Naturally Elevated Radionuclides Encountered at Environmental Radiation Remediation Sites – 15224

Jough Donakowski, USACE Kansas City District

ABSTRACT

Many FUSRAP sites contain contamination that is similar to naturally occurring radioactive material (NORM) which may lead to improper characterization of elevated material, resulting in additional effort and cost. Several instances of NORM or other non-FUSRAP related material present at cleanup locations are discussed herein.

INTRODUCTION

In recent years USACE has encountered many types of natural material at sites and developed tools to explain the occurrences, so as to not chase natural material not related to site contaminants. Rare earth analysis can assist in differentiating between Site related contamination and Naturally Occurring Radioactive Material (NORM). Some substances like coal ash and other common fill materials often contain elevated uranium and thorium at or above FUSRAP project cleanup criteria levels.

Elevated natural Uranium can be found in wetlands and in other swamp material at concentrations in the tens or even hundreds of picocuries per gram and out of equilibrium with thorium daughter products.

Elevated Pb-210 in soils is often encountered, due to natural processes involving radon-222 daughter washout from rain events and accumulation, and is not uncommon. Slightly elevated levels of Pb-210 are commonly found in low lying areas where rain collects and concentrates. Pb-210 levels ranging up to 20 pCi/g or even higher can be found when analyzing soil and sediment samples collected from these areas.

All of these types of excess natural radiological material can be accounted for using tools such as rare earth element analysis, close examination of radiological concentration ratio, and process knowledge.

RARE EARTH ELEMENT ANALYSIS

One particular challenge when dealing with large remediation sites contaminated with radiological material is differentiating between low-level Site related contamination and naturally occurring radioactive material (NORM) or technologically enhanced naturally occurring radioactive material (TENORM). Sites like thorium processing and milling facilities generate wastes that contain contaminants which are similar to naturally occurring radioactive material deposits and other industry wastes. This issue is further compounded when addressing radiological contamination present at locations which have been subject to past filling with fly ash, coal ash, or other higher activity material. These materials may have activity levels near, or even well above, a

project's cleanup goals. At the Welsbach/General Gas Mantle Superfund Site (Site), many of the suspected fill areas are comingled with fly ash or coal ash.

To assist in determining whether or not historical fill materials are present at known or suspected Site fill locations, the rare earth element ratios of un-impacted fill areas were compared to rare earth element ratios of the fill at the former Welsbach facility. Based on this study of rare earth element ratios, particularly lanthanum, cerium, and neodymium, a higher background activity for these fill areas was developed. Though the ROD defined cleanup level remained unchanged, the higher background derived from the fly ash/coal ash material, as identified by rare earth analysis, resulted in higher project screening levels, ensuring that excavation activities did not "chase" material that was not contamination related to the Site. The ratios of three key rare erath elements for Welsbach impacted material and materials impacted by coal ash can be seen in Figure 1.

Data from technical literature reports and previous New Jersey Department of Environmental Protection investigations into local rare earth concentrations was also used as an information source, which assisted in the overall analysis and regulatory acceptance.

