Alternative Sample Preparation of Soils Containing Thorium at an Environmental Protection Agency Superfund Site - 9380

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ABSTRACT

This paper describes an alternative method to determine radon levels in soil contaminated with thorium. Traditional measurements of radon and the 21-day in-growth waiting period are not time and cost effective for remediation and restoration efforts of residential and industrial properties. A review of actual sample results and calculated theoretical radon daughter yields was performed and is reviewed as a continual process to substantiate alternative method goals. A former gas mantle manufacturing facility used thorium to create the "glow" for gas lamps from about 1912 to 1944. Upon termination of the mantle production, thorium wastes were scattered over a small metropolitan area that later became a Superfund site. The material pre-dates the Atomic Energy Commission and the Nuclear Regulatory Commission, is of low activity and is generally disposed of as ore processing materials generated prior to 1978. Remediation of the site involves excavating soil/debris on impacted residential and industrial properties. An alternative approach has been developed that significantly reduces sample preparation time for soil samples and provides comparable results to the standard method. Some radon is released during the collection and sample preparation process that is to be accounted for. Radon in-growth analysis was reviewed for the wet and dry corrected samples. A correction factor is applied to account for radon in-growth depending on the sample activity. Radon correction factors at low activity show good correlation, but show variability at higher activity. Actual radium/thorium counts were compared to derived radium/thorium activity and a good correlation exists.

INTRODUCTION

The Welsbach gas mantle manufacturing facility in Gloucester City, New Jersey used raw monazite sand to concentrate thorium. The General Gas Mantle facility in Camden, New Jersey used refined thorium to produce gas mantles. The gangue material from both manufacturing facilities and later demolition debris were disposed of at various industrial and residential locations near Camden and Gloucester City, New Jersey (Figure 1).

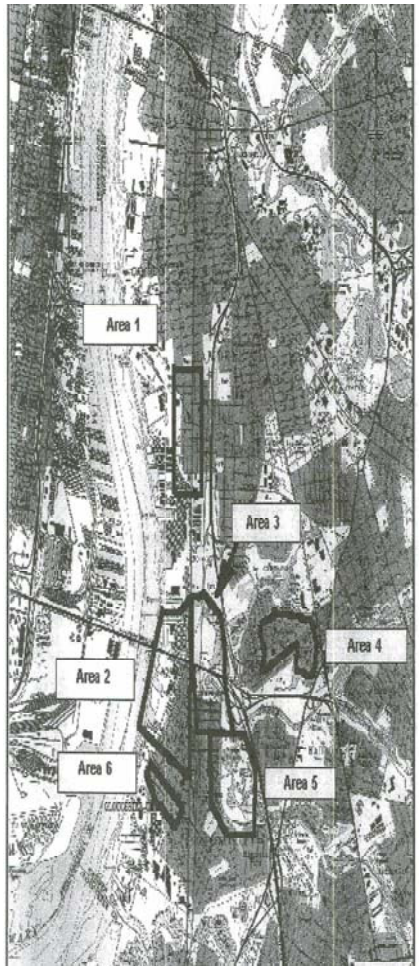

Figure 1 – Welsbach General Gas Mantle Contamination Site –
Study Areas. [1] Correlation Report No.7, Sevenson, 2008

Different ratios of Th-230 and Th-232 are present in the waste material depending on the waste components. The waste material is in equilibrium conditions, but sampling and remediation activities liberate Rn-220 and Rn-222.

The initial procedure used to evaluate soil samples for thorium and related daughter products by gamma spectroscopy uses standard drying and grinding. The drying of soil samples by oven or ambient air takes many hours to days of drying time. Dried samples are then sieved and ground to improve the homogenization. The alternative method utilizes blending of soil by hand followed by a moisture analysis measurement to determine the percent moisture in each sample, which takes only 15 to 45 minutes of sample preparation. The actual weight of the sample is moisture corrected in order to report results in equivalent dry weight. Thorium contaminated soils were evaluated using this method and correlation analysis showed a strong linear fit. A correction factor could
not be used for the moisture analyzed samples due to high variability of moisture content, therefore the individual moisture measured sample provides a more accurate estimate of activity.
Use of the alternative moisture correction method provides a tool to faster evaluate site remediation and is used for backfill decisions. Standard methods of sample preparation and analysis are used for Final

Status Surveys. Applying radon correction factors for low activity samples can be used to avoid waiting for the 21-day decay period. This radon calculation will be investigated in this paper.

Additionally, thorium activities are calculated using a correlation factor for quick pre final status survey decisions but for Final Status Survey results, the actual Th-230 alpha spectroscopy results and Th-232 gamma spectroscopy results are reported.

