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Ohio Environmental Protection Agency PRS 322 Dayton Unit III Soil Screening Results Report

February, 1998

Executive Summary

PRS 322 (Dayton Unit III) is one of over 400 PRS's (Potential Release Sites) included in the DOE Mmiamiaburg Environmental Management Project (MEMP) Mound cleanup decision-making strategy known as "Mound 2000". PRS 322 is located in the City of Dayton and is owned and occupied by the Dayton Board of Education. The site has a history of polonium-210 processing that occurred during the federal government's Manhattan Project. Because no known analytical soil sampling data had been collected for the site, further information was needed to determine if the site is contaminated. DOE MEMP, USEPA, Ohio EPA and the city of Dayton agreed that Ohio EPA would sample soils at PRS 322 to screen for potential contamination.

Sampling was conducted on August 27, 1997. Four soil samples were collected from PRS 322 and two soil samples were collected from the Grace E. Green School property across Edison Street from PRS 322. Of the six samples collected, two exhibited levels of polonium-210 and lead-210 above expected values. However, further sampling and analysis of soils at PRS 322 is recommended to further define contamination and to evaluate potential health risks at the site.

1.0 Introduction

The U.S. Department of Energy (DOE) MEMP Mound facility contains over 400 sites (called Potential Release Sites or PRS's) where potential releases of hazardous materials may have occurred. In order to accelerate cleanup of the Mound facility and make it available for economic development, a decision-making strategy was developed which addresses the potential for contamination at each of the PRS's. This process is known as "Mound 2000". PRS 322 (known also as Dayton Unit III) is located in the City of Dayton and is not part of the Mound property. DOE Mound consulted with Ohio EPA, Ohio Department of Health and USEPA Region V regarding how to proceed with PRS 322 since it appears in the Mound Operable Unit 9 Site Scoping Report (DOE, 1994). Consistent with the Mound 2000 process, DOE MEMP solicited public input before concluding that PRS 322 required no further assessment. No known analytical soil sampling data had been previously collected for the site. As a result, the public and the City of Dayton expressed their concerns through the public comment process. The state, DOE MEMP and property owners agreed that the Ohio Environmental Protection Agency (Ohio EPA) would obtain samples at PRS 322 to screen for potential contamination,

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For a timeline of the Dayton Project, see Appendix A.

3.0 <u>Sampling Objectives</u>

Ohio EPA's initial objective for sampling at PRS 322 was to screen for residual ²¹⁰Po in the soils due to historical polonium processing at the site, although with a half-life of 138 days, any residual polonium should have decayed to stable lead-206. Cobalt-60 was suspected as having been released at PRS 322, a possible activation product from the irradiation of bismuth. No data had been previously collected for cobalt-60 (DOE, 1994). Therefore, soils were analyzed for cobalt-60 and gamma emitters using gamma spectroscopy. Supplemental analyses included plutonium-238 and total lead, 23.6% which is lead-206, the final decay product of ²¹⁰Po. PRS 322 has no known history of plutonium-238 use, but similar federal sites across the state have shown the presence of this radionuclide.

Due to the discovery of additional information regarding the lead dioxide process used at PRS 322, the sampling objectives were expanded. At the time when the first set of sample results were received, little information was provided regarding the methods used in processing polonium at PRS 322. Repeat analyses were performed on the samples in order to confirm the presence of ²¹⁰Po in the samples. As more information on the processes employed at PRS 322 became available, additional relevant laboratory analyses were performed in an attempt to identify a possible source of the analytes found in soil samples taken at the site. These analyses are presented in detail in Sections 4-8.

4.0 Sampling and Analytical Methods

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4.1 Sample Collection and Field Methods

On August 27, 1997 Ohio EPA conducted surface soil sampling at PRS 322 and nearby areas in accordance with the Mound PRS 322 Field Sampling and Analysis Summary (Appendix B). Sampling was performed to screen for the potential presence of residual ²¹⁰Po and plutonium-238, cobalt-60 and gamma-emitting radionuclides, and total lead. Sampling locations are described in Appendix A and shown in map form in Appendix B. A total of six locations were sampled; four at PRS 322 and two on the Grace A. Green School property across Edison Street and north of PRS 322.