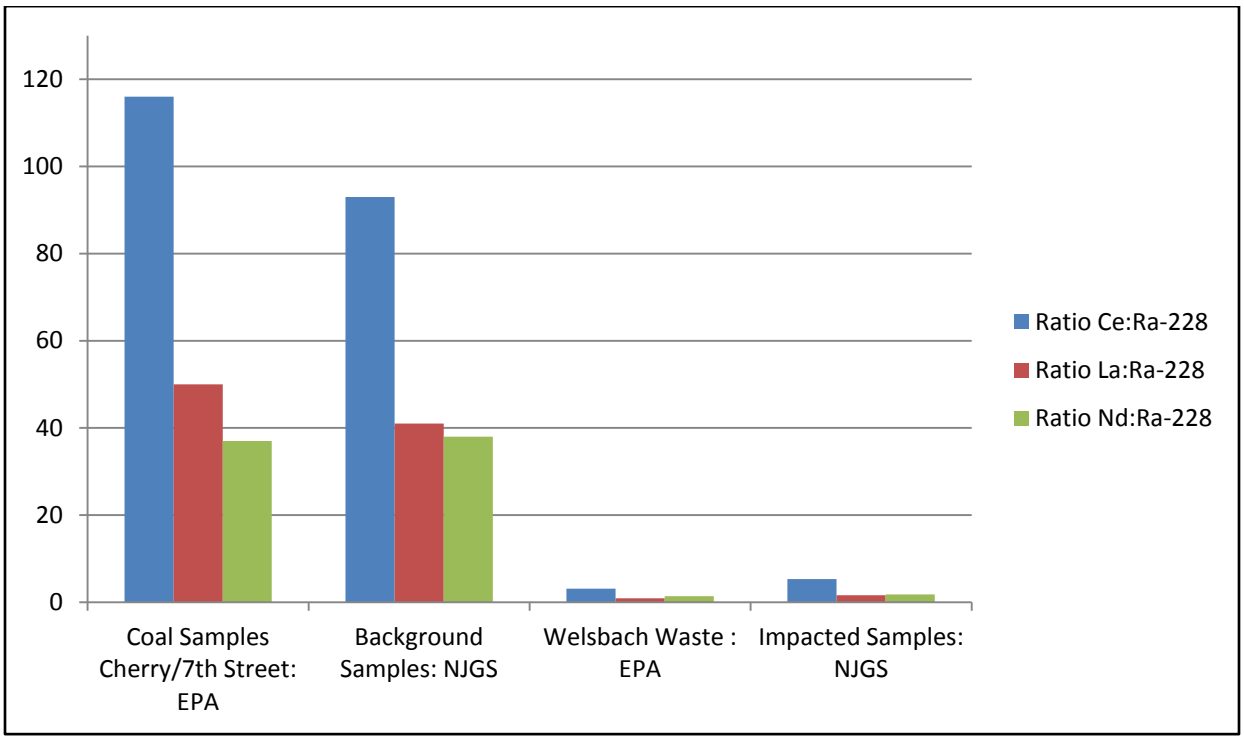

Figure 1 – Welsbach Rare Earth Element Ratios

Rare earth analysis has also been used at the FUSRAP Maywood Superfund Site (FMSS).

Rare earth and thorium processing operations at the FMSS during the first half of the last century generated wastes containing thorium, radium, and uranium. These wastes entered the environment primarily from disposal in burial pits, through transport via surface/storm water, and use of the material as fill. NORM is known to occur in the FMSS area. Addressing these natural materials is

outside the authority of FUSRAP. The RE content of soils on properties with elevated NORM have been used by DOE and USACE as one dataset to determine if soils with elevated radionuclides are consistent with FMSS wastes, natural sources, or other potential sources.

The RE content of background soil, soil at selected FMSS properties, and in FMSS wastes was presented in the FMSS 1992 Remedial Investigation Report for the Maywood Site. The RI stated "Analytical data indicate a good correlation between the presence of radioactive contamination and the occurrence of various rare earth elements." In areas where radioactive contamination is absent, rare earth elements were detected infrequently and at trace levels. Cerium, lanthanum, and neodymium were detected with greater frequency and at higher concentrations than other rare earth elements. Concentrations of rare earth elements encountered at the Maywood Site are shown in Figure 2.