METHODS

The radionuclides of concern at the Superfund project are Ra-226, Ra-228, Th-230, and Th-232. For residential excavated soils less than or equal to 2.44 meters (8 feet) below ground surface (bgs), goals are ≤ 0.11 Becquerels (Bq) (3 pCi/g)for Ra-226 plus Ra-228 above background. For Th-230 plus Th-232 above background the goal is also ≤0.11 Bq (3 pCi/g). For residential ≥2.44 meters bgs, and commercial property at all depths, the remediation goal is ≤0.18 Bq(5 pCi/g) for combined radium plus background and combined thorium plus background. Background radium and thorium levels are shown in Table 1.

Radionuclide	Mean Conc. (pCi/g)	Std. Deviation (pCi/g)
Th-230	1.095	0.705
Th-232	0.872	0.515
Ra-226	0.877	0.490
Ra-228	1.048	0.685
Th-230 + Th-232	1.97	
Ra-226 + Ra-228	1.93	

Table I: Background Isotope Data

[2] Sevenson 2008

Manufacturing processes were terminated in 1944. Due to the elapsed time, all Th-232 and Ra-226 progeny will be present at the same concentration as that of the parent, in secular equilibrium. Measurements of the progeny are analyzed by gamma spectroscopy with their weighted average to calculate the amount of thorium and radium alpha emitters. Ra-226 is quantified from the B-214 and Pb-214 daughters; Ra-228 and Th-232 are quantified by the surrogate Ac-228. Thorium-230 is analyzed by alpha spectroscopy by an off-site laboratory.

Counting and collecting radon can be a difficult process. Radon generation and transport measurements can be at different rates and theoretical inconsistencies based on air-phase emanation can result in activities not representative of true radon conditions [3] Rogers, 91. The solid, liquid and gas phases in the environment can affect radon emanation. Methods for radon sampling are indirectly measured from the daughter isotopes at equilibrium conditions. Ra-220 is measured indirectly by the Pb-212 Lead and the Ra-222 by the Pb-214 [4] Yamashita, 87.

Table 2 illustrates a comparison of dry sample activity without a 21 day in-growth, and actual 21-day in-growth sealed in a Marinelli beaker and then counted. At low sample activity, of one to a few pCi/g, the radon disequilibrium impact is very small, but cannot be entirely disregarded.

Table II: 21-Day vs. Dry Ratio

	Dry	Dry	21 Day	21 Day	21 Day vs. Dry Actual	
Sample ID	Ra-226 pCi/g	Th-232 pCi/g	Ra-226 pCi/g	Th-232 pCi/g	Radium Ratio	Thorium Ratio
2	0.78	1.12	0.82	1.13	1.05	1.01
1	0.90	1.01	0.85	1.03	0.94	1.02
A5-2	1.58	0.92	0.92	1.08	0.58	1.17
3	0.92	1.06	0.76	1.13	0.83	1.07
6A-2	1.50	1.93	1.62	1.66	1.08	0.86
A1.3-U01-001	0.54	1.84	0.65	2.52	1.2	1.37
A5-1	0.73	0.83	0.77	0.98	1.05	1.18
SWC-1	0.98	1.10	0.81	1.04	0.83	0.95
1780-004	0.54	0.92	0.52	0.85	0.96	0.92
7A-1	1.53	1.76	1.75	1.89	1.14	1.07
7A-2	1.80	2.33	1.77	2.12	0.98	0.91
NS-1	0.88	1.11	0.84	1.06	0.95	0.95
F27#4	2.36	1.94	2.66	1.87	1.13	0.96
1780-001	1.05	2.32	1.07	2.38	1.02	1.03
1780-003	0.75	1.96	0.88	1.91	1.17	0.97
6A-1	0.52	2.90	0.56	2.75	1.08	0.95
B23#2	0.62	0.65	0.51	0.68	0.82	1.05
H/127	1.29	1.78	1.32	1.87	1.02	1.05
RCS-079	1.69	2.76	1.99	3.24	1.18	1.17
B23#1	0.47	0.44	0.44	0.45	0.94	1.02
F27	3.89	3.02	4.55	3.26	1.17	1.08
D24/25	1.15	3.03	1.19	3.49	1.03	1.15
6/7-F/G	1.55	4.15	1.58	4.44	1.02	1.07
5/6G	2.12	3.16	2.26	3.47	1.07	1.10
F4	1.83	3.30	2.14	3.04	1.17	0.92
G5	0.47	0.59	0.52	0.69	1.11	1.17
H5A	0.28	0.43	0.31	0.36	1.11	0.84
H5B	0.43	0.75	0.48	0.64	1.12	0.85
A5.1-U03-004	0.75	0.83	0.74	0.86	0.99	1.04
A5.1-U03-008	1.85	2.68	1.89	2.88	1.02	1.07
A5.1-U03-011	0.66	0.97	0.65	0.99	0.98	1.02
A5.1-U03-012	1.07	1.61	1.03	1.64	0.96	1.02
A5.1-U03-102	1.48	2.28	1.45	2.31	0.98	1.01
A5.1-U03-103	0.87	0.98	0.80	1.01	0.92	1.03
A5.1-U03-104	0.99	1.11	0.92	1.09	0.93	0.98