Deviations from the Field Sampling and Analysis Summary include the addition of one sampling location on the west side of Building 4 (see map of PRS 322, Appendix B). Sample location 32206 was added due to discussion among the current occupants that Building 4 had once contained laboratories. Aside from the addition of one sampling location, no deviations from the Field Sampling and Analysis Summary or Standard Operating Procedures occurred.

4.2 Analytical Methods

The samples were analyzed for various radionuclides utilizing both gamma spectroscopy and alpha spectroscopy methods. The samples were initially analyzed specifically for isotopic plutonium (Pu-238 and Pu-239/240) and ²¹⁰Po by alpha spectroscopy. Gamma spectroscopy was used to screen the sample for any unknown gamma emitting radionuclides that may be present in the sample.

The alpha spectroscopy methods are performed by chemically separating the element of concern, i.e. plutonium and/or polonium. The separated element is "plated" onto a planchette which is then counted in a specially designed alpha particle counter, which measures the number of alpha particles emitted as well as the energy of the emitted alpha particle. This information allows the radiochemist to determine which isotopes of the separated element are present in the sample.

The gamma spectroscopy method is performed by placing a prepared portion of the soil sample in a container which is placed under a high purity germanium detector. This detector counts the emitted gamma rays and their specific energies. The resulting spectrum acts as a "fingerprint" for the sample. A computer code is used to separate the spectrum into specific radionuclides present in the sample.

For total lead analysis (non-radiological) the soil sample is digested in concentrated nitric acid and hydrogen peroxide. The sample is then introduced into a graphite furnace equipped with an electrothermal atomizer, and the concentration of lead in the sample is measured by atomic absorption.

4.3 Data Validation

Radiological data was validated by the Ohio Department of Health. Total lead data was validated by Environmental Quality Associates.

5.0 Results

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5.1 Gamma Spectroscopy Results- See table 5.1.

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	32201	32202	32203	.32204	32205	32206
Bi-210	NR	NR	· NR	NR	NR	0.21
Cs-137	0.51	1.1	0.60	0.39	2.2	1.6
K-40	16	15	6,4	12	12	16
Ra-226	1.9	1.4	0.43	0.88	1.2	2.1
Ra-228	1.3	1.5	NR	0.91	1.3	1.5
Ac-228	1.3	1.5	NR	0.91	1.3	1.5
Bi-214	1.9	1.4	NR	0.91	NR	2.1
РЬ-210	NR	NR	NR	NR	41	NR
РЪ-212	1.3	0.91	NR	0.63	0.79	1.3
РЪ-214	2.1	2.2	0.74	1.1	1.7	2.3
Th-228	1.3	1.2	0.63	0.69	NR	1.3
Th-232	1.3	1.5	NR	0.91	1.3	1.5
T1-208	1.2	1.1	0.58	0.64	NR	1.2

Table 5.1 Results are reported in pCi/g.

NR=Not reported.

5.2 Isotopic Plutonium Results - See table 5.2

Tab	le 5.2	Results	ате	reported	in	DCi/2

· ·	32201	32202	32203	32204	32205	32206
Pu-238	< 0.039	< 0.039	< 0.047	< 0.023	< 0.030	< 0.023
Pu-239/240	< 0.030	< 0.034	< 0.034	< 0.023	< 0.038	<0.030

< indicates result was less than the minimum detectable activity (MDA).

5.3 210Po Results - See table 5.3

Table 5.3 Results are reported in pCi/g.