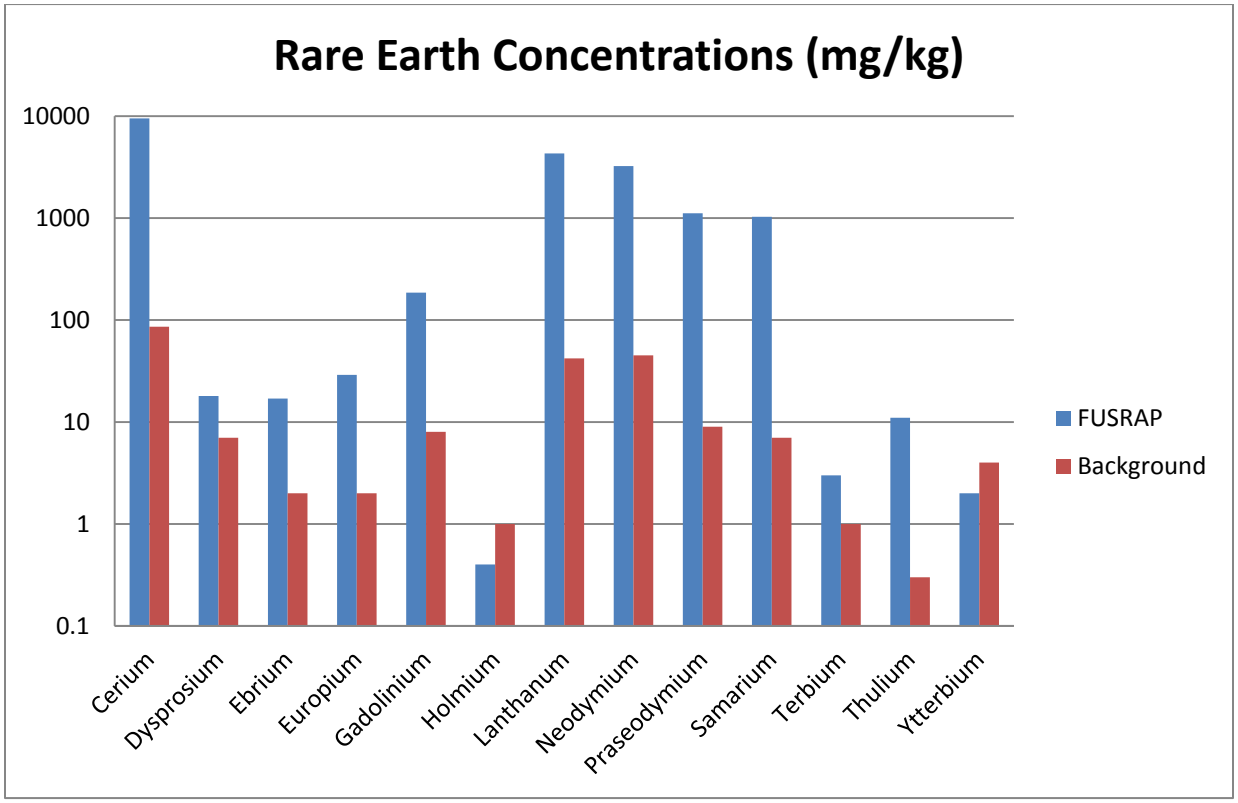

Figure 2 – Maywood Rare Earth Element Concentrations

The distinctions between waste and background concentrations provide a basis for determining if a material is related to FUSRAP wastes or not. Elevated cerium, lanthanum, neodymium, praseodymium, samarium, and to a lesser extent gadolinium concentrations are representative of FUSRAP wastes. USACE primarily utilizes cerium, lanthanum, neodymium, praseodymium, and samarium concentrations to make FUSRAP waste determinations.

Key FUSRAP waste indicators of rare earths are cerium, lanthanum, neodymium, praseodymium, samarium, and to a lesser extent dysprosium. While these RE are also found in background samples, the magnitude of concentrations varies significantly.

Rare earth analysis is a relatively inexpensive, quick, and straightforward analysis to perform on soil samples and, as long as a good rare earth profile of both contaminated and natural (in this case fly ash and coal ash) material can be established, the results can be quite definitive, resulting in significant volume reduction, minimizing the amount of material excavated and disposed of, leading to cost and time savings for what could be an otherwise costly and potentially complex excavation. The differences in rare signatures between FUSRAP wastes and background are significant. Accordingly, the use of rare analysis is an effective tool to aid in identifying potential FUSRAP wastes and conversely in releasing properties with NORM from FUSRAP consideration. Real case examples demonstrate the importance of understanding the methods of distinguishing wastes.

