analysis (INAA).

b. MMAD = mass median aerodynamic diameter.

c. GSD = geometric standard deviation.

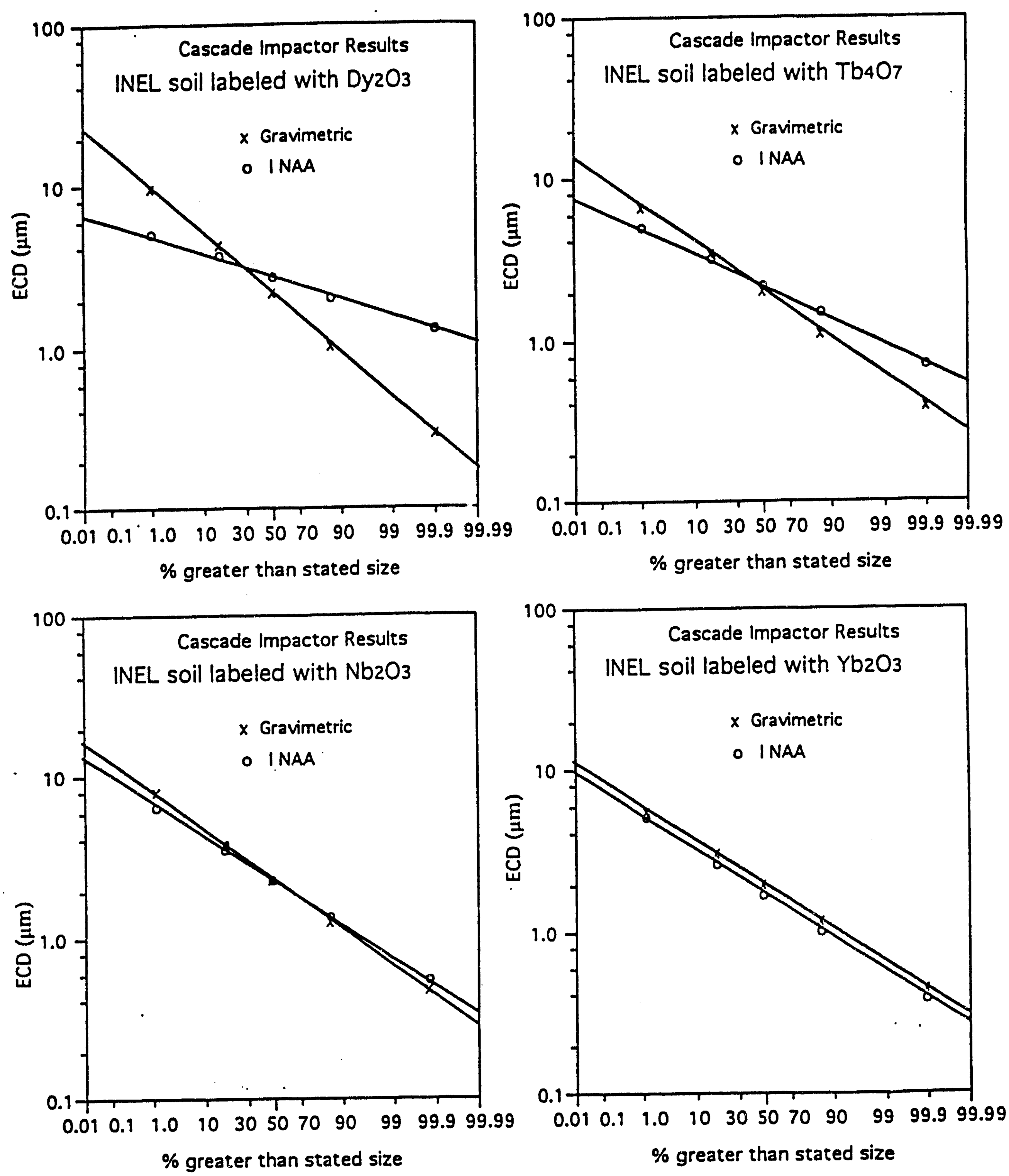


Figure 2. Log-probability plots showing percent greater than stated size for each INEL soil/lanthanide and the corresponding INAA results for each lanthanide.

4. PLUTONIUM/SOIL RESULTS FROM ITRI EXPERIMENTS

Experiments were conducted at ITRI using poly-dispersed PuO_2 particles combined with INEL clean soil. The activity of the mixture was less than 2 nCi/g. The percent of total mass for INEL clean soil and percent of total mass and activity for a three run average of INEL/PuO_2 is given in Table 6. The activity follows the soil size with only a slight shift toward smaller diameters. The percent of total mass and activity greater than particle diameter for the INEL clean soil mass, INEL soil/PuO_2 mass, and INEL soil/PuO_2 activity is presented in Table 7. All three curves follow similar paths indicating that the activity is well dispersed throughout the sample sizes.