	32201	32202	32203	32204	32205	32206
Po-210	2.6	3.2	14	2.6	39	2.9

5.4 Total Lead Results - See Table 5.4

Table 5.4 Results are reported in mg/kg

	32201	32202	32203	32204	32205	32206
Total Pb	67	117	300	67	64	476

6.0 Discussion of Results

The isotopic plutonium results did not indicate the presence of any significant amounts of these radionuclides in the samples taken. This result was anticipated and consistent with the process history of the site.

The gamma spectroscopy results were also consistent with the known process history of the site with the exception of sample 32205. Sample 32205 had a reported concentration of 41 $pCi/g^{-210}Pb$.

The ²¹⁰Po results, however, were inconsistent with the process history. Samples 32201, 32202, 32204, and 32206 all appear consistent, and would appear to be in the range of expected background concentrations. (Typical background concentrations for ²¹⁰Po are not available at this time, but should be in the same range as ²³⁸U and other isotopes within the radiological decay chain which is 1 - 3 pCi/g). Samples 32203 and 32205, however, were reported higher than anticipated. The reported results were 14 and 39 pCi/g respectively.

Due to the short half-life of ²¹⁰Po (138 days) and the time since operations ceased at PRS 322 (45+ years) virtually all of the raw ²¹⁰Po used at this facility would have decayed to stable lead-206. ²¹⁰Po would be expected to be in equilibrium with its parent radionuclides in the naturally occurring ²¹⁸U decay chain. ²¹⁰Po concentrations should be attributable to natural or background concentrations present in the area.

Sample 32205 appeared to exhibit equilibrium between ²¹⁰Pb and ²¹⁰Po as expected, but

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²²⁶Ra, which is in the same decay chain, was not detected in the gamma spectroscopy results.

Due to these inconsistencies, the ²¹⁰Po planchettes were recounted to ensure that no laboratory counting or computational errors had occurred. The results of the recount were consistent with the initial count verifying the laboratory counting and computational methods. The results of the initial and the recount are shown in table 6.1

Po-210	32201	32202	32203	32204	32205	32206
Initial	2.6	3.2	14	2.6	39	2.9
Recount	2.6	4.7	14	2.6	37	2.7

Table 6.1 Results are reported in pCi/g.

Since the inconsistencies were not resolved through this recount, the laboratory was requested to perform an enhanced analytical suite to aid in determining the source of the elevated ²¹⁰Po concentrations.

In addition to the radiological recounts, a second set of total lead analyses was requested to be performed on a fresh aliquot from the original soil sample. The results of the first and second total lead analyses are shown in Table 6.2.

Table 6.2	2 Results	are reported	i in mø/ke.
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Total Pb	32201	32202	32203	32204	32205	32206
Analysis l	67	117	300	67	64	476
Analysis 2	70	114	308	66	63	412

The second set of total lead results confirm the accuracy of the first. The lead background concentrations for urban/industrial areas in Ohio have been found to have a geometric mean of 161 mg/kg, with a 95% upper confidence limit of 2400 mg/kg (Cox-Colvin, 1995).

7.0 Enhanced Analytical Methods

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The laboratory was tasked to recount the original planchettes prepared for ²¹⁰Po 60 days after the original count. This method should ensure that the radionuclide being counted is truly ²¹⁰Po.

The laboratory was tasked to observe the original gamma spectroscopy results specifically for ²³⁵U. The ²³⁵U data will be helpful in determining the source of the ²¹⁰Po.

The laboratory was also tasked to prepare new samples for analysis from the soils collected previously. The samples will be analyzed for the following radionuclides: ²¹⁰Po, ²¹⁰Pb, and ²²⁰Ra to aid in the determination of the source of the ²¹⁰Po.

²¹⁰Po was analyzed by alpha spectrometry methods to again determine the concentration of ²¹⁰Po present in the soil samples. This was also intented to determine if a microscopic portion of the sample may have had a relatively elevated concentration which was not representative of the whole sample.

