LESSONS LEARNED FROM THE IMPLEMENTATION OF A MODERN PARTICULATE RADIONUCLIDE AIR MONITORING PROGRAM IN SUPPORT OF OUTDOOR ENVIRONMENTAL TENORM REMEDIATION

M. S. Winters
FUSRAP Maywood Team - Shaw Environmental, Inc.
100 West Hunter Ave., Maywood, NJ 07607

D. C. Hays Jr.
FUSRAP Maywood Team - United States Army Corps of Engineers
700 Federal Building, 12th St., Kansas City, MO 64106

ABSTRACT

This paper discusses recent improvements to the environmental particulate radionuclide air-monitoring program at the FUSRAP Maywood Superfund Site (FMSS), which can be applied to other radiation protection programs conducting similar work or working in similar environments.

This paper will present the regulatory back-drop and the mechanisms and assumptions by which the Maywood Team evaluates the relationships between parent nuclides and daughter progeny within the Thorium and Uranium Decay Series. Discussions into the historical perimeter air monitoring and analysis methods previously used at the FMSS and commonly used at other environmental cleanup sites follow. The focus will then shift to lessons learned from the Maywood Team’s efforts to continuously improve the quality of the air-monitoring program over recent years.

Readers with similar programmatic responsibilities will benefit from exposure to methodologies that are manageable and technically effective in this type of environment.

INTRODUCTION

Shaw Environmental, Inc. is teamed with the United States Army Corps of Engineers (USACE) at the Maywood Site. The mission of the FUSRAP Maywood Team is to safely and effectively remediate a waste stream consisting primarily of soils and debris contaminated with the waste product of an industrial thorium extraction process. Although Maywood wastes are not legally classified as Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM), the chemically manipulated radioactive waste stream is primarily thorium-232 (Th-232) with lesser amounts of radium-226 (Ra-226); uranium-238 (U-238); and other associated naturally occurring radionuclides. The current phase of the remediation effort includes the remediation of the FMSS, which includes the 4.7 hectare Maywood Interim Storage Site (MISS) and 26 vicinity properties (commercial, state and federal) located in the densely populated and commercially active area of Bergen County, New Jersey. In this environment, accurate and dependable air monitoring technologies are essential to ensure that remedial actions are protective of public health and the environment.

Cleanup of radiologically contaminated soils in a densely populated area can be challenging. For the Health Physics Team one of the biggest challenges is to ensure that fugitive emissions, generated during excavation and handling of contaminated soils, are limited to protect workers, nearby residents, and the general public. An effective environmental and occupational air-monitoring program is essential to verify that fugitive emission controls are adequately protective. When working with TENORM, it is critical to evaluate the potential for disruption of the equilibrium conditions within the waste stream.

REGULATORY BACK-DROP

Airborne Public Exposure

Limiting particulate radionuclide emissions to the public from remediation activities is the key challenge for the Health Physics Program. The National Emission Standard for Hazardous Air Pollutants (NESHAP) 40 CFR 61.102 [1] and 10 CFR 20.1101(d) [2] limit off-site exposures at 0.1 milliSieverts/year (mSv/y). Specific isotopic concentration limits can be determined by taking twenty percent of the values specified in 10 CFR 20, Appendix B, Table 2, Column 1, which are based on the NRC limit of 0.5 mSv/y.

Formal regulatory compliance at Maywood is demonstrated by the use of the Clean Air assessment Package (CAP-88) computer code. The Maywood Team uses field air sampling to track and trend site perimeter air conditions.

Occupational Exposure

The USACE Engineering Manual (EM 385-1-80) Safety-Radiation Protection Manual [3] established a "never-to-exceed" limit, equivalent to the standard in 10 CFR 20.1201(a), of 50 mSv/year and a 10 percent operating limit, applicable to Maywood, of 5 mSv/year.

The Maywood Team uses field air sampling to assess occupational exposures from airborne contaminated particulates.

IDENTIFYING THE ISOTOPIC DISTRIBUTION IN THE WORKPLACE

One of the great challenges of any particulate radionuclide air monitoring program comes from trying to understand the relationship between the activity deposited on a given filter, the related gross counting activity, and the precise measurement of the isotopic content in the sampled filter. Understanding these relationships is an ongoing process at Maywood.