Calculations were made to determine the mass mean aerodynamic diameter (MMAD) for the clean INEL soil (1.95 μm) and soil/PuO_2 (2.07 μm) as well as the activity mean aerodynamic diameter (AMAD) for the PuO_2 (1.81 μm). The activity AMAD was less than the soil/PuO_2 by 0.26 μm, and the geometric standard deviation (σg), which is also an indicator of the dispersion was only slightly larger by 0.03. These parameters are given in Table 8 and were also used to obtain the log-probability plots in the three sets of data shown in Figure 3.

The radioactivity per gram was calculated for each sample size (see Table 9). This increase in activity as particle size gets smaller occurs because plutonium is an alpha emitter, and the alpha particles are generated from near the particle surface. As the particle size decreases the surface area increases per gram of mass. Thus the activity varies as the surface area of the particles, and the mass varies as the volume of the particles.

Table 6. Percent of total mass for INEL clean soil and percent of total mass and activity for a three run average of INEL soil/PuO_2 from ITRI data.

ECAD[a] (μm)	INEL clean soil Percent of total clean soil	INEL soil/PuO_2 Percent of total activity	Three run average INEL soil/PuO_2 Percent of total soil/PuO_2
0.3	3.57	6.3	2.52
0.8	20.71	21.37	17.31
1.4	35	39.33	34.62
2.2	30	22.28	27.46
3.5	10	7.62	12.82
5.7	0.71	2.02	3.15
9	0	0.64	1.18
14	0	0.44	0.94
Total	100	100	100

a. Effective cut-off aerodynamic diameter.

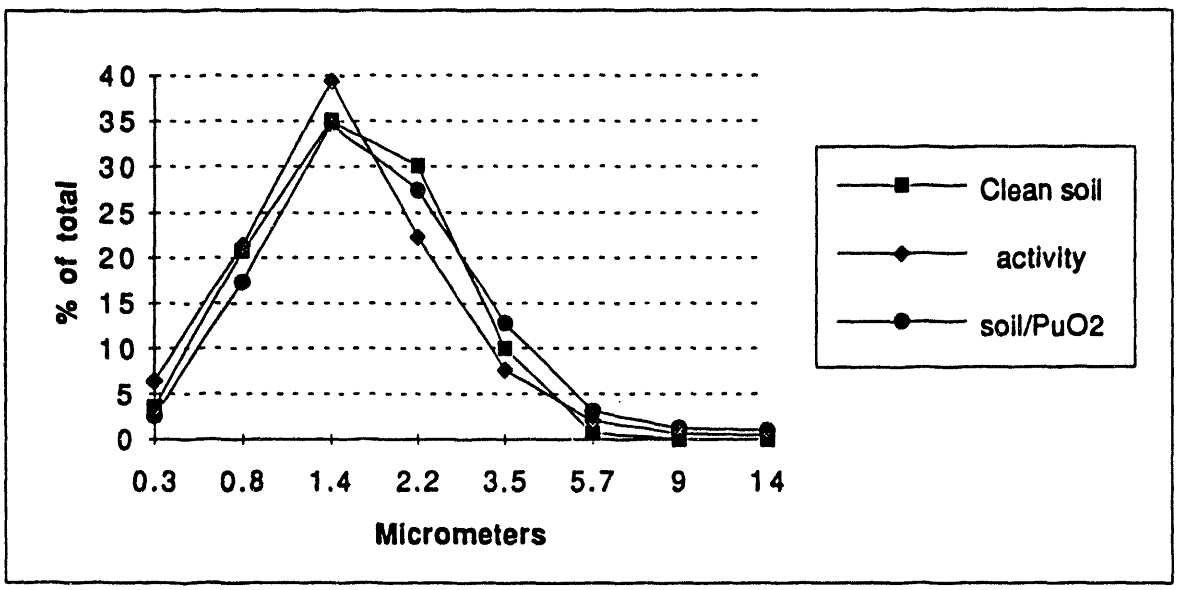


Table 7. Percent of total mass or activity greater than particle diameter for INEL clean soil mass, INEL soil/PuO_2 mass, and INEL soil/PuO_2.

ECAD[a] (μm)	Percent of total mass >d for clean soil	Percent of total >d for soil/PuO_2 activity	Three run average Percent of total mass >d for soil/PuO_2
0.3	100	100	100
0.8	96.42	93.7	97.48
1.4	75.71	72.33	80.17
2.2	40.71	33	45.55
3.5	10.71	10.72	18.09
5.7	0.71	3.1	5.27
9	0	1.08	2.12
14	0	0.44	0.94

a. Effective cut-off aerodynamic diameter.

