Distinguishing Between Site Waste, Natural, and Other Sources of Contamination at Uranium and Thorium Contaminated Sites - 12274

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ABSTRACT

Uranium and thorium processing and milling sites generate wastes (source, byproduct, or technically enhanced naturally occurring material), that contain contaminants that are similar to naturally occurring radioactive material deposits and other industry wastes. This can lead to misidentification of other materials as Site wastes. A review of methods used by the US Army Corps of Engineers and the Environmental Protection Agency to distinguish Site wastes from potential other sources, enhanced materials, and natural deposits, at three different thorium mills was conducted. Real case examples demonstrate the importance of understanding the methods of distinguishing wastes.

INTRODUCTION

In today's world of budget constraints and limited funding, distinguishing between Site related wastes and other sources of radioactive materials is of greater importance than ever. Additionally, programs such as the US Army Corps of Engineers (USACE) Formerly Utilized Sites Remedial Action Program (FUSRAP) have specific authorities which preclude them from addressing natural materials or non FUSRAP contamination.

The most obvious and effective method for reduction of cost impacts is excluding the other (non site related) source areas from remediation and additional consideration. In order to utilize this method, Site personnel require knowledge of methods to differentiate multiple sources from Site wastes as well as a process to do so.

A review of three different Thorium (Th) mills' remediation and waste distinguishing approaches was conducted. Each facility had complex contaminant migration conceptual models in urban environments. Each facility is also located in areas where significant filling of areas has occurred over many years. The various periods of fill and unknown sources of fill materials contribute to the necessity to have methods established to distinguish waste materials. Failing to distinguish between site wastes and natural or other sources of contamination in these environments would and in some cases did increase project costs.

METHOD

Each facility had some common as well as unique conditions and materials that required varying approaches to waste identification. The specific conditions (other sources, natural processes, and natural deposits) at each facility are not specifically discussed herein. Each facility's approach to distinguishing Site wastes from other materials were reviewed and organized to develop a process of determination.

DETERMINATION PROCESS

A formal approach to distinguishing Site wastes is important to reduce the costs associated with doing so. See Figure 1 for an illustration of the Determination Process developed from this review. The process consists of 4 phases. These are Historical Records, Site Process/Waste Knowledge, Characterization, and Documentation.

