SLUDGE ASH FOR ENVIRONMENTAL AVAILABILITY AND RESPIRABLE ACTIVITY KISKI VALLEY WATER POLLUTION CONTROL AUTHORITY LEECHBURG, PENNSYLVANIA

INTRODUCTION

The Kiski Valley Water Pollution Control Authority (KVWPCA) operates a wastewater treatment plant in Leechburg, Pennsylvania. Solids from the treatment process are dewatered by vacuum filtration and incinerated producing sludge ash. Until 1993, the ash was slurried and transferred to an on-site lagoon for storage. In August 1994, the Pennsylvania Department of Environmental Resources notified the U.S. Nuclear Regulatory Commission (NRC) that they had measured elevated uranium activity in a sludge ash sample obtained from the ash lagoon. A radiological characterization survey of the ash lagoon was performed in December 1994 to determine the extent of radioactive contamination (ORISE 1995). Additional actions are pending the establishment of guidelines for remediation of the site.

OBJECTIVE

At the request of the NRC, the Environmental Survey and Site Assessment Program (ESSAP) of Oak Ridge Institute for Science and Education analyzed five sludge ash samples for environmental availability of uranium, respirable uranium activity and respirable mass. Environmental availability is an indication of the potential for contamination to be available to drinking water, plants or higher organisms.[2] This information is necessary in assessing potential doses from residual radioactive material migrating into the environment. The dose pathway depends on contaminant solubility, which is the characteristic facilitating movement into surface or groundwater, uptake by plants and higher organisms and eventually, human ingestion. Therefore, environmental availability is assessed by measuring solubility of the contaminant. Respirable activity is an indication of potential doses from inhalation of residual radioactive contamination. The potential for inhalation is dependent on the particle size of airborne material. Therefore, respirable activity is measured by separating

particles of a size that may be inhaled and measuring contaminant levels in these particles. Respirable mass measures the quantity in milligrams of sample material in each respirable particle size fraction.

PROCEDURES

Environmental availability was evaluated through two leaching processes, total available uranium (TAU) and readily available uranium (RAU). In each case, an aliquot of an ash sample was mixed with acid for a predetermined time. The resulting leachate was separated from the solids, and the dissolved activity in the leachate solution was measured. TAU was evaluated by a single 16-hour leach with dilute hydrochloric acid. RAU was evaluated using five dilute acetic acid leaches for 24 hours each. The five solutions were combined and the dissolved activity measured to obtain RAU. For RAU, the original procedure prescribed the combination of five 16-hour leaches. After consultation with NRC staff, it was determined to extend the time to 24 hours. All resulting leachates were measured for isotopic uranium activity by alpha spectroscopy.

Respirable activity was determined by separating the sludge ash into constituent particle size fractions and measuring activity in each respirable size fraction. To obtain respirable activity, a 0.5 gram aliquot of dry ash was enclosed in a four liter flask connected in-line to a cascade impaction sampler and calibrated vacuum pump. Air was pulled through the flask to entrain respirable particles. Entrained particles were carried from the flask into the cascade impaction sampler. Respirable particle fractions were separated into eight respirable fraction stages and one submicron, non-respirable fraction in the cascade impaction sampler. Respirable particle size fractions in microns were 0.43-0.65, 0.65-1.1, 1.1-2.1, 2.1-3.3, 3.3-4.7, 4.7-5.8, 5.8-9.0 and 9.0-10. The final, non-respirable fraction was below 0.43 microns. Isotopic uranium activity in each stage and the final fraction was measured using alpha spectroscopy analysis. Respirable mass was measured by a tared glass fiber filter on each stage of the cascade impaction sampler. Loaded and unloaded filters were weighed to a precision of 0.1 milligrams to measure respirable mass (ASI 1984). A diagram of the sampling apparatus is shown in Figure 6.

RESULTS AND DISCUSSION

TAU ranged from 74% to 91%, while RAU ranged from 2% to 4%. The values for TAU indicate that a large fraction of the uranium activity was in a soluble chemical form. However, the relatively low values for RAU indicate that the activity would not dissolve readily under normal environmental conditions. These studies gave a general overview of potential environmental availability. More complex studies would provide more detailed information regarding chemical speciation and potential mobility of uranium contamination (PNL 1994). Values for environmental availability are presented in Table 1.

Measurement of respirable activity indicated that a normalized average of approximately 39% of the activity was present in the 9-10 micron range, 25% in the 5.8-9 micron range, 21% in 4.7-5.8 micron range, 10% in the 3.3-4.7 micron range, 4% in the 2.1-3.3 micron range, 1% in the 1.1-2.1 micron range and 0% in the .43-1.1 micron range. This was demonstrated graphically by the plot of cumulative fraction less than size range which is the sum of all activity present in all respirable particles less than the lower limit of a given particle size range. A comparison of total activity per gram in the sludge ash to the respirable activity per 0.5 grams analyzed gives an average of about 10% of the total activity being respirable. Measurement of respirable mass showed a direct correlation between activity and mass. Values for respirable activity per unit mass of respirable material may be calculated and applied to modeling or *in situ* ambient air samples. Results for respirable activity and mass are presented in Tables 2 through 6. Plots of cumulative fraction less than size range versus particle size ranges sampled are presented in Figures 1 through 5.