Currently all Maywood air samples are collected on general-purpose glass fiber filters with an acrylic resin binder. The acrylic resin binder is added to the filter during fabrication to maintain integrity in high-pressure drop applications. A significant drawback to these type filters is caused by the resin binder interfering with the lab analysis digestion process. In preparation for isotopic analysis, the filters are digested using a combination of strong acids. The acrylic binder does not readily dissolve and retains an indeterminate percentage of the radionuclides captured on the filter. The radionuclide retention characteristics of the acrylic resin binder were identified by gross beta counting. Some empirical relationships are possible to establish, but they are limited in usefulness. For example, a comparison of gross beta counting of the filter vs. gross beta counting of the collected resin binder would produce a rough estimate of the radionuclide concentrations retained within the binder. It is a rough estimate for two main reasons. The first is that little is currently understood about the elemental preference to the resin binder that may be occurring during digestion. The second reason is that the comparison of gross counting measurements of two distinctly different matrices is unsound, unless the differences in self-absorption characteristics are empirically established. The Maywood Team is currently evaluating whether an "AE" grade glass fiber filter, with no resin binder, can be used for extended operating cycles without breakthrough.

To date, the Maywood Team has relied primarily on soils isotopic data to establish appropriate relationships for the various Uranium and Thorium Series products present at the site. The weighted relationship is built from waste screening data for the primary contaminants of concern (CoC) Th-232, Ra-226, and U-238, and is updated annually. Table I presents the waste screening data, based on the ten soil load-out evolutions performed at the MISS in 2003 and the calculated averages for each CoC:

Table I. Weighted Averages for Maywood Contaminants of Concern Based on Waste Disposal Soil Screening Concentrations

Load-Out	Tons	Weighting Ratio	Th-232 (pCi/g)		Ra-226 (pCi/g)		U-238 (pCi/g)	
			Sampled	Weighted	Sampled	Weighted	Sampled	Weighted
1	5648	20.33%	2.67	0.54	0.89	0.18	3.53	0.72
2	2459	8.85%	2.11	0.19	0.65	0.06	2.04	0.18
3	1390	5.00%	2.81	0.14	0.7	0.04	1.74	0.09
4	1496	5.39%	6.86	0.37	1.04	0.06	3.67	0.20
5	1604	5.77%	3.83	0.22	1.03	0.06	2.13	0.12
6	4167	15.00%	7.47	1.12	1.38	0.21	2.72	0.41
7	2223	8.00%	12.28	0.98	1.87	0.15	8.94	0.72
8	3874	13.95%	14.12	1.97	2.09	0.29	10.03	1.40
9	3034	10.92%	7.66	0.84	1.3	0.14	6.34	0.69
10	1883	6.78%	9.96	0.68	1.64	0.11	7.85	0.53
SUM	27778	1	69.77		12.59		48.99	
AVG				**7.05**		**1.29**		**5.05**

[a] Soil Mass & Concentration Data Source: Ref. 4

After a relationship of the primary contaminants is established, an Effective Derived Effluent Limit (DEL_{eff}) can be determined using the following formula:

$$DEL_{eff} = f_a \left[\sum_i \left(f_i / DEL_i \right) \right]^{-1} \qquad \text{(Eq. 1)}$$

Where:

DEL_{eff} – Derived Effluent Limit @ 0.1 mSv/y

f_a – The ratio of total alpha activity in the mixture to the total activity of the mixture

f_i – The ratio of each radionuclide to the total activity of the mixture

DEL_i – The DEL for each nuclide in the mixture from 10 CFR 20, Appendix B, Table 2 "Effluent Concentrations", Column 1, "Air", reduced by 80% to achieve 0.1 mSv/y.