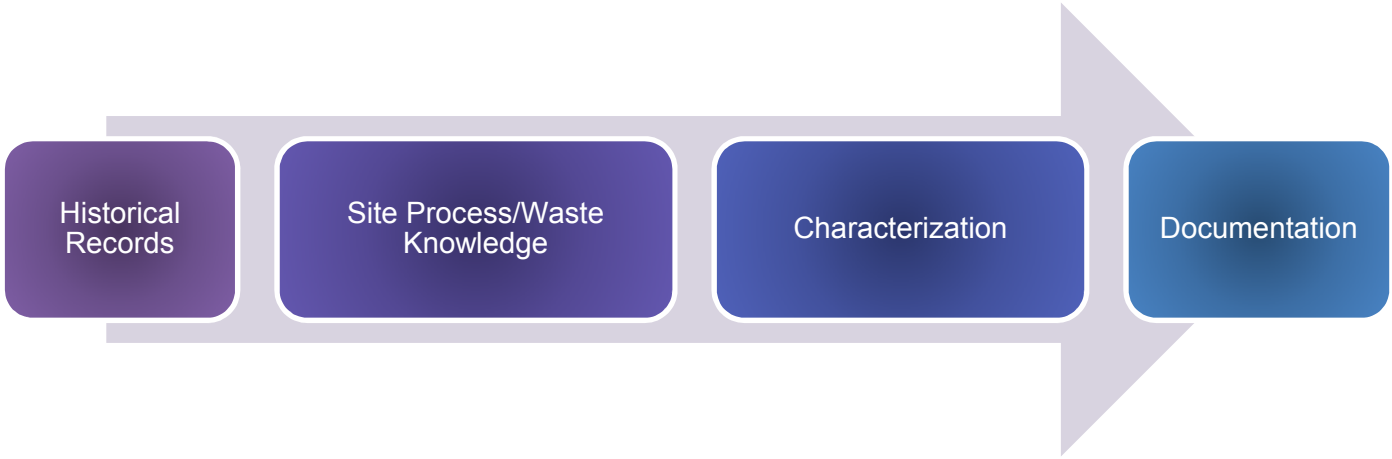

Fig. 1 Determination Process

DISCUSSIONS

There are many potential sources of radioactive materials that may be encountered at a remediation Site. The most common is material found naturally in the Site's environment. This material is referred to as Background and consists of Naturally Occurring Radioactive Material (NORM) at typically very low concentrations. These background materials typically are primordial (potassium-40), originate from the natural decay chains (Uranium, Thorium, Actinium chains), or are produced by natural interactions. These are Site specific and concentrations can vary significantly. Distinguishing between typical background and Site wastes is the subject of many papers and guidance documents and is not specifically discussed further in detail. Distinguishing from altered or enhanced background sources and Site wastes may be a key aspect to controlling costs. The term enhanced background is used here to identify NORM that is elevated in radioactivity above typical Site Background levels.

When a natural radioactive material concentration is concentrated or enhanced by man or nature and that material is located on a Site, two primary issues are encountered (Site overall risk and distinguishing from wastes). The two issues are related and can be quite complex. Often Site personnel opt to remediate any radioactive material above the Sites criteria due to concerns over total site risk or dose. This may significantly increase the cost of remediation.

Examples of NORM enhanced by man include coal ash, oil industry wastes, byproduct materials, sources, fertilizers, and many others, See table I. Natural processes such as physical deposition (wind and placer deposits) and chemical mobility (ground water) can also alter or

enhance NORM concentrations in specific areas. Sites may address these specific areas in several ways but exclusion from remediation or further consideration is often the least expensive approach.

Table I Radioactivity in Man and Naturally Enhanced Background Materials

Material	Process	Potential Isotopes	Literature maximum values (rounded)
Coal Ash	Burning of coal results in concentrating NORM. The ash has been used as fill and is used as an ingredient in many types of concrete	Uranium, Radium-226, Thorium-232 and daughters	U - 100 ppm Ra-226 – 0.4 Bq/g Th-232 – 0.8 Bq/g
Phosphate Fertilizers	Phosphate ore materials often contain NORM, U tends to stay with the phosphate in the manufacturing process	Uranium isotopes and short lived daughters	2 Bq/g
Fertilizer production Tailings	Phosphate ore materials often contain NORM, Radium tends to remain with the tailings from phosphate extraction	Ra-226 and daughters	2 Bq/g
Oil and Gas industry	Process waters contain elevated concentrations of NORM, this NORM can plate out in piping and accumulate in soils under lagoons holding this water	Ra-226, Pb-210, and daughters	125 Bq/g 86% of data < 3.7 Bq/g
Ore Milling	NORM isotopes can be concentrated in milling many types of metal ores. Aluminum, Zirconium, and magnesium production can generate significant NORM radioactivity in tailings	Th-232, Ra-226, U-238 and daughters	370 Bq/g
Ceramics	Some Clays and glazes have natural radioactive material	Th-232, Ra-226, U-238 and daughters	2 Bq/g
Groundwater	Groundwater dissolves many natural radioisotopes from the deposits which it flows. Note: Enrichment of U-234 in ground water does occur , U-234 tends to mobilize more than U-238 and U-235	Ra-226, Ra-228, Pb-210, Uranium,	Ra-226 – 1.15 Bq/L Ra-228 – 2.72 Bq/L Pb-210 – 0.28 Bq/L Uranium – 10,000ug/L
Plants	Certain plants accumulate radioactive materials and some are used to remediate contaminated areas	Uranium	Approximately 50% to 80+% of water concentration retained in plants
Wetlands	Wetlands accumulate many metals including radioactive material	Uranium	Approximately 50% of water concentration retained in wetland

Historical Records

Historical Site Assessments (HSA) are often focused on a facilities use of materials and Site waste areas. Less focus is placed on other wastes and or other materials that may be of concern at this stage of most projects. The same information that may be used to demonstrate where Site materials may be expected, may also provide information on potential other materials. Information on other potential sources of radioactive materials should be noted during HSA.