Figure 1: Location 35N, 15E (150-170 cm) Cumulative Plot

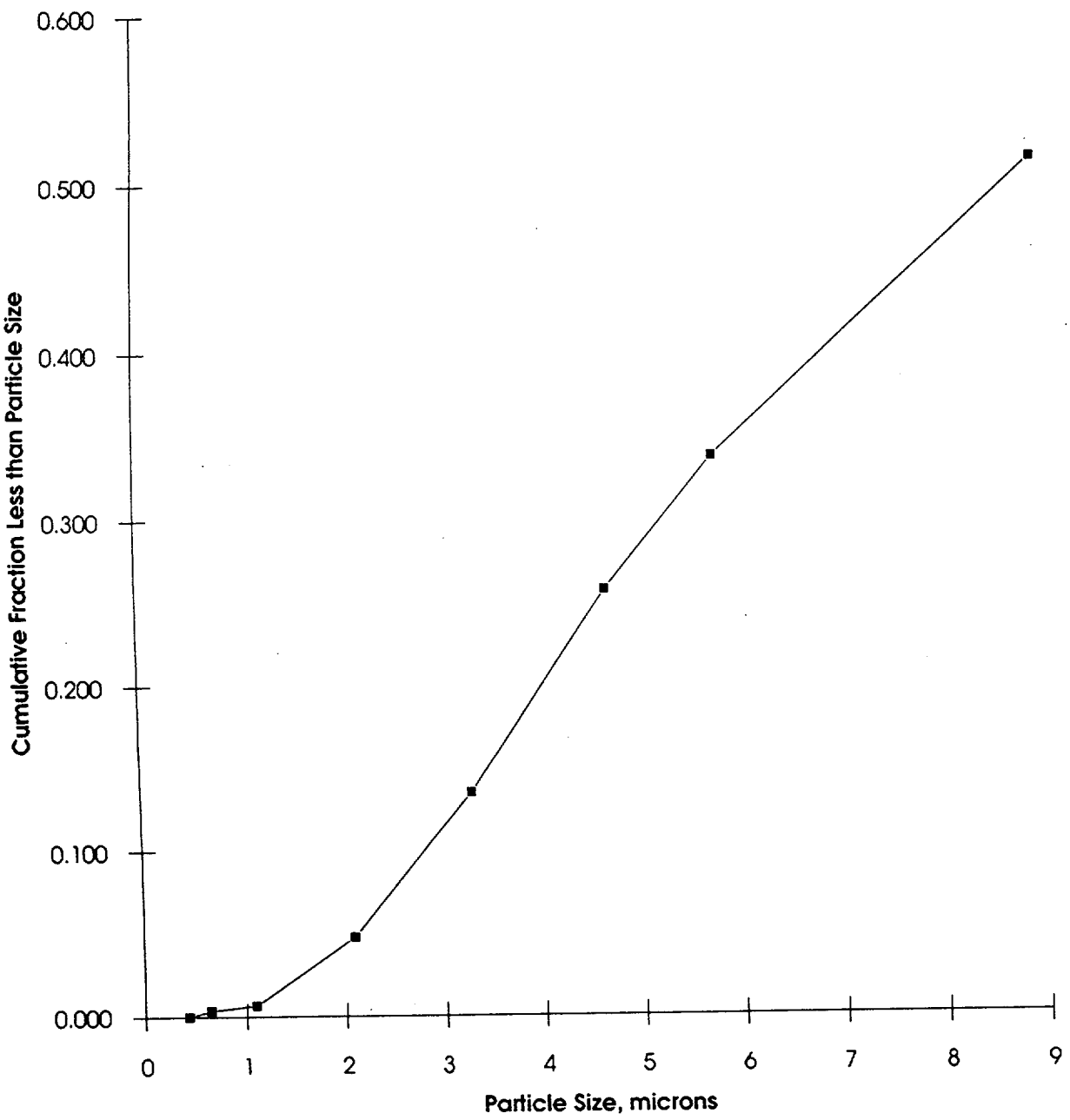

Figure 2: Location 35N, 15E (200-215 cm) Cumulative Plot