NATURALLY OCCURING URANIUM

Wetlands are known to accumulate U. In fact, artificial wetlands are being used to remediate some U contaminated water. U disequilibrium has been used in numerous studies to age ground water.

The exact reasons wetlands accumulate U and other metals depends on the makeup of the wetlands but involves bioorganisms, plants, redox potentials, carbon content, pH, and soil types.[7] It is also noted that the accumulation is most prevalent in the shallow surface of the wetland (to a depth of roughly 20 cm).

Two natural processes (Alpha recoil and alpha track) can result in enhancing the activity concentration of U-234 in ground water. The first method is from alpha recoil. When an atom of U-238 decays the force of expelling an alpha particle may eject the resultant Th-234 atom from the structure that the atom is in, into the pore space. The Th-234 then decays to Pa-234m, then to U-234 which is moved by water in the pore space. The other method is the creation of alpha tracks in the solid structure allowing the ultimate U-234 atoms to be transported by pore space water. An illustration of this process is shown in Figure 3.

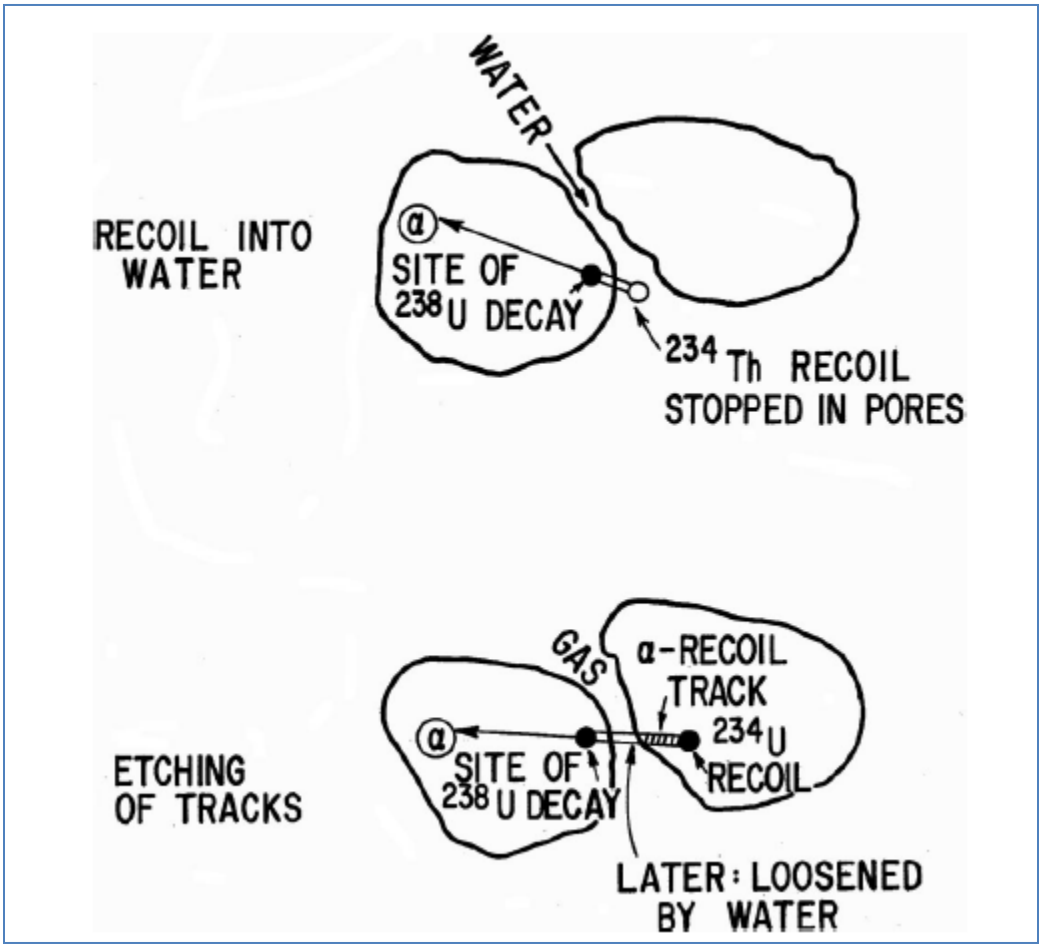

Figure 3 – Alpha Recoil Process

These two methods result in the enrichment of U-234 in ground waters. This is most common with waters moving through U bearing materials. U-234 to U-238 ratio in sea water is 1.14. The same ratio in groundwater can vary significantly between water sources and ranges from 1 to 3 with a typical mean of around 2.

LEAD-210 AND RADON DAUGHTERS

Elevated Pb-210 in soils is often encountered, due to natural processes involving radon-222 daughter washout from rain events and accumulation, and is not uncommon. Slightly elevated levels of Pb-210 are commonly found in low lying areas where rain collects and concentrates. Pb-210 levels ranging up to 20 pCi/g or even higher can be found when analyzing soil and sediment samples collected from these areas. The following quotation is taken directly from the Multi-Agency Radiation Survey and Assessment of Material and Equipment (MARSAME) manual (USEPA, 2009)