The underlying principles are an assumption of equilibrium between the reported CoCs and their daughter progeny, and an assumption that all radionuclides are Class Y, except radium. Each isotope is weighted against all radionuclides likely to be present in the mixture during analysis. Note that radon isotopes and associated progeny are not included, as several iterations of calculations were performed with varying assumptions of radon daughter activities resulting in limited impact to results. Given this and assuming that radon isotopes and associated progeny are unlikely to be present on the filter after a reasonable decay period, they are excluded from the evaluation. Finally, the alpha emitting isotopes are isolated to establish a measurable limit for use in comparison to gross alpha air sample counting. The following Microsoft Excel™ worksheet output, presented as Table II, shows the DEL_{eff} calculation and the result for 2003 data inputs:

Table II. Effective DEL Calculation Methodology Worksheet Output

Isotope	DEL (uCi/ml)	Activity (pCi/g)	Activity Fraction (Fi)	Emission	Fi/DEL
Th-232	1.20E-15	7.05	0.1115	Alpha	9.29E+13
Ra-228	4.00E-13	7.05	0.1115	Beta	2.79E+11
Ac-228	1.20E-11	7.05	0.1115	Beta	9.29E+09
Th-228	4.00E-15	7.05	0.1115	Alpha	2.79E+13
Ra-224	4.00E-13	7.05	0.1115	Alpha	2.79E+11
U-238	1.20E-14	5.05	0.0800	Alpha	6.67E+12
Th-234	4.00E-11	5.05	0.0800	Beta	2.00E+09
Pa-234	1.80E-09	5.05	0.0800	Beta	4.44E+07
U-234	1.00E-14	5.05	0.0800	Alpha	8.00E+12
Th-230	6.00E-15	5.05	0.0800	Alpha	1.33E+13
Ra-226	1.80E-13	1.29	0.0204	Alpha	1.13E+11
U-235	1.20E-14	0.23	0.0037	Alpha	3.07E+11
Th-231	1.80E-09	0.23	0.0037	Beta	2.04E+06
Pa-231	1.60E-15	0.23	0.0037	Alpha	2.30E+12
Ac-227	1.20E-15	0.23	0.0037	Beta	3.07E+12
Th-227	1.00E-13	0.23	0.0037	Alpha	3.68E+10
Ra-223	1.80E-13	0.23	0.0037	Alpha	2.04E+10
	Total	63.18	1	17	1.55E+14
				Total alpha	
				11	
				Fraction alpha (Fa)	**Fi/DEL sum ^-1**
				0.647058824	6.44261E-15
			Effective DEL for Mixture (uCi/ml)	**4.17E-15**	

[b] Effective DEL Methodology Source: Ref. 6

HISTORICAL PERIMETER AIR MONITORING PRACTICES AT MAYWOOD

From inception through 2002, the Maywood Project utilized standard rotary vane air samplers, commonly referred to as a "Lo-Vol", to collect particulate emission data. Rotary vane style samplers are commonly encountered in the nuclear industry with varying designs and features, but a similar basic function. They are relatively reliable and usually can be handled by a single technician. There are more costly rotary vane style samplers available that have robust electronics and pump control packages. However, this discussion relates to the standard air sampler, more commonly encountered in the industry, where operators make manual flow adjustments and view flow rates from a float indicator. Typical costs for a standard manual control low-volume air sampler are in the range of $700-$1000. Annual maintenance costs at Maywood average $150/sampler.

There are several distinct functional characteristics and lessons learned from deployment of the basic rotary-vane style samplers at Maywood:

- A weather house is needed to protect the sampler from damage in outdoor applications. At a minimum, a stand is required to keep the sampler off of the ground.
- Due to their relatively small size, they are easily stolen. Air sampler thefts have occurred at Maywood.
- Samplers are calibrated under weather conditions (e.g., temperature and barometric pressure) that often differ from the changing conditions encountered during the monitoring period.
- Samplers lack the capability to manage pump speed and maintain preset flow rates under pressure-drop conditions expected as particulate filter loading occurs and weather conditions change. The lack of certainty in day-to-day flow rates can lead to less than accurate sample volume calculations and require more frequent technician observations.
- Power interruptions and mechanical failures can go unnoticed in the more basic models. The lapses in sampling can lead to volume calculations that are much higher than actual and result in the underreporting of air concentrations. In the worst case, where a sampler failed at an unknown point, the collected sample is virtually useless.
- Rotary vane style samplers typically require preventative maintenance on at least an annual basis
- Under continuous operating conditions, the sampler flow rate is limited to around 0.06 cubic meters per minute or 60 liters per minute (lpm). Average flow rates at Maywood are 56 lpm.
- Samples require extended count times in order to approach the necessary minimum detectable activities (MDA).
- Average Noise Level @ 1 meter - 66.5 decibels - "A" weighted (dBA)
- Operating current range is from 4-7 Amps.