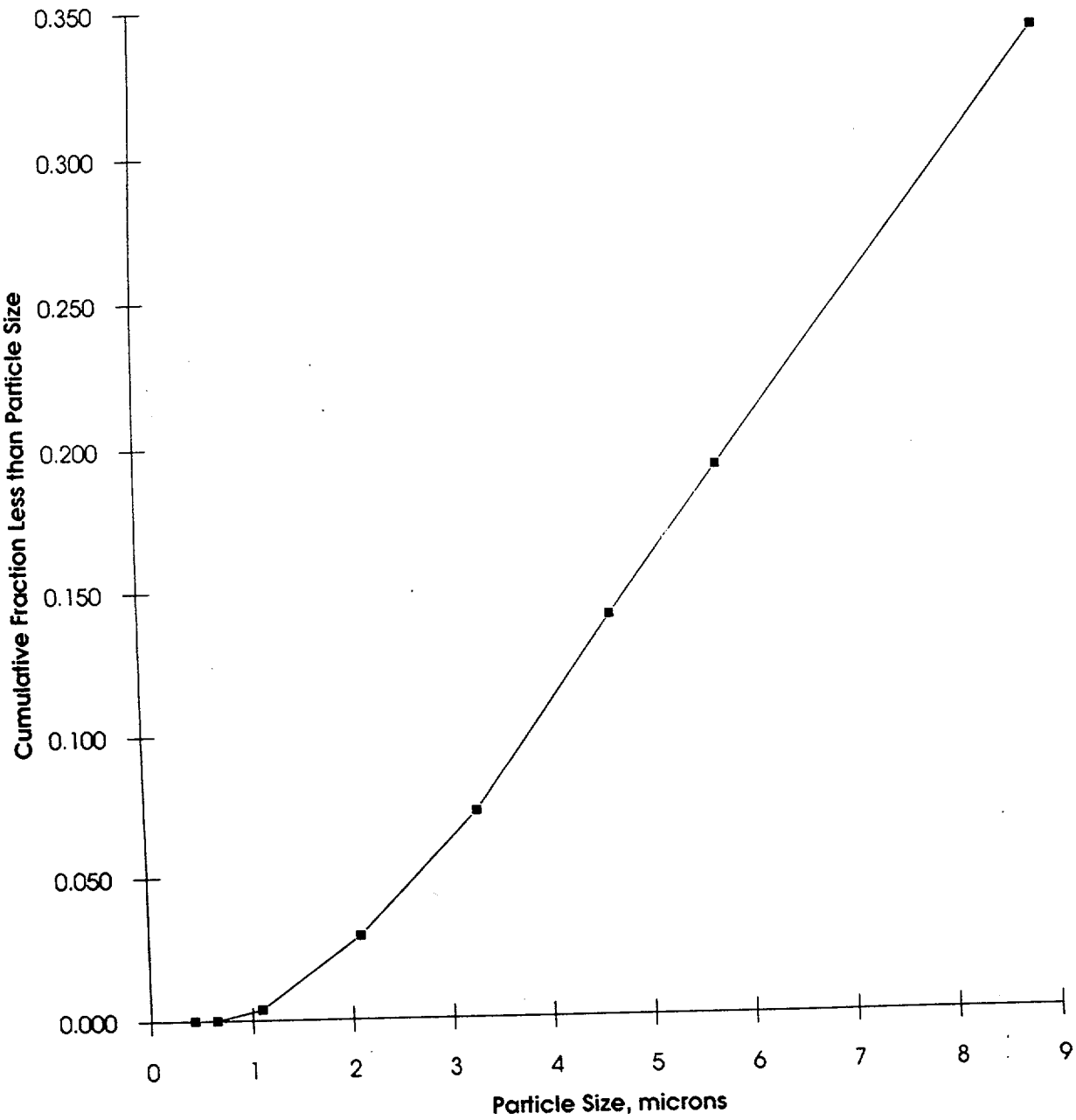

Figure 3: Location 30N, 15E (160-175 cm) Cumulative Plot

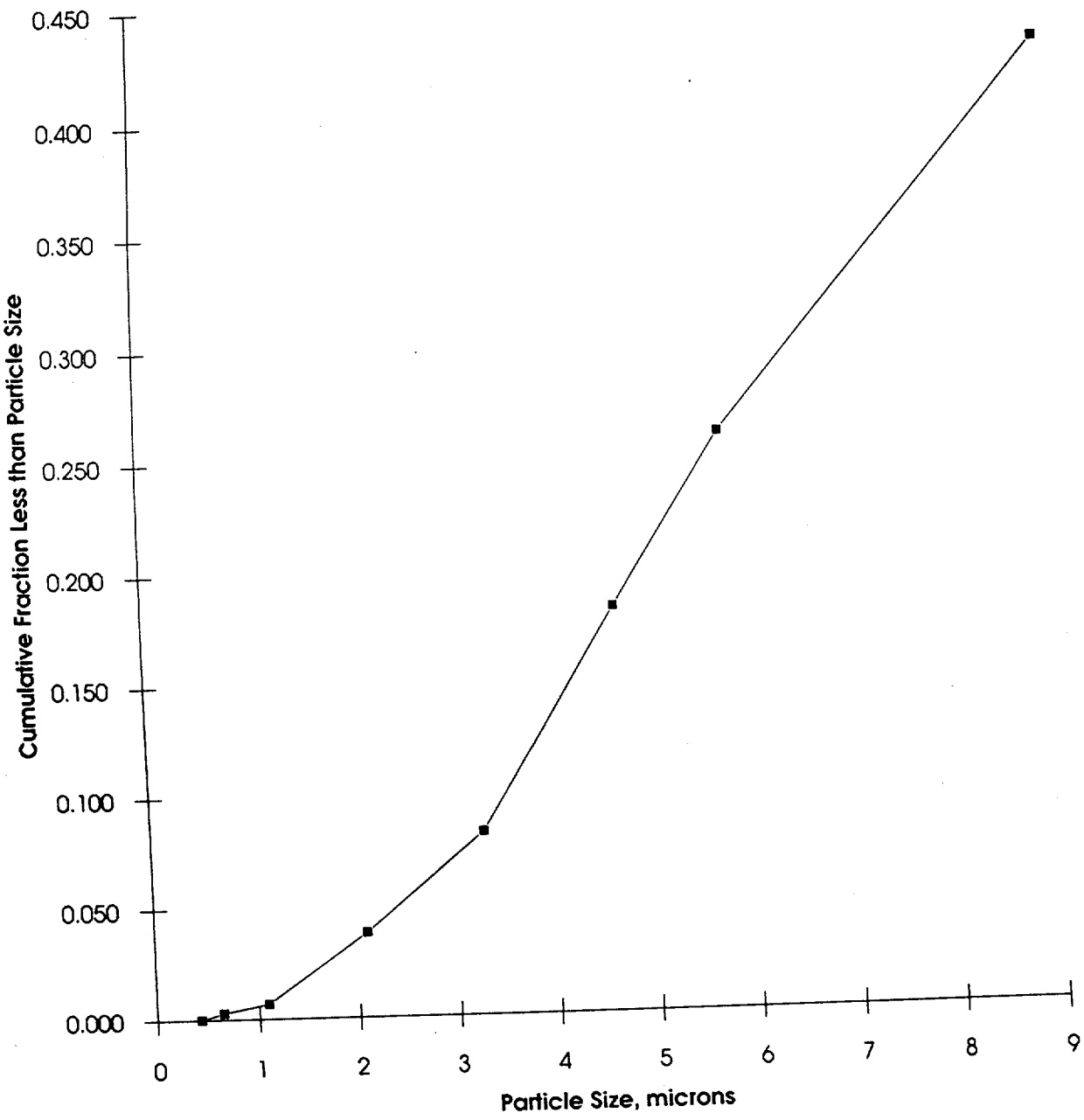

Figure 4: Location 45N, 25E (150-170 cm) Cumulative Plot

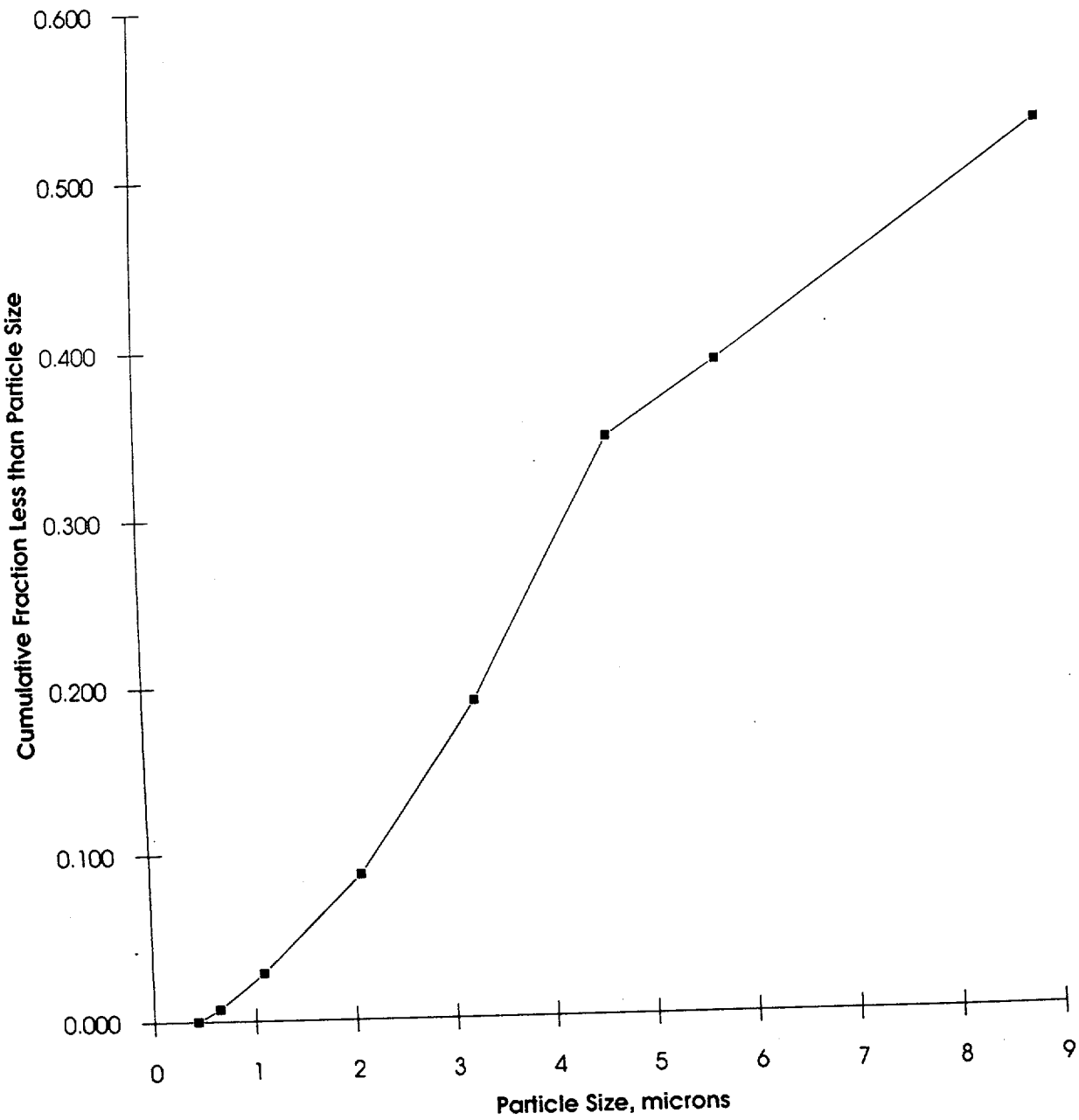

Figure 5: Location 45N, 5E (115-130 cm) Cumulative Plot

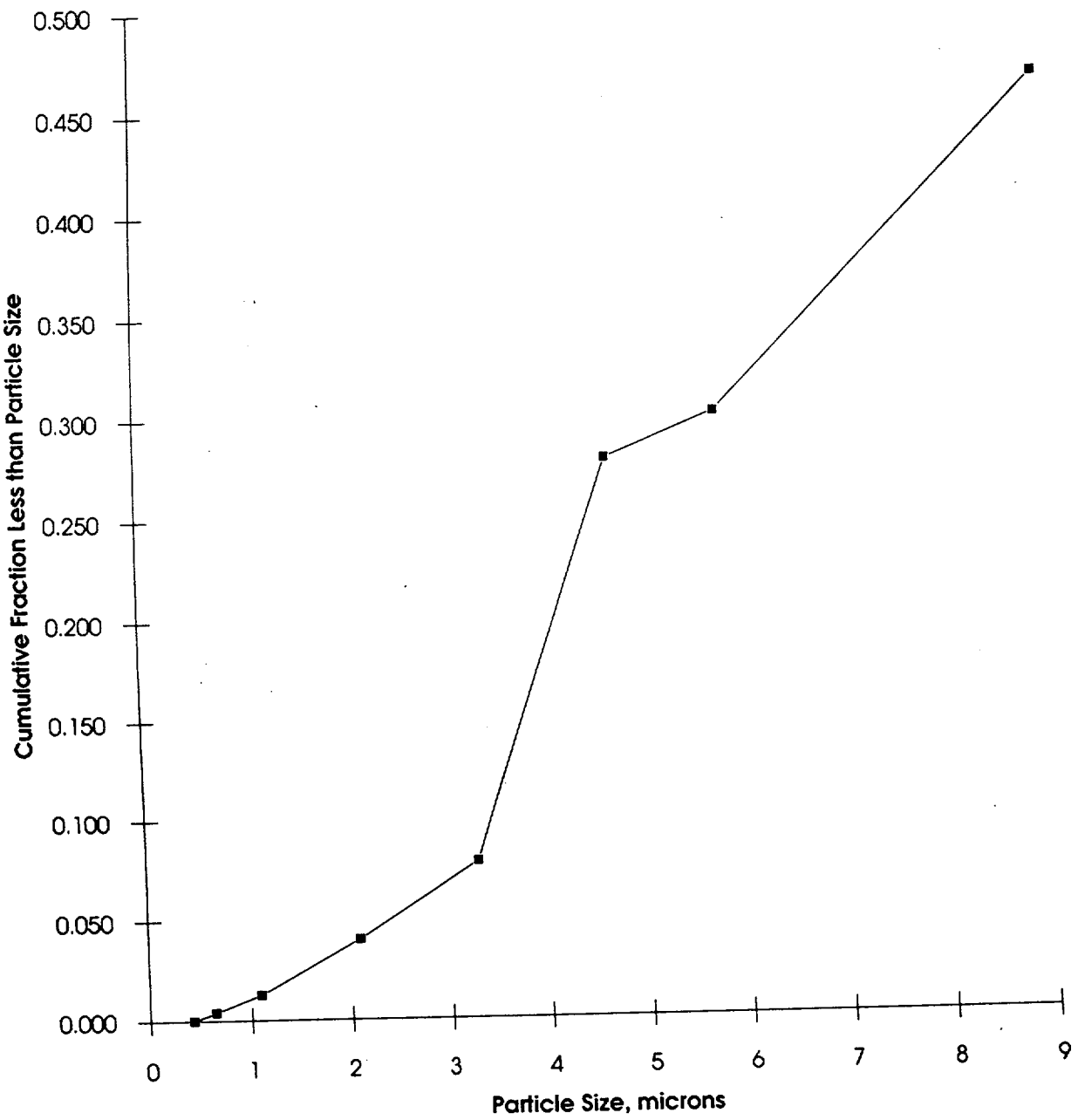

FIGURE 6: Sampling Apparatus for Separation of Respirable Particles in Sludge Ash From Kiski Valley Water Pollution Control Authority Leechburg, Pennsylvania