> *"Radon emissions vary significantly over time based on a wide variety of factors. For example, relatively small changes in the relative pressure between the source material and*

the atmosphere (indoor or outdoor) can result in large changes in radon concentrations in the air. Soil moisture content also has an effect on the radon emanation rate.

Radon progeny tend to become fixed to solid particles in the air. These particles can become attached to surfaces as a result of electrostatic charge or gravitational settling. Air flow through ventilation ducts can produce an electrostatic charge that will attract these particles. A decrease in atmospheric pressure often precedes a rainstorm, which increases the radon emanation rate. Immediately prior to an electrical storm, an electrostatic charge can build up on equipment resulting in elevated radiation levels from radon progeny. Rainfall acts to scavenge these particles from the air, potentially resulting in elevated dose rates and surface activities during and immediately following rainfall.

Pb-210 is a decay product of 222Rn and 238U. The 22-year half-life provides opportunities for buildup 210Pb and progeny in sediments and low-lying areas. As mentioned previously, rain acts to scavenge radon progeny from the air. Areas where rain collects and concentrates can result in elevated levels of 210Pb and progeny over time. In addition, lead is easily oxidized and can become fixed to surfaces through corrosion processes. Rust or oxide films on equipment can be indicators of locations with a potential for elevated background radioactivity."

This radon daughter washout process is a known and established natural process. Indeed, Pb-210 accumulation and radiometric dating is an established technique used in geology to compare sedimentation rates and determine the age of deposited material. An illustration of this process is given below in Figure 4.