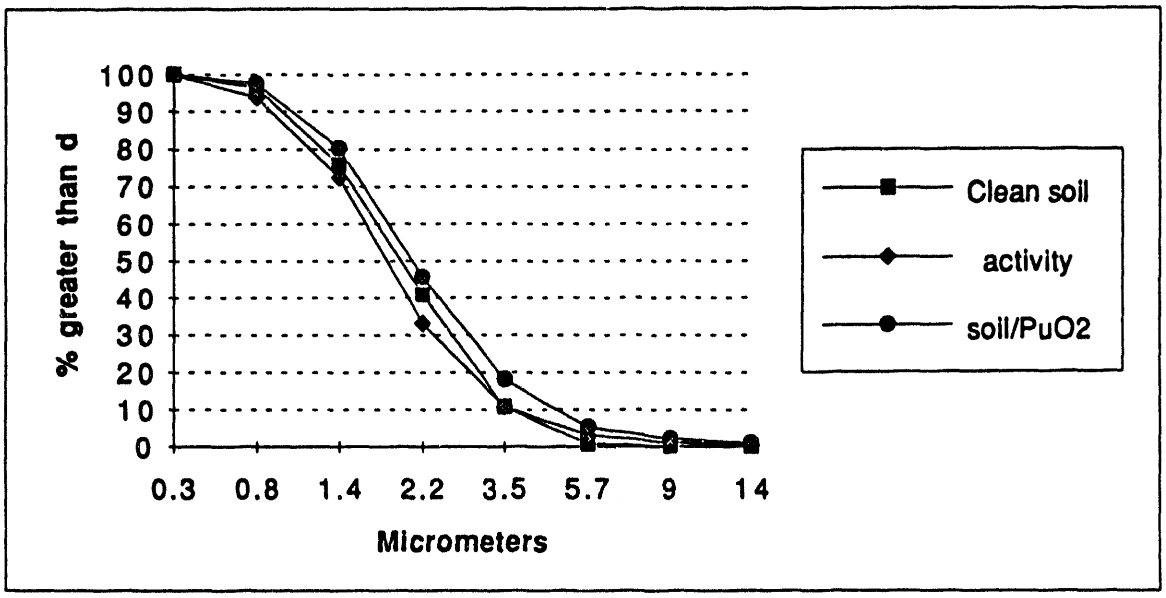


Table 8. Soil/PuO_2 mixture MMAD, AMAD, and associated σg parameters for mass and activity from ITRI test data.

	MMAD or AMAD[a]	σg
Clean soil	1.95	1.72
Soil/PuO_2	2.07	1.86
Soil/PuO^2 activity	1.81	1.89

a. MMAD = Mass median aerodynamic diameter.

AMAD = Activity mean aerodynamic diameter.

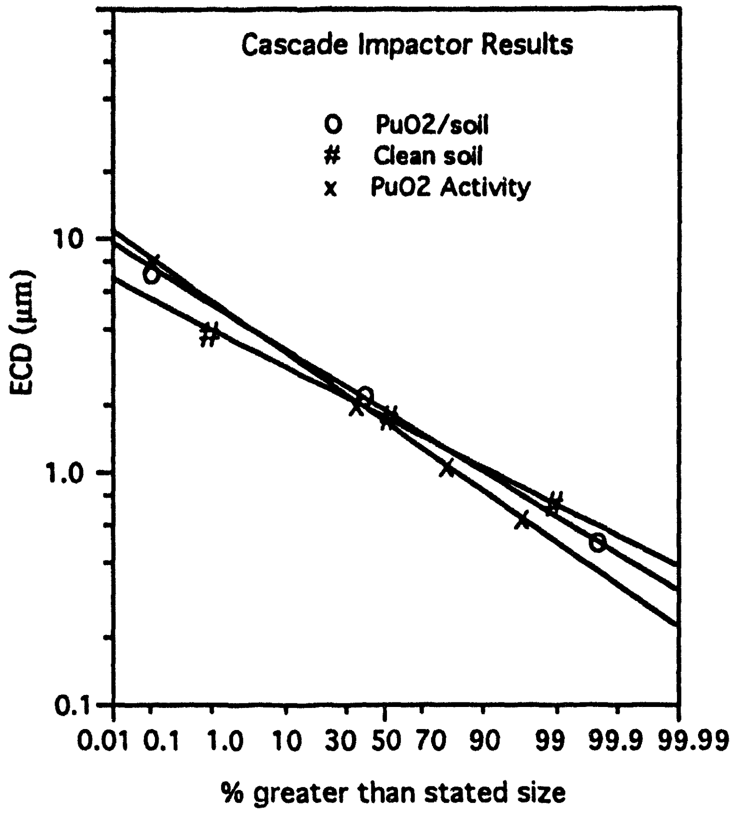
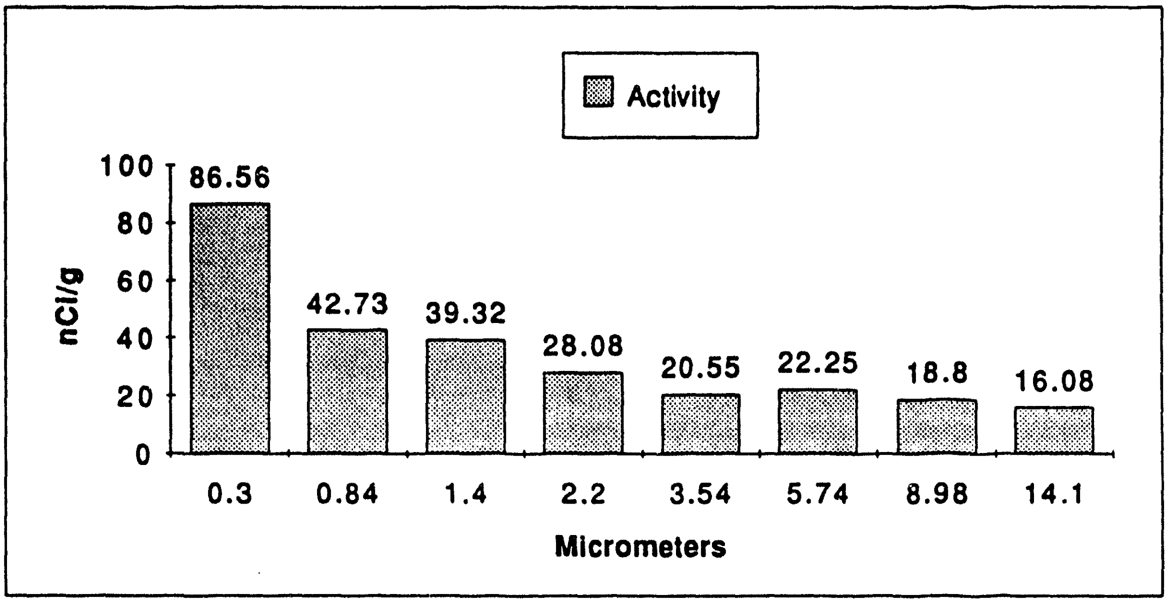