³¹⁰Pb was analyzed by beta proportional counting by the in-growth of ³¹⁰Bi. ²¹⁰Pb is the longest lived isotope prior to ²¹⁰Po in the decay series. The ²¹⁰Pb and ²¹⁰Po concentrations should be comparable and demonstrate secular equilibrium.

²³⁶Ra was analyzed by alpha scintillation methods by the in-growth of ²²²Rn. Again, this analysis was performed to determine if secular equilibrium exists in the matrix and, if not, to help determine where disequilibrium occurs and what likely causes may be attributable to this condition.

8.0 Results of Enhanced Analytical Suite

8.1 ²¹⁰Po Results- The results from all four ²¹⁰Po analysis are shown in table 8.1.

Po-210	32201	32202	32203	32204	32205	32206
Initial	2.6	3.2	14	2.6	39	2.9
Recount	2.6	4.7	14	2.6	37	2.7
60-days	2.6	3.3	13	2.5	43	3.1
Split	3.4	3.9	11	3.2	33	3.2

Table 8.1 Results are reported in pCi/g

Note: all results are corrected for decay.

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Table 8.2								
	32201	32202	32203	32204	32205	32206		
U-238	< 3.0	< 3.1	< 1.9	< 2.2	< 5.4	< 3.1		
Ra-226	1.5	1.2	0.42	1.5	1.1	1.7		
РЬ-210	2.2	2,8	8.9	3.5	36	2.0		

8.2 Uranium-238, Radium-226, and Lead-210 Results - See table 8.2

9.0 Discussion of Results from Enhanced Analy

The results indicate definitively that the source for the elevated ²¹⁰Po is ²¹⁰Pb. The data also suggests that the ²¹⁰Pb and ²¹⁰Po are not present in naturally occurring concentrations in samples 32203 and 32205. Table 9.0 indicates what concentrations would be expected from ²³⁸U decay chain as compared to samples 32203 and 32205.

Table 9.0 Results are reported in pCi/L.

	Naturally Occurring*	32203	32205
Uranium-238	1-3	< 1.9	< 5.4 (1.4)
Radium-226	1-3	0.42	1.1
Lead-210	1-3	8.9	36
210Po	1-3	11	33

* levels commonly found in this region, assuming isotopes are in secular equilibrium,

Some natural phenomenon may affect the secular equilibrium of any one sample but the overall concentrations would remain relatively the same. As table 9.0 illustrates, equilibrium is not present in samples 32203 and 32205, indicating that ²¹⁰Pb is the source radionuclide for the ²¹⁰Po.

²¹⁰Pb is still present due to its relatively long half-life of 22 years as compared to ²¹⁰Po (138 days). Concentrations of ²¹⁰Pb may have been as much as four times higher in 1945.

9.1 Conclusion

Prior to using irradiated bismuth as the source for ²¹⁰Po, radioactive lead, ²¹⁰Pb, was used to harvest ²¹⁰Po chemically. Residue from the Port Hope radium refinery, lead dioxide, which

contained 1-3 Ci of ²¹⁰Po per ton was used to harvest ²¹⁰Po. Thirty-seven tons of lead dioxide was received and processed yielding approximately 40 Ci of ²¹⁰Po. The data indicates that the source of the elevated concentrations of ²¹⁰Po is likely due to ²¹⁰Pb contamination.

10.0 Recommendations

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The soil sampling results indicate that the concentrations of ¹¹⁰Pb and ²¹⁰Po are greater than expected. The process history for the site indicates that the presence of these radionuclides may be due to contamination from the former processes at the facility. A more thorough sampling regimen is recommended to evaluate the nature and extent of contamination at this location and identify any potential health risks.