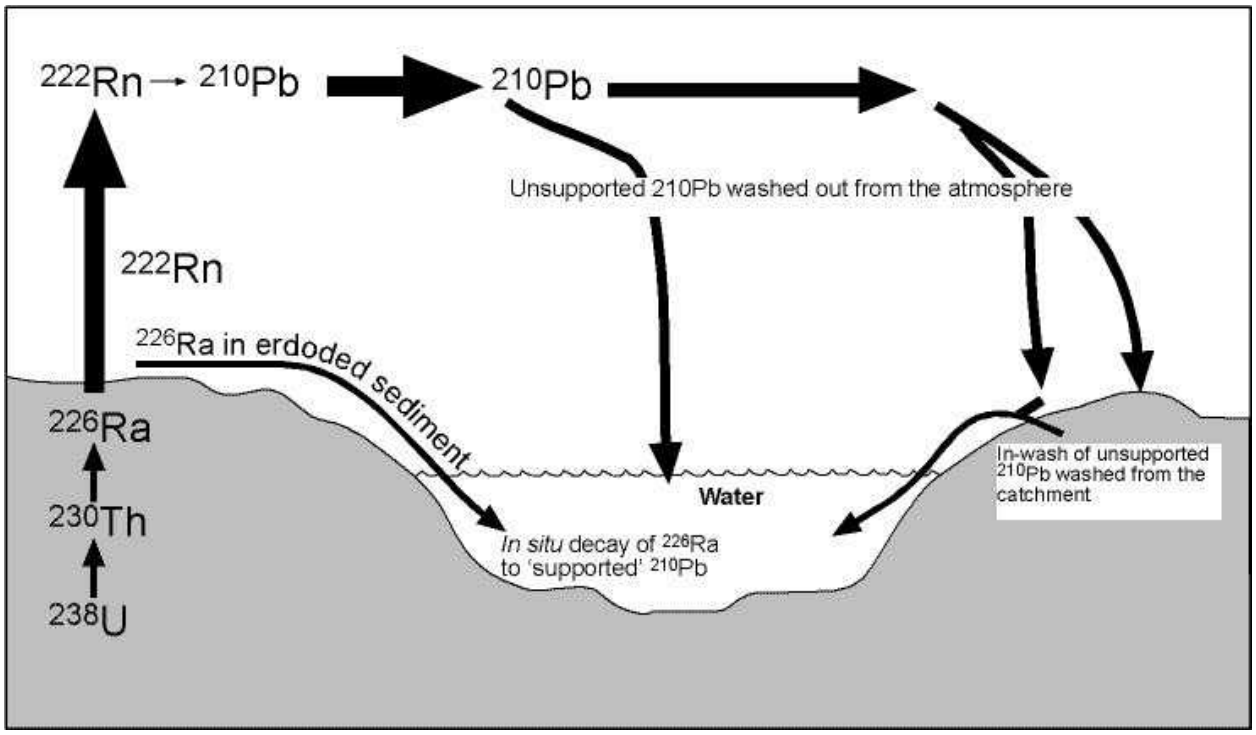

Figure 4 – Pb-210 Washout Process

Elevated Pb-210 levels were encountered in several Dayton, Ohio FUSRAP Site samples collected by USCAE. These samples were considered to be within normal background for the areas they were collected.

Radioactive material reported to exist in sediment outside the Dayton Unit III (Bonebrake) site is not outside the range of naturally occurring background levels for Pb-210. One might also find Pb-210 in underground confined spaces, such as a manhole, that tend to concentrate naturally occurring radioactive materials from the decay of radon gas. Pb-210 that was detected in sediment from a manhole at a concentration of 11 pCi/g is within the range of values reported for some background sediments in the state of Ohio. Ohio sediments in nature may contain Pb-210 at 5.1 to 19 pCi/g as measured in an Ohio no-till basin. At the Dayton Unit III Site, a “relatively elevated” concentration of Pb-210 measured in sediment from the manhole could be attributable to the run-off and concentration of surface soil fines containing recently deposited Pb-210 fallout. Airborne Pb-210 is generated continuously by the emanation of radon from soils containing the naturally occurring uranium decay chain.

SUMMARY AND CONCLUSIONS

In summary, many naturally occurring radionuclides can be found comingled with potential Site contamination across all types of media. Rather than simply accepting the potential for additional expenditure of resources to deal with elevated areas, a variety of analysis tools and techniques exist to differentiate between Site contamination and non-site related material. As part of any attempt to distinguish between contamination and natural material it’s important to maintain a

consistent and technically defensible approach and ensure the proper formal processes are in place. By distinguishing between Site and non-Site material significant project cost and schedule savings can be realized.

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