Figure 3. Log-probability plot for ITRI experiment showing PuO_2/soil, INEL clean soil, and activity percent greater than stated particle size.

Table 9. Plot of nCi/g versus particle size for INEL soil/PuO_2 mixture from data collected on ITRI experiment.

Micrometers	nCi/g activity
0.3	86.56
0.84	42.73
1.4	39.32
2.2	28.08
3.54	20.55
5.74	22.25
8.98	18.8
14.13	16.08



5. PLUTONIUM/SOIL RESULTS FROM ELECTROSTATIC CURTAIN EXPERIMENTS

In addition to the plutonium data from the ITRI experiments, plutonium aerosol data were taken from two test series of the INEL electrostatic curtain experiments.[1,2] The first series was the electrostatic curtain demonstration experiments that used a mixture of INEL clean soil and poly-dispersed PuO_2. This soil mixture was prepared by ITRI and had a radioactivity level of less than 2 nCi/g.

The results are presented in the same format as the results from the ITRI experiments. The Andersen cascade eight stage impactor used in these experiments had a preseparator that separated out particles larger than 10 μm. The soil was aerosolized as received; the mean aerosol particle size was about 5.8 μm; and the mean activity was associated with 4.7-μm particles. This is slightly larger than the 2 to 3-μm mean particle size found in the ITRI experiments. The percent mass and activity in particle size range follows a pattern similar to the ITRI experiments as can be seen in Table 10, and the cumulative percent of total mass and activity in particle size range is given in Table 11.

The second set of electrostatic curtain test data was taken using a blend of Rocky Flats contaminated soil and INEL clean soil with an activity level of less than 2 nCi/g. The mean particle diameter was about 6 μm, and mean activity was associated with 5.8-μm particles. The activity and mass followed trends similar to the results from the ITRI experiments and electrostatic curtain experiments using the ITRI mixed soil/PuO_2. The percent of total mass and activity collected on each stage is given in Table 12, and the cumulative percent of total mass as well as activity is presented in Table 13.

The size distribution parameters for these two electrostatic curtain experiments are given in Table 14. These parameters were used to plot the log-probability lines in Figures 4 and 5. The results are similar to those obtained in the ITRI experiments.

Table 10. Percent mass and activity in particle size range for ITRI mixed INEL soil/PuO_2 used in electrostatic curtain demonstration experiment.

ECAD[a] (µm)	Percent of total activity	Percent of total mass
0.2	0.06	0.03
0.4	0.28	0.14
0.7	2.82	1.42
1.1	12.75	4.91
2.1	20.88	11.94
3.3	10.75	17.15
4.7	6.27	12.93
5.8	31.28	28.24
9	14.92	23.24
10	0	0
Total	100.01	100

a. Effective cut-off aerodynamic diameter.