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REFERENCES

- Monsanto Research Corporation, 1979. Historical Resume of Monsanto Operation of the Dayton Project Sites - Units I, II, III, IV, V and others. Waste Disposal 1943-1980. Unpublished Report (December 1979).
- 2. DOE, 1994. Operable Unit 9 Site Scoping Report: Volume 12 Site Summary Report, Final, Appendix A.1 (December 1994).
- 3. U.S. AEC, 1956. Polonium. Harvey V. Moyer, editor. U.S. Atomic Energy Commission (July 1956).
- 4. Lamberger, 1998. Personal interview with Paul Lamberger, former Mound Plant employee (January 1998).
- 5. Cox-Colvin & Associates, 1995. Investigation of Background Metal Concentrations in Ohio Soils (March 1995).

TIMELINE FOR DAYTON PROJECT

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Appendix B

Ohio Environmental Protection Agency Field Sampling and Analysis Summary Mound PRS 322

Site Background

Potential Release Site (PRS) 322 is the former site of Bonebrake Theological Seminary. The property was used by Monsanto, Inc. under the federal government's Manhattan Project for chemical and metallurgical processing of polonium-210. Although the original building was removed, six other primary buildings which were built during the 1940's still remain at the site. Known by the Department of Energy as "Unit III", PRS 322 was turned over to the City of Dayton Board of Education in 1950. The city has occupied the site since that time.

Sampling Objectives

Sampling is being performed by Ohio EPA at PRS 322 in order to screen for residual polonium-210 and cobalt-60, due to historical use. Gamma spectroscopy will be performed as a general check for the potential presence of gamma emitters. Supplemental analyses will include lead, the final decay product of polonium-210, and plutonium-238. Although the site has no history of plutonium-238 use, similar sites across the state have shown evidence of the presence of this radionuclide.

Sampling Locations and Designation

Three surface soil samples will be obtained from the City of Dayton Board of Education property at 1601 West First Street in the following locations:

1 - in the grassy area near the northeast corner of building 4

(see PRS package page 72)

- 2 in the grassy area between Building 6 and Building H, the former location of a guard shack
- 3 as close as possible to the former location of the original seminary building

Two samples will be obtained from the Grace A. Green school property at 503 Edison Street in the following locations:

1 - in the grassy area in front of the building; south side of building

2 - in the athletic field to the east of the building

Samples will be numbered as follows:

322XX

where 322=PRS number

XX-sequential sample number

Coordinates for the sampling locations will be obtained by using a global positioning system (GPS).

Sampling Equipment and Procedures

Collection and handling of samples will be performed in accordance with appropriate Standard Operating Procedures (SOP's) as specified in the DOE Mound Methods Compendium (DOE, 1997) and the Mound Operable Unit 9 Quality Assurance Project Plan (QAPP, DOE, 1993). Samples will be obtained using a stainless steel scoop and bowl. The following SOP's apply to this sampling event:

Methods Compendium:

S-001 General Instruction for Field Personnel

S-002 Soil Sampling with a Spade and Scoop

QAPP:

SOP 1.3 Sample control and documentation

SOP 1.4 Sample Containers and Preservation

SOP 1.5 Guide to Handling, Packaging and Shipping of Samples

SOP 1.6 General Equipment Decontamination

Any deviations from SOP's will be noted.

Sample Handling and Analysis

Samples will be handled in accordance with SOP's 1.3, 1.4 and 1.5. Chain of custody forms will be used to record required custody, and laboratory information. The following laboratory analyses will be performed:

polonium-210 isotopic plutonium gamma spectroscopy (includes cobalt-60) lead

Radiological anlayses will be performed by Thermo NuTech and lead analysis will be performed by Ross Analytical Laboratories.

Data Validation

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Radiological data will be submitted for validation to the Ohio Department of Health Bureau of Radiological Protection. Non-radiological data will be reviewed for validity by Environmental Quality Associates.

References

DOE, 1997. Methods Compendium, Mound Plant. U.S. Department of Energy Ohio Field Office (June 199)7.

DOE, 1993. Operable Unit 9 Quality Assurance Project Plan. U.S. Department of Energy Albuquerque Field Office (June 1993).

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Appendix C

(location of Grace A. Green school)

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PRS 322 - Dayton Unit III Sampling Locations

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