A1.6-U01-001	0.45	0.59	0.48	0.60	1.07	1.02
A1.6-U01-002	0.50	0.51	0.46	0.49	0.92	0.96
TSF-132	3.05	77.0	3.63	76.5	1.19	0.96
TSF-133	2.91	85.9	4.05	95.7	1.39	1.11
TP2-2	1.65	29.0	1.70	28.7	1.03	0.99
TP6-4	2.64	3.64	2.62	3.45	0.99	0.95
TP7-5	2.59	3.44	2.70	3.22	1.04	0.94
TP7-6	2.54	3.34	2.57	3.32	1.01	0.99
TP9-6	2.66	3.42	2.71	3.50	1.02	1.02
TP8-6	1.73	2.27	1.78	2.17	1.03	0.96
INFO-009	0.65	0.67	0.67	0.65	1.03	0.97
INFO-010	0.77	1.57	0.82	1.55	1.06	0.99
INFO-012	2.58	19.9	3.48	19.8	1.35	0.99
INFO-021	0.94	2.63	1.00	2.64	1.06	1.00

[5] Sevenson, 2008

Radon is only a concern for Final Status Survey samples. Soils are analyzed wet using a Denver Model 2000 moisture analyzer to determine activity for backfill decisions without waiting on the complete dry time. Final Status Survey samples are dried using standard approved methods, counted and then sealed in a Marinelli beaker for 21 days and re-analyzed. The New Jersey Department of Environmental Protection requires radon to be accounted for in Final Status Survey samples. The average ratio for Ra-226 is 1.04 with
a standard deviation of 0.13 and a median of 1.05. For Th-232, the average ratio is 1.03
with a standard deviation of 0.11 and a median of 1.02 [6] Sevenson, 08. Therefore, a ratio of 1.05 is conservative and will be used to calculate the radon contribution for Final Status Survey samples as a proposal for future approval.

A summary of radium decay is provided by [7] Evans, 69 to explain equilibrium conditions and to eliminate direct radon sampling. The growth of radioactive series activities is complimentary to their decay. The summation rules, for example deal with 1 curie of purified radium in a glass sealed ampoule will accumulate daughter product radon, according to exponential growth ($1-e^{-\lambda t}$), and that radon's daughter product, RaA, and all the subsequent series of decay products in-growth will each have an activity of 1 curie [8] Evans, 91. All radionuclide decay products are in equilibrium with the parent 1 curie of radium. Open the ampoule, radon escapes and you no longer have equilibrium. If the lost radon could be captured, it would be found to grow daughter products at the same rate at a predicted growth curve. Activity is based on the activity of the collected parent sample.

DISCUSSION

The concern for radon at this site from any indoor residual thorium left in a residential home is from any potential inhaled dose. Overall levels of thorium contamination prior to remediation do not present high exposure scenarios. After remediation, once the 3 pCi/g combined thorium plus background goal has been obtained, exposure scenarios are at ALARA and the potential for radon generation will be minimized.

The evaluation of Table 2 data show that calculated radon activity using the ratio of 1.05 is conservative and could be used rather than performing a 21-day in-growth count. Higher activity samples do not follow the ratio, due to the variability in source term thorium. The high radium activity samples will be screened out by walk over gamma surveys and gamma counting of samples at that location, to require further remediation and removal of contaminated soil. Low activity samples follow ratio trends and radon levels can be predicted within a range based on dry activity of gamma spectroscopy analysis.

A statistical comparison of data was performed. The null hypothesis was accepted that there is no difference between moisture corrected or actual dry radon data for both radium and thorium than their respective 21-day equilibrated in-growth. Continued data evaluation may allow the 21-day in-growth procedure to be eliminated. Radon disequilibrium correction combined with sample specific moisture correction significantly reduces time and costs, this costs savings may be realized by other projects if similar methods were adopted.

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