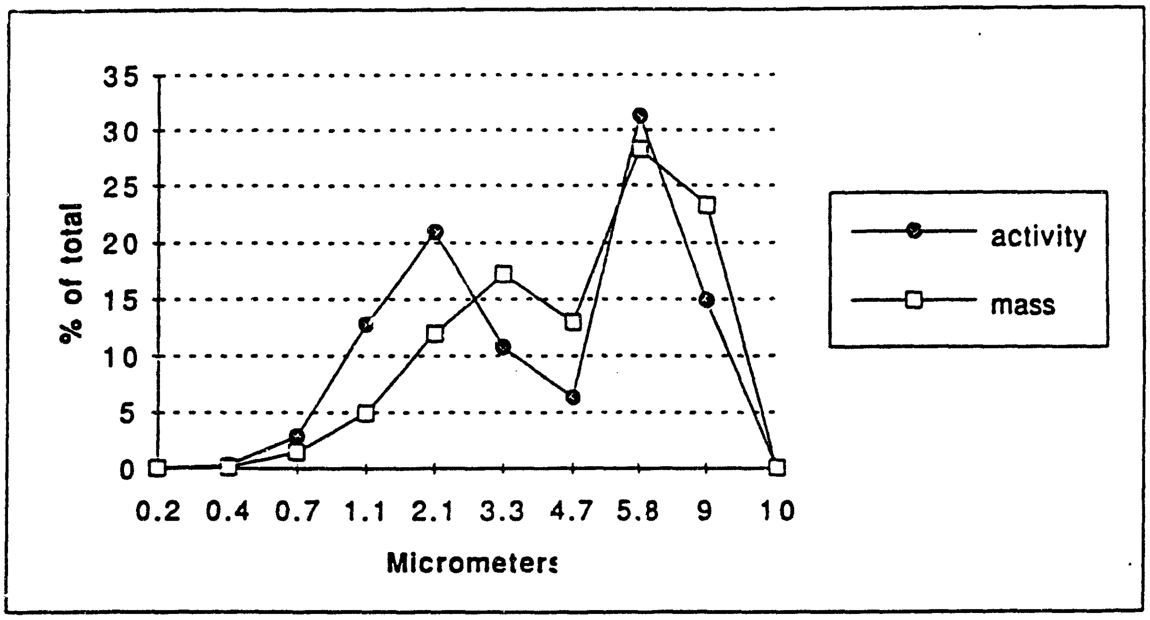


Table 11. Cumulative percent of total mass and activity in particle size range for ITRI mixed INEL soil/PuO_2 used in electrostatic curtain demonstration experiment.

Micrometers	Activity	Mass
0.2	100.01	100
0.4	99.95	99.97
0.7	99.67	99.83
1.1	96.85	98.41
2.1	84.1	93.5
3.3	63.22	81.56
4.7	52.47	64.41
5.8	46.2	51.48
9	14.92	23.24
10	0	0

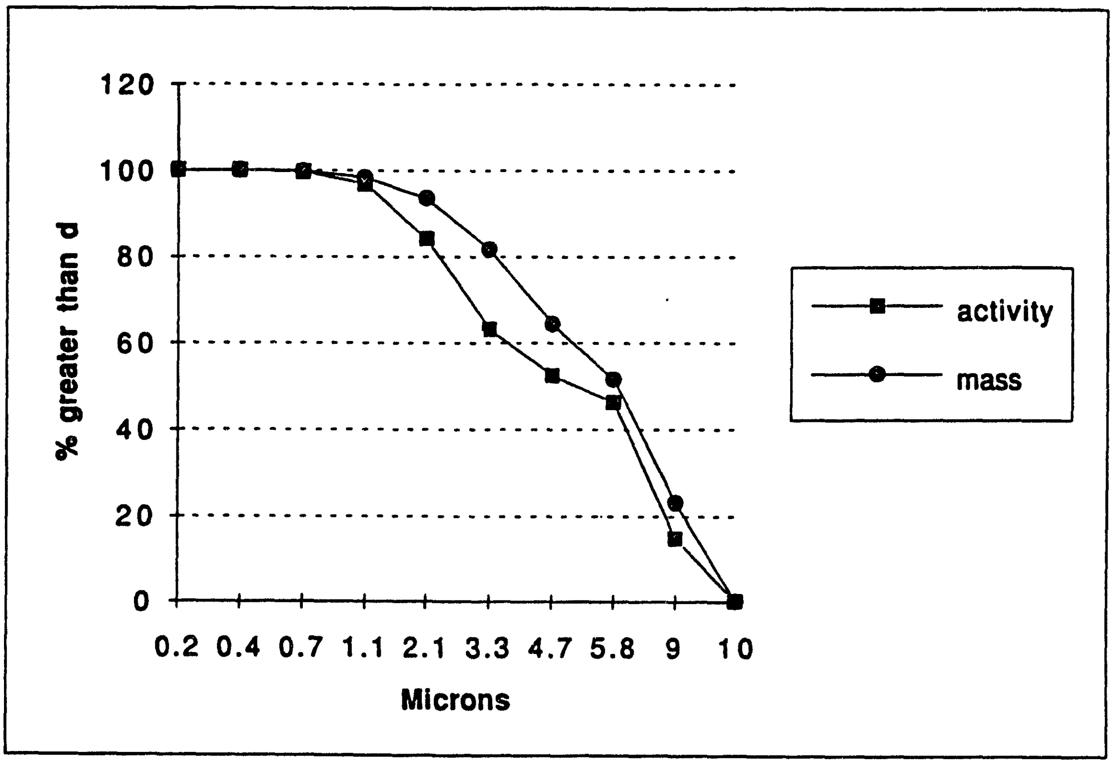


Table 12. Percent of total mass for clean soil and percent of total mass and activity for RWMC/Rocky Flats PuO_2-contaminated soil collected on the INEL electrostatic curtain experiments.