Navigation maps, aerial photography, fire insurance maps, photographs, and other historical records may provide key understandings that help limit cost and schedule impacts during construction [1]. Understanding key historical information such as the development history of an area, probable fill areas, and important local features effecting contamination distribution not only provides a better understanding of Site contamination it also provides a basis to question if elevated materials are actually Site related. This can be used to delineate areas for further review or sampling as discussed herein.

In one example a former creek was identified in historical maps. Other maps and figures demonstrated that the creek had been filled in prior to the time of Site operations impacting the area. The dates of fill helped to highlight that elevated radioactive materials in the fill may not be from the Site. An additional review of data from the area was prompted and additional sampling conducted to distinguish Site contamination from above background NORM. This resulted in specific areas being excluded from further consideration and significant cost avoidance for the project.

Site Process/Waste Knowledge

As in the case of historical records understanding the process generating a waste can be used to identify Site contaminants and also aid in distinguishing wastes from above background NORM. Knowledge of what other materials and chemicals are associated with Site wastes can be used to facilitate or eliminate the Characterization phase.

As an example, at a gas mantle production facility both thorium and cerium (Ce) were required to create coatings for mantles. These elements are often found together in natural deposits and as such, the ores brought to the facility for processing contained both thorium and cerium. These ores also have elevated concentrations of other rare earth elements. Waste from the facility would therefore be expected to contain both Th and Ce as well as other associated rare earths. Sampling for rare earth content could be added to the characterization phase in suspect areas.

Understanding how wastes were handled may provide key information as to the probability of a material being site related. Other wastes associated with other Site activities may prove useful as well.

At the above gas mantle production Site example, material appearing to be elevated background NORM just over the Site release criteria was discovered in an area not suspected of containing Site wastes. Site personnel considered further data review and characterization to prove the material was not Site related. In this example however, the fact that mantle cloth was also found in the area provided sufficient justification to include as Site waste. Although a modest savings, the cost and schedule impacts of additional characterization was minimized by understanding the facilities other processes.

Characterization

There are many NORM characteristics that can be used to distinguish site wastes from natural and other enhanced back ground materials. Chemical, isotopic ratios, and Uranium (U) isotopic natural abundance are just a few.

Chemical and other by products associated with a process serve as finger prints to identify Site waste. A good example of this is thorium milling wastes which typically contain rare earth elements. Vicinity properties at two thorium mills were identified as having above criteria thorium contamination. A rare earth characterization sampling approach was implemented on properties that were identified through the Historical or Process Knowledge phases as unlikely to contain Site wastes. Rare earth analysis was conducted on typical site wastes as well as on elevated background soils from the vicinity properties. The rare earth analysis comparison proved that material on the properties in question was not Site related and was likely natural.

Isotopic ratios of wastes versus other materials can also be used to distinguish between Site related wastes and enhanced background materials. At the thorium mill sites studied, typical Th-232 to Radium-226 (Ra-226) ratios in waste are on the order of 4:1 to over 20:1. This ratio in natural background materials is closer to 1:1. The Th-232:Ra-226 ratio in enhanced NORM such as coal ash can vary significantly but experience on 2 of the 3 thorium mill sites demonstrates Th-232:Ra-226 ratios of 1:1 to 2:1 in coal ash. It is notable that on the 3rd Site coal ash ratio was 1:1 but in coal ash with cinder chunks the ratio was as high as 20:1.

Given its historical use in industry and home heating in the areas of these sites, coal ash is ubiquitous and a significant contributor to local and specific background at all three study sites. Depending on the type and grade of coal burned, the ash can contain significant radioactivity compared to background soils. Use of isotopic ratios has been used to distinguish Site wastes from coal ash on many Sites.