Lo-Vols at Maywood utilize a 47mm glass fiber filter for the collection of airborne particulates. Samples are typically counted for gross alpha activity in a Protean™ Model WPC-9550 Gas Flow Proportional Counter with an alpha counting efficiency that ranges between 20% and 29% and a background range between 0 and 0.13 counts per minute (cpm). Counting efficiencies are generated from an electroplated stainless steel source and require a correction for alpha particle self-absorption and shielding that is present in the actual samples.

It should be noted that for routine monitoring of general area airborne radiological conditions, in temporary work areas, and under occupational dose limits, the standard rotary vane type pump is ideal. The main challenge arises when the dose limits drop to public levels. The challenge is sampling a volume that is sufficient to meet required sample MDAs and have acceptable uncertainty without requiring extensive laboratory resources. It can become almost impossible to accurately trend airborne radiological conditions at public dose levels routinely because of the required MDA and large individual sample uncertainties.

MODERNIZED PERIMETER AIR MONITORING PRACTICES AT MAYWOOD

In 2002, the Maywood Team procured high volume air samplers to replace the rotary vane samplers in use at perimeter monitoring stations. The specific model selected was the Hi-Q™ Model HVP-3800AFC (~$3,600). Lessons learned from the deployment of the HVP-3800AFC at Maywood identified several key characteristics worth noting:

- Weather-proof
- Small footprint
- Easily secured to prevent theft and minimize vandalism
- Brushless 3-stage maintenance-free centrifugal blower motor
- Self-contained electronics package that can be easily replaced by a technician with little training
- Maintains critical sampling information (i.e., run times and volume sampled) in static memory in the event of a power interruption or mechanical pump failure
- Can be simply modified to sample for total airborne particulate or for particulate matter averaging 10 microns or less (PM-10).
- Uses a mass flow sensor to continuously adjust motor speeds to maintain sustainable flow rates under high-pressure drop conditions and in variable weather conditions.
- Typical operating flow rate of 13 CMM
- Average Noise Level @ 1 meter - 68 dBA
- Operating current range is from 12-14 Amps.

HVPs at Maywood utilize an eight by ten inch glass fiber filter for the collection of airborne particulates. The same filter array is used in either the Total Airborne Particulate (TAP) or PM-10 application. Samples are counted for gross alpha activity in a system comprised of a Ludlum™ Model 2000 Scaler and NE™ Model AP-6 600cm^2 Scintillation Detector. The AP-6 is a floor-wall hand held detector adapted for use in this fixed geometry application. Typical instrument alpha counting efficiencies are between 14 percent and 17 percent with background count rates between six and eight cpm.

Using the AP-6 detector is advantageous because it allows for the entire filter to be counted without dissection. To establish more realistic counting efficiencies for air filters, a source standard was created that is the same dimension as a sample requiring counting. Liquid Th-230 standard was sprayed on a thin film surface, and then adhered to a standard air sample filter. The benefits of using a source standard in this configuration are three-fold. By using a check source that is the same size as the samples being counted, areas of the detector with poorer efficiency are properly accounted. Second, no assumption about the uniform distribution of contaminants on the filter is required since dissection is not required. Finally, using a source that more closely emulates an actual sample reduces the need to adjust for self-absorption and shielding.

UNDERSTANDING THE DATA QUALITY BENEFITS OF SAMPLING SYSTEM MODERNIZATION

In order for an air-monitoring program to be effective, the sampling data must be of sufficient quality to be useful. Considering the regulatory back-drop discussed earlier, generating quality data can be challenging. A useful measure of the quality of data generated is the "Minimum Detectable Concentration" or MDC. MDC is defined as the minimum activity concentration on a surface or within a material volume that an instrument is expected to detect with a 95 percent confidence [5]. Calculated sample-specific MDC values ideally should be as small a percentage of the action level as is reasonably achievable. The target goal for MDC values at Maywood is 10 percent of the applicable limit. For 2003, the calculated DEL_{eff} for Maywood was 4.7E-15 uCi/ml. The MDC target goal is 10 percent of the DEL_{eff} or 4.7E-16 uCi/ml.