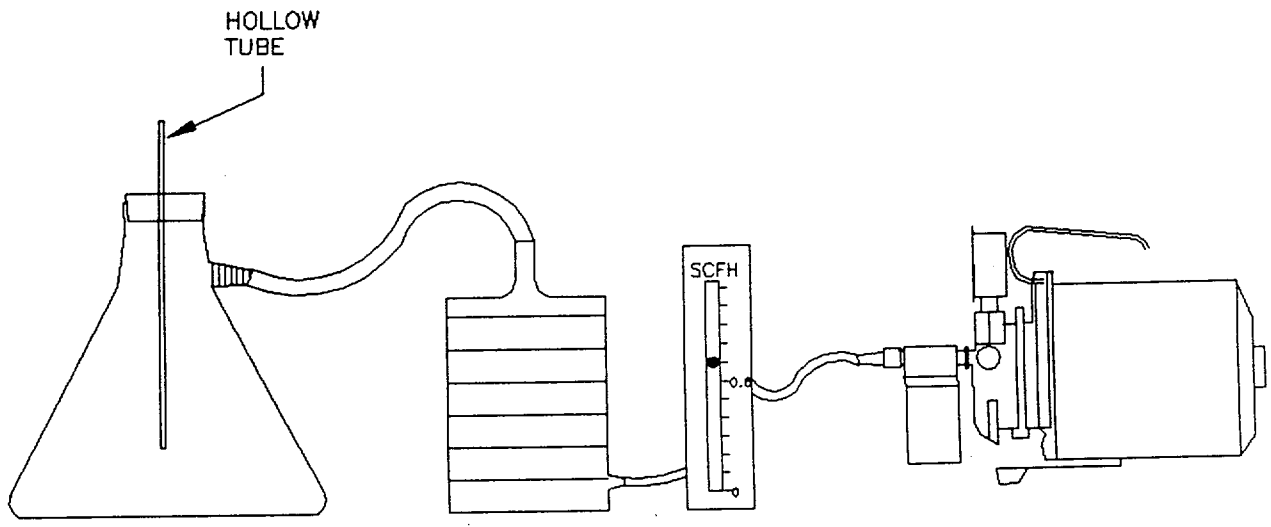

TABLE 1

ENVIRONMENTAL AVAILABILITY IN SLUDGE ASH SAMPLES
KISKI VALLEY POLLUTION CONTROL AUTHORITY
LEECHBURG, PENNSYLVANIA

TOTAL AVAILABLE URANIUM

Location	Depth, cm	Leached Activity, pCi/g				Quantity Analyzed, grams	pCi/ml of Leachate	Fraction of Total Leached	Total Uranium in Sample, pCi/g
		U-234	U-235	U-238	Total Uranium				
35N, 15E	150-170	534.15 ± 37.43	20.71 ± 2.32	103.6 ± 7.97	658.46	0.1919	25.27	0.912	721.71
35N, 15E	200-215	418.57 ± 27.76	17.55 ± 1.89	82.17 ± 6.05	518.29	0.2079	21.55	0.741	699.86
30N, 15E	160-175	594.47 ± 42.39	23.39 ± 2.5	110.3 ± 8.54	78.16	0.2073	30.19	0.874	833.44
45N, 25E	150-170	442.71 ± 31.1	17.33 ± 2.02	73.54 ± 5.85	533.58	0.1918	20.47	0.809	659.36
45N, 5E	115-130	635.56 ± 43.42	26.52 ± 2.68	126.22 ± 9.32	788.3	0.192	30.30	0.854	922.85

READILY AVAILABLE URANIUM

Location	Depth, cm	Leached Activity, pCi/g				Quantity Analyzed, grams	pCi/ml of Leachate	Fraction of Total Leached	Total Uranium in Sample, pCi/g
		U-234	U-235	U-238	Total Uranium				
35N, 15E	150-170	25.78 ± 1.78	1.09 ± 0.11	4.87 ± 0.36	31.74	5.0014	0.794	0.0440	721.71
35N, 15E	200-215	15.98 ± 1.15	0.71 ± 0.08	3.15 ± 0.25	19.84	5.0009	0.496	0.0283	699.86
30N, 15E	160-175	17.92 ± 1.29	0.79 ± 0.09	3.37 ± 0.27	22.08	5.0003	0.552	0.0265	833.44
45N, 25E	150-170	15.28 ± 1.13	0.62 ± 0.08	2.5 ± 0.21	18.40	5.0014	0.460	0.0279	659.36
45N, 5E	115-130	13.64 ± 1	0.59 ± 0.07	2.75 ± 0.23	16.98	5.0004	0.425	0.0184	922.85