ECAD[a] (μm)	Percent of total mass for clean soil[b]	Percent of total mass for RWMC/Rocky Flats soil[c]	Percent of total RWMC/Rocky Flats soil activity[d]
0	0	0	0
0.4	0.02	0	0
0.7	0	0.11	0.27
1.1	1.07	0.53	1.31
2.1	5.7	4.73	11.94
3.3	17	13.14	25
4.7	13.9	11.46	11.97
5.8	37.6	32.91	28.51
9	24.8	37.12	20.99
10	0	0	0
Total	100	100	100

a. Effective cut-off aerodynamic diameter.

b. Total mass of clean soil collected on the cascade impactor samples was 57.82 mg.

c. Total mass of RWMC/Rocky Flats contaminated soil collected on the cascade impactor samples was 9.51 mg.

d. Total activity collected on the RWMC/Rocky Flats contaminated soil cascade impactor samples was 0.837 pCi.

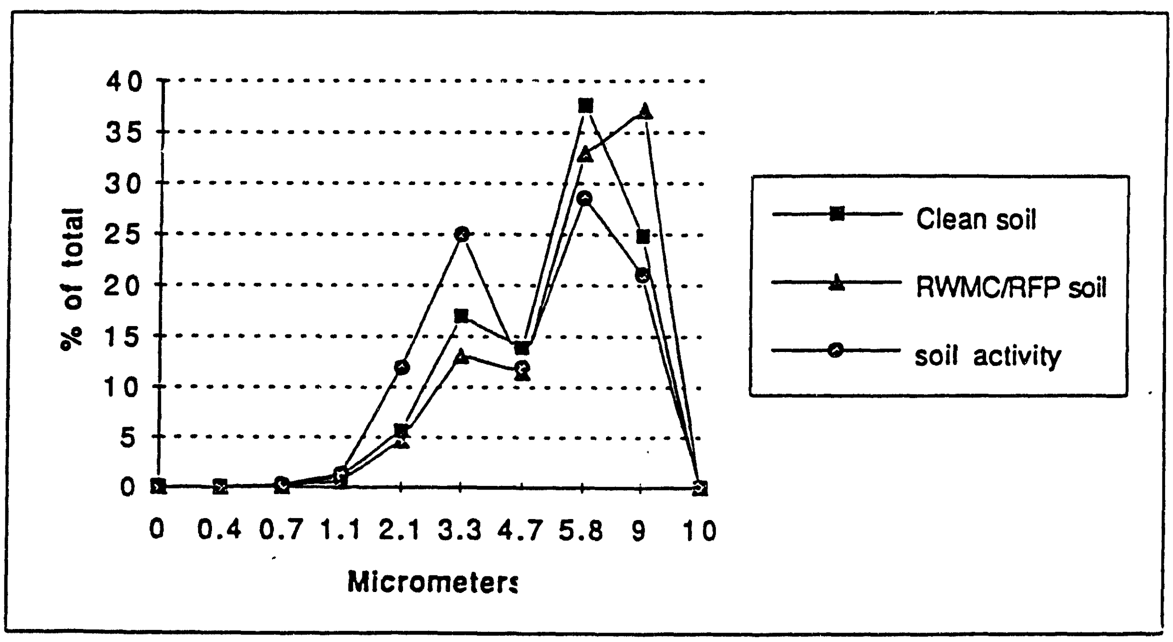


Table 13. Cumulative percent of total mass for clean soil and cumulative percent of total mass and activity for RWMC/Rocky Flats PuO_2-contaminated soil collected on the INEL electrostatic curtain demonstration experiment.

Micrometers	Clean soil	RWMC/Rocky Flats soil	Soil activity
0	100.09	100	99.99
0.4	100.09	100	99.99
0.7	100.07	100	99.99
1.1	100.07	99.89	99.72
2.1	99	99.36	98.41
3.3	93.3	94.63	86.47
4.7	76.3	81.49	61.47
5.8	62.4	70.03	49.5
9	24.8	37.12	20.99
10	0	0	0