There are several different ways to calculate MDC. The formula selected for application at Maywood allows for variances between the sample and background count times and is presented as follows:

$$MDC = \frac{3 + 3.29\sqrt{(R_b)(T_s)\left(1 + \frac{T_s}{T_b}\right)}}{\left(E_i\right)(T_s)(SAF)(vol)(2.22E6)} \qquad \text{(Eq. 2)}$$

Where:

MDC – minimum detectable concentration in microcuries per milliliter (uCi/ml)

Rb – background count rate in cpm

Ts – sample count time in minutes (min)

Ei – counting instrument efficiency expressed as a decimal

SAF – Self-Absorption Factor to account for alpha self-absorption and shielding

2.22E6 – value to convert disintegrations per minute (dpm) to uCi

vol – sample volume in ml

An alpha self-absorption factor of 0.7 is introduced in situations where instrument calibration occurs via a stainless steel electroplated source. The self-absorption factor accounts for the deeper particulate embedding and self-shielding that occurs when glass-fiber filters are operated for extended periods or in high dust environments. The self-absorption factor may be adjusted as isotopic analysis results are compared to gross counting results.

As discussed previously, the target sample-specific MDC for Maywood Site perimeter samples is 10 percent of the most restrictive annual limit, or 4.7E-16 uCi/ml. Table III compares the sampling and counting conditions (typical) for the historical and modern perimeter air monitoring methods used at Maywood, and their resultant impact on MDC values:

Table III. Comparison of Calculated MDC Values based on Typical Operating Conditions

	LoVol & 47mm Proportional Counting	**HVP & 600cm^2 Scintillation Counting**
Sample Volume (ml)	4.2 E 8	4.0 E 10
Counting Instrument Efficiency (Ei)	25%	15%
Alpha Self Absorption Factor (SAF)	0.7	N/A
Background Count Rate (Rb)	0.1 cpm	7 cpm
Sample Count Time (Ts)	120 min	60 min
Background Count Time (Tb)	120 min	600 min
Minimum Detectable Concentration (MDC)	9.8 E -16 uCi/ml	9.2 E -17 uCi/ml

As expressed, the MDC calculated for the typical HVP sample is much lower than the typical LoVol sample and is less than the target MDC of 4.7E-16 uCi/ml. This is true, even though the proportional counter registers a significantly lower background count rate (Rb). This is because the single greatest factor affecting this comparison is the sample volume. The HVP’s ability to draw air at a much higher and constant rate, for a significantly longer time period, is a significant advantage when evaluating air concentrations at such restrictive levels.

CONCLUSION

Evaluating the distribution of radionuclides likely to be present in airborne particulates and implementation of a modern air monitoring program are two steps that the Maywood Team has taken to ensure radiation exposures to the public and workers from site activities are maintained As-Low-As-Reasonably-Achievable (ALARA) and within prescribed limits. These techniques can help sites in similarly constrained situations assess radiological constituents in a more realistic manner and produce sampling data that is of sufficient quality to be useful in evaluating exposure trends.

REFERENCES

[1] 40 CFR 61, National Emission Standards for Hazardous Air Pollutants Washington, D.C.: Environmental Protection Agency. Current Revision July, 2004.
[2] 10 CFR 20. Standards for Protection against Radiation, Washington, D.C.: Nuclear Regulatory Commission. Current Revision January, 2004.
[3] EM-385-1-80, Safety-Radiation Protection Manual, Washington, D.C.: United States Army Corp of Engineers. Current Revision May, 1997.
[4] Annual NESHAP Compliance Report - Year 2003; Calculation 11, "MISS 2003 Annual NESHAPS Calculation." Prepared by Shaw Environmental, Inc. for the Army Corp of Engineers. June 2004.
[5] NUREG-1507. Minimum Detectable Concentrations with Typical Radiation Survey Instruments for Various Contaminants and Field Conditions, Washington, D.C.: Nuclear Regulatory Commission. December, 1997.
[6] Keele, B., Reider, B., *Technical Basis for the Dosimetry and Air Sampling Programs*, Shaw Environmental & Infrastructure, USACE Maywood FUSRAP Site, 2002

FOOTNOTES

[a] See Ref. [4].

[b] See Ref. [6].