TABLE 2

LOCATION 35N, 15E (150-170 cm)
RESPIRABLE URANIUM ACTIVITY AND MASS IN SLUDGE ASH SAMPLES
KISKI VALLEY POLLUTION CONTROL AUTHORITY
LEECHBURG, PENNSYLVANIA

Micron Range	Measured Activity, pCi/g				Respirable Mass, mg	Cumulative Fraction Less Than Size Range
	U-234	U-235	U-238	Total Uranium		
9 - 10	14.13 ± 1.12	0.68 ± 0.15	2.66 ± 0.31	17.47	13.9	0.513
5.8 - 9	5.2 ± 0.53	0.22 ± 0.09	0.85 ± 0.17	6.27	4.9	0.338
4.7 - 5.8	2.34 ± 0.29	0.13 ± 0.06	0.41 ± 0.11	2.88	2.0	0.258
3.3 - 4.7	3.68 ± 0.4	0.17 ± 0.08	0.55 ± 0.12	4.40	3.1	0.135
2.1 - 3.3	2.59 ± 0.32	0.11 ± 0.08	0.44 ± 0.12	3.14	2.3	0.047
1.1 - 2.1	1.21 ± 0.2	0.04 ± 0.05	0.22 ± 0.08	1.47	1.7	0.006
0.65 - 1.1	0.08 ± 0.11	0.03 ± 0.05	<0.07	0.11	0.2	0.003
0.43 - 0.65	0.09 ± 0.06	<0.02	0.03 ± 0.03	0.12	0.0	0
<0.43	0.37 ± 0.1	0.01 ± 0.03	0.45 ± 0.11	0.83	---	---
			Total Respirable Uranium, pCi	35.86	---	---
			Sample Quantity, grams	0.5026	---	---

TABLE 3

LOCATION 35N, 15E (200-215 cm)
RESPIRABLE URANIUM ACTIVITY AND MASS IN SLUDGE ASH SAMPLES
KISKI VALLEY POLLUTION CONTROL AUTHORITY
LEECHBURG, PENNSYLVANIA

Micron Range	Measured Activity, pCi/g				Respirable Mass, mg	Cumulative Fraction Less Than Size Range
	U-234	U-235	U-238	Total Uranium		
9 - 10	30.2 ± 2.31	1.31 ± 0.23	5.73 ± 0.56	37.24	12.3	0.341
5.8 - 9	6.7 ± 0.68	0.36 ± 0.12	1.33 ± 0.23	8.39	4.9	0.192
4.7 - 5.8	2.32 ± 0.29	0.1 ± 0.06	0.5 ± 0.12	2.92	1.4	0.140
3.3 - 4.7	3.12 ± 0.37	0.17 ± 0.08	0.54 ± 0.13	3.83	2.7	0.072
2.1 - 3.3	1.95 ± 0.34	0.13 ± 0.08	0.36 ± 0.13	2.44	2.2	0.029
1.1 - 2.1	1.16 ± 0.2	0.06 ± 0.06	0.22 ± 0.09	1.44	1.1	0.004
0.65 - 1.1	0.14 ± 0.08	0.03 ± 0.04	0.04 ± 0.05	0.21	0.3	0.000
0.43 - 0.65	<0.09	<0.03	<0.02	0.00	0	0
<0.43	0.38 ± 0.12	0.01 ± 0.04	0.36 ± 0.1	0.75	---	---
			Total Respirable Uranium, pCi	56.47	---	---
			Sample Quantity, grams	0.4998	---	---

TABLE 4

LOCATION 30N, 15E (160-175 cm)
RESPIRABLE URANIUM ACTIVITY AND MASS IN SLUDGE ASH SAMPLES
KISKI VALLEY POLLUTION CONTROL AUTHORITY
LEECHBURG, PENNSYLVANIA

Micron Range	Measured Activity, pCi/g				Respirable Mass, mg	Cumulative Fraction Less Than Size Range
	U-234	U-235	U-238	Total Uranium		
9 - 10	17.01 ± 1.41	0.61 ± 0.16	3.28 ± 0.38	20.90	11.8	0.430
5.8 - 9	4.9 ± 0.52	0.21 ± 0.1	1.12 ± 0.21	6.23	3.3	0.260
4.7 - 5.8	2.21 ± 0.29	0.09 ± 0.05	0.53 ± 0.13	2.83	1.6	0.183
3.3 - 4.7	2.94 ± 0.34	0.12 ± 0.07	0.62 ± 0.13	3.68	3.0	0.082
2.1 - 3.3	1.28 ± 0.2	0.06 ± 0.06	0.27 ± 0.09	1.61	2.1	0.038
1.1 - 2.1	0.91 ± 0.17	0.04 ± 0.04	0.21 ± 0.08	1.16	1.0	0.007
0.65 - 1.1	0.13 ± 0.09	<0.04	0.02 ± 0.07	0.15	0.2	0.003
0.43 - 0.65	0.08 ± 0.06	0.01 ± 0.02	0.01 ± 0.01	0.10	0.1	0
<0.43	0.46 ± 0.12	0.02 ± 0.03	0.46 ± 0.12	0.94	---	---
			Total Respirable Uranium, pCi	36.66	---	---
			Sample Quantity, grams	0.503	---	---