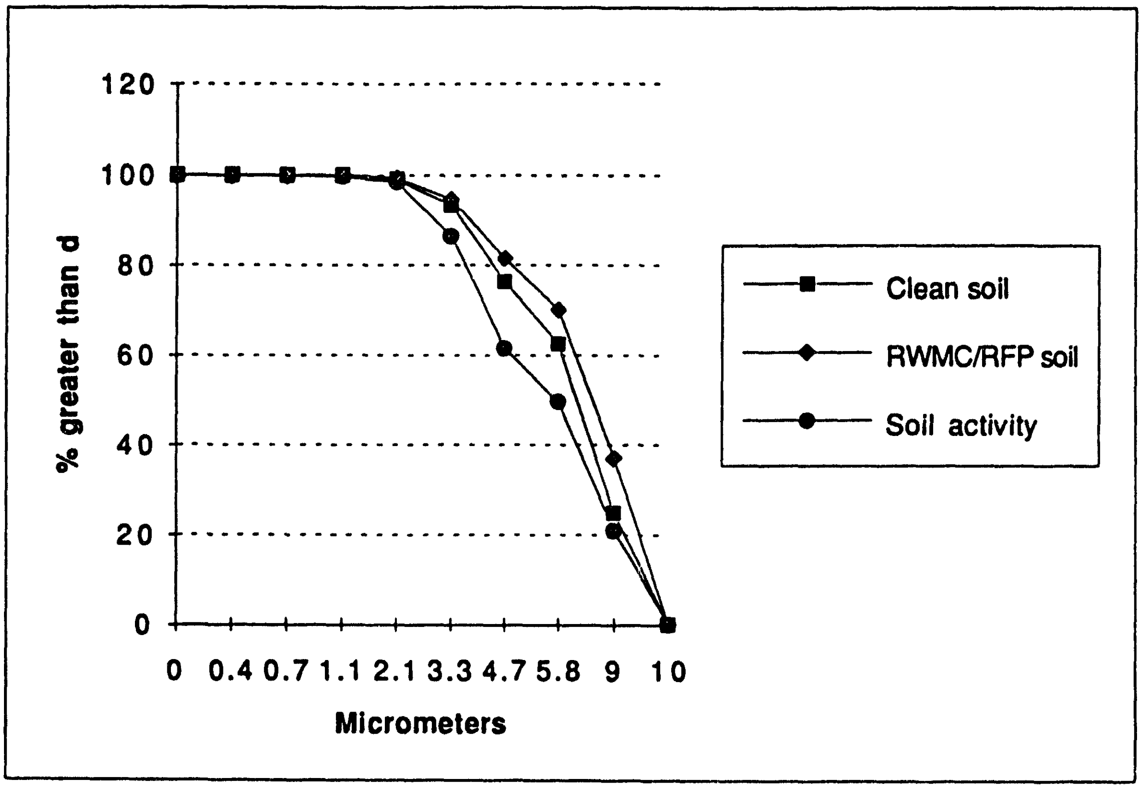


Table 14. Size distribution parameters from electrostatic curtain experiments for INEL soil mixed with Rocky Flats PuO_2-contaminated soil and ITRI mixed PuO_2.

	MMAD (μm)	GSD (σg)
INEL soil/PuO_2 (ITRI mix)	5.94	1.84
Activity INEL soil PuO_2 (ITRI mix)	5.12	2.07
Clean INEL soil	6.7	1.54
RWMC/Rocky Flats PuO_2 soil	7.577	1.54
Activity RWMC/Rocky Flats PuO_2 soil	5.76	1.64

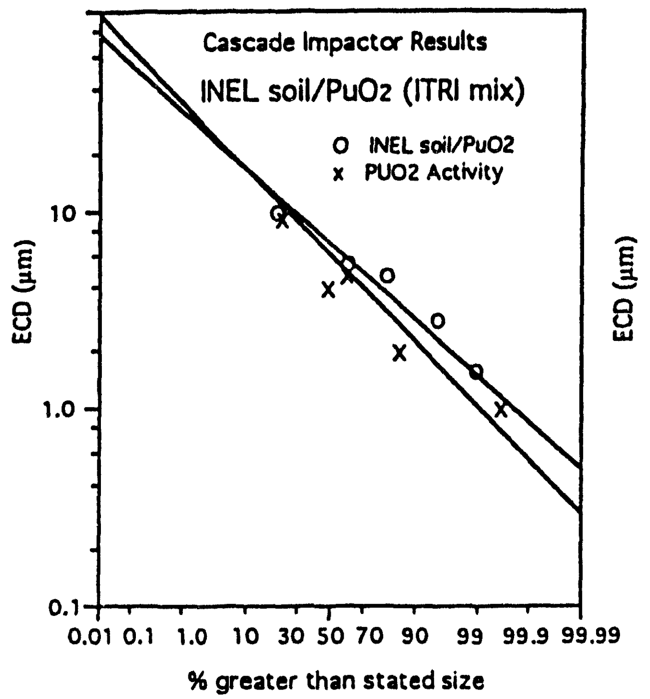


Figure 4. Log-probability plot for INEL soil/PuO_2 (ITRI mix) showing percent greater than stated particle size.

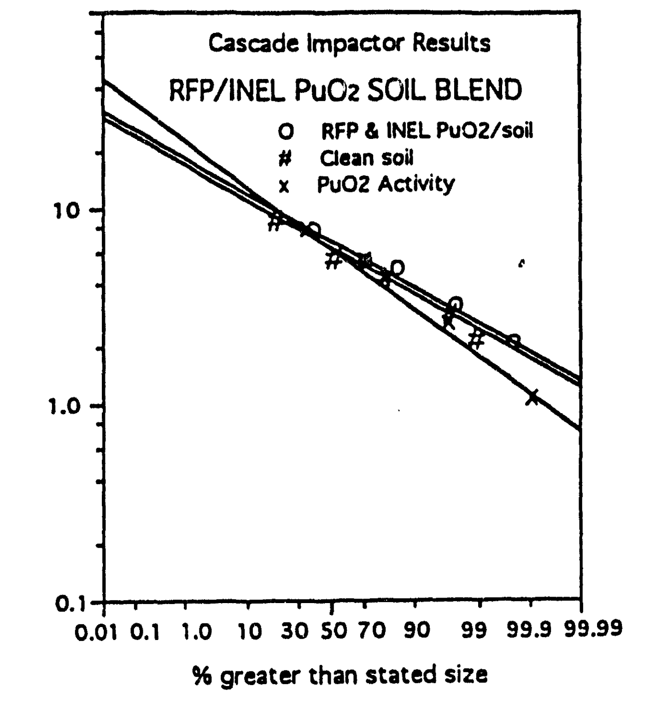


Figure 5. Log-probability plot for Rocky Flats PuO_2-contaminated soil blended with clean INEL soil showing percent greater than stated particle size.