Uranium is found in most Th ores and a typical ratio of U to Th can be calculated. This isotopic ratio is disturbed by milling but the waste may still exhibit a consistent Th-232 to U-238 ratio. At one mill Site this ratio was used to demonstrate that natural deposits were not Site related.

Another key approach to identifying wastes is the natural U isotopic abundance. Natural U consists of U-238, U-234, and U-235 in a 1:1:0.046 activity ratio respectively. Obvious disruption of this ratio occurs during U enrichment processes. Enrichment results in enriched U (U

enriched in U-235 content) and depleted U (U depleted in U-235 content). Uranium enrichment or depletion is used at many Sites to distinguish between Site wastes and natural materials.

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. [2] 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. [3] The same ratio in groundwater can vary significantly between water sources and ranges from 1 to 3 with typical mean around 2. [4]

A key aspect of all 3 study sites was borings. Boring logs provide valuable characterization data if completed properly and by a geologist. The geologist can identify soil types as well as other sources of radioactivity such as coal ash in the logs.

Physical traits and materials types such as color and composition can also be indicators of non Site wastes. Elevated gamma scan readings at a Site were associated with road pavers referred to as Belgian Block. The blocks were not considered site wastes but the soil immediately beneath the blocks also contained elevated radioactivity. A comparison of isotopes and isotopic ratios demonstrated that the soil was impacted by fine particles eroding from the blocks rather than site wastes.

Documentation

The Determination Process should be documented for the Site or project files. Often Site regulators request to review the documentation, so it should be prepared accordingly. USACE uses many methods to document decisions regarding Site wastes. The least onerous method is preparation of a Technical Memorandum.

EXAMPLE (ACTUAL) CASE

The following is an actual example of using the Determination Process on an actual Site area. Site related wastes are from thorium milling and contain Th-232, Ra-226, U-238, and their associated daughter products. This example case resulted in hundreds of thousands of dollars of cost avoidance for the project. Water (surface and ground) control, water treatment, excavation depths, and shoring costs would have resulted in increased costs, as well as the increased material handling and disposal costs.

Example Historical Records

The study Property, hereafter referred to as the Property, is bordered on the West by a railroad and on the East by a Brook. On the East bank of the Brook is an Apartment Complex Property (ACP).

A review of historical information and aerial photographs (1931 to present) demonstrates the following:

- The Property at one time was a vacant wetland area that was later filled in by debris and other fill. The property is triangular in shape.
- Prior to the ACP being built in 1948 the area East of the Brook was farmland with an irregular shaped boundary with the former wetland area. After the ACP were built the Brook was almost a straight non vegetated channel between the Property and ACP.
- The railroad is evident on the earliest photograph from 1931. The railroad line is documented as being built in the 1902-1908 timeframe.
- The X Chemical Co. transported thorium and cerium process building debris and other wastes to the Property during 1964.
- Additional fill was placed on the Property after 1964 by unknown parties.

Further research identifies the Brook as a perennial spring fed stream that originates just south of the property under a parking lot and flows north approximately 3 miles to a River. The Brook is in a culvert until it daylights at the Property, approximately 100 meters from its origin. The Brook sub-watershed drains approximately 17 acres and includes the Property. Land use in the local Brook watershed is mixed undeveloped, residential, and commercial.

Previously a series of soil borings on the ACP were done as a supplemental characterization effort to determine if Site related materials may be East of the Brook. Eighteen borings were installed and sampled between the Brook and the apartments. Elevated U results were found in a dense organic layer approximately 3 meters below ground surface (bgs).

The dense organic layer had been identified across the Property at 1 to 2 meters bgs and the railroad property at 1 to 2 meters bgs. This organic layer (termed “Swamp mat” by work crews) represents the historical (pre-fill) wetland surface of the properties. When the different depths of fill on each property are considered the Swamp mat layer is fairly flat (at the same elevation).

Example Site Process and Waste Knowledge

It is clear that Site waste is present on the Property. Given that Site wastes do contain U the