TABLE 5

LOCATION 45N, 25E (150-17 cm)
RESPIRABLE URANIUM ACTIVITY AND MASS IN SLUDGE ASH SAMPLES
KISKI VALLEY POLLUTION CONTROL AUTHORITY
LEECHBURG, PENNSYLVANIA

Micron Range	Measured Activity, pCi/g				Respirable Mass, mg	Cumulative Fraction Less Than Size Range
	U-234	U-235	U-238	Total Uranium		
9 - 10	5.63 ± 0.51	0.21 ± 0.08	0.97 ± 0.16	6.81	4.2	0.527
5.8 - 9	1.58 ± 0.27	0.09 ± 0.07	0.3 ± 0.11	1.97	4.6	0.391
4.7 - 5.8	0.64 ± 0.42	<0.16	<0.16	0.64	1	0.346
3.3 - 4.7	1.98 ± 0.27	0.03 ± 0.04	0.24 ± 0.09	2.25	0.3	0.190
2.1 - 3.3	1.25 ± 0.22	0.09 ± 0.06	0.15 ± 0.08	1.49	0.6	0.087
1.1 - 2.1	0.7 ± 0.14	0.01 ± 0.02	0.13 ± 0.06	0.84	0.3	0.028
0.65 - 1.1	0.27 ± 0.09	0.01 ± 0.03	0.03 ± 0.04	0.31	0.1	0.007
0.43 - 0.65	0.09 ± 0.06	<0.02	0.01 ± 0.04	0.10	0	0
<0.43	0.36 ± 0.1	0.02 ± 0.03	0.3 ± 0.09	0.68	---	---
			Total Respirable Uranium, pCi	14.41	---	---
			Sample Quantity, grams	0.5025	---	---

TABLE 6

LOCATION 45N, 5E (115-130 cm)
RESPIRABLE URANIUM ACTIVITY AND MASS IN SLUDGE ASH SAMPLES
KISKI VALLEY POLLUTION CONTROL AUTHORITY
LEECHBURG, PENNSYLVANIA

Micron Range	Measured Activity, pCi/g				Respirable Mass, mg	Cumulative Fraction Less Than Size Range
	U-234	U-235	U-238	Total Uranium		
9 - 10	17.39 ± 1.35	0.71 ± 0.16	3.31 ± 0.36	21.41	15.6	0.465
5.8 - 9	5.23 ± 0.53	0.16 ± 0.07	1.13 ± 0.19	6.52	8.9	0.302
4.7 - 5.8	0.69 ± 0.15	0.03 ± 0.03	0.19 ± 0.08	0.91	3.6	0.279
3.3 - 4.7	6.56 ± 0.59	0.29 ± 0.1	1.19 ± 0.19	8.04	4.3	0.078
2.1 - 3.3	1.17 ± 0.2	0.1 ± 0.06	0.26 ± 0.09	1.53	2.7	0.040
1.1 - 2.1	0.85 ± 0.15	0.07 ± 0.05	0.19 ± 0.07	1.11	1.1	0.012
0.65 - 1.1	0.23 ± 0.09	0.01 ± 0.03	0.08 ± 0.05	0.34	0	0.004
0.43 - 0.65	0.1 ± 0.07	0.02 ± 0.04	0.04 ± 0.05	0.16	0	0
<0.43	0.61 ± 0.14	0.02 ± 0.03	0.52 ± 0.12	1.15	---	---
			Total Respirable Uranium, pCi	40.02	---	---
			Sample Quantity, grams	0.5023	---	---

REFERENCES

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APPENDIX A

MAJOR INSTRUMENTATION

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The display of a specific product is not to be construed as an endorsement of the product or its manufacturer by the authors or their employers.

LABORATORY ANALYTICAL INSTRUMENTATION

Alpha Spectrometry System
Tennelec Model 256
(Oxford, Oak Ridge, TN)
Used in conjunction with:
Surface Barrier and Ion Implanted Detectors
(EG&G ORTEC, Oak Ridge, TN and Canberra, Meriden, CT) and
Multichannel Analyzer
3100 Vax Workstation
(Canberra, Meriden,CT)

Alpha Spectrometry System
Canberra Model 7401VR
(Canberra, Meriden, CT)
Used in conjunction with:
Ion Implanted Detectors and
Multichannel Analyzer
3100 Vax Workstations
(Canberra, Meriden, CT)

APPENDIX B

ANALYTICAL PROCEDURES

APPENDIX B

ANALYTICAL PROCEDURES

ALPHA SPECTROSCOPY

Samples were dissolved by potassium fluoride and pyrosulfate fusion and the elements of interest were participated with barium sulfate. Barium sulfate precipitate was redissolved and the specific elements of interest were individually separated by liquid-liquid extraction and re-precipitated with a cerium fluoride carrier. The precipitate was then counted using surface barrier and ion implanted detectors (ORTEC), alpha spectrometers (Tennelec and Canberra), and a multichannel analyzer (Nuclear Data).