6. AEROSOL THEORY AND ANALYSIS

Environmental factors that may influence particle behavior are humidity and static electricity from charged particles and surfaces. PuO_2 particles are self-charging, and other particles acquire a natural static charge. The static electricity is controlled by using an enclosure that is made of conducting materials. The enclosure and equipment is then electrically grounded. These items were covered in the electrostatic curtain experiments and are not included in this study.

The theoretical analysis included the La-oxide and PuO_2 properties that are needed to accurately predict tracer or PuO_2 molecular aerosol behavior in air. This involves knowledge of their physical, chemical, and molecular properties, of which density, molecular weight, molecular diameter, and diffusivity are of primary importance. The PuO_2 had a higher density than the La-oxides, but density turned out to be less important than the aerodynamic particle size. The physical, chemical, and molecular properties of La-oxides and PuO_2 are important to accurately predict tracer or PuO_2 molecular aerosol behavior in air (see Table 15).

An aerosol is a two-phase mixture of solid particles or liquid suspended in a gas or air. Airborne PuO_2 particles exhibit aerosol behavior. If this aerosol behavior can be simulated by one or more of the La-oxides, it would be a good nonradioactive surrogate for use in the cold waste demonstration pit. The method used in this study to show similar aerosol behavior is to compare important aerosol parameters and verify those parameters by experiment. The important aerosol parameters in addition to molecular properties are

- Particle diameter
- Agglomeration coefficient
- Aerodynamic diameter
- Particle diffusivity
- Density
- Shape factor
- Size distribution
- Slip factor.

The laws governing aerosol behavior are complex and change with particle size. Particle terminal velocity is proportional to the density and particle diameter squared. Thus, particle size is the most important parameter for dictating aerosol behavior with density being of secondary importance.

A cascade impactor measures particle size as the aerodynamic diameter (d_a), which is defined as the diameter of the unit density sphere that has the same settling velocity as the particle.

Table 15. Physical, chemical, and molecular properties of La-oxides and PuO_2 important to an accurate prediction of tracer or PuO_2 molecular aerosol behavior in air.

Chemical composition	Density (g/cm^3)	Molecular weight (g/mole)	Molecular diameter d_m (cm)	Diffusivity estimate (cm^2/s)[a]
Dy_2O_3	8.2[b]	372	5.33 E-8	9.6 E-8
Ty_4O_7	7.7[b]	748	6.87 E-8	1.0 E-7
Yb_2O_3	9.2[b]	394	5.23 E-8	9.18 E-8
Nb_2O_3	7.3[b]	336	5.36 E-8	1.03 E-7
PuO_2	10.5[c]	271	4.42 E-8	8.60 E-8
INEL soil	2.55[d]	66	5.16 E-8	2.20 E-7

a. Diffusivity estimates for a 1-μm diameter particle.

b. UNOCAL-Molycorp., Technical Data Sheet.

c. L. Leibowitz, "Properties for LMFBR Safety Analysis," ANL-CEN-RSD-76-1, April 1976.

d. Department of Metallurgical and Materials Engineering, Colorado School of Mines.

For example, a 1-μm PuO_2 particle would have an equivalent d_a of 2.29 μm; d_a for 1-μm particles of La-oxides ranged from 1.91 to 2.14 μm; and for soil, it was 1.13 μm.

The effects of size and concentration, gas suspension properties, particle motion, and particle agglomeration were examined in the ITRI experiments and analysis. The analytical study showed that the rare earth tracers should move like the PuO_2 with the soils as long as the particle size of the contaminant is similar.[c] Particle size effects were found to be the most significant factor and more important then the density of the PuO_2 or La-oxide contaminant.

c. B. Y. H. Liu, personal communication, September 1993.

7. CONCLUSIONS

La-oxides offer good simulation of PuO_2/soil aerosol behavior, as long as proper attention is given to adequate size simulation of the PuO_2 waste form.

The various La-oxide/soil, PuO_2/soil mixtures and INEL clean soil had similar aerosol size distributions. Thus, the addition of trace quantities of oxide material did not alter basic aerosol dispersion characteristics from that using INEL uncontaminated soil.

These experimental results are supported by analysis, indicating that particle density effects are of second order importance compared to particle size effects.

If moisture content, inadequate mixing, or other factors should produce seed surrogate-oxide particles that have large different size characteristics than the actual PuO_2 waste form, one would not expect the soil/La-oxide aerosol to accurately simulate aerosolized soil/PuO_2 mixtures.

8. REFERENCES

1. L. C. Meyer, *Electrostatic Curtain Studies*, EG&G Idaho, Inc., EGG-WTD-10255, May 1992.

2. L. C. Meyer, *Engineering Scale Electrostatic Enclosure Demonstration*, EG&G Idaho, Inc., EGG-WTD-10988, September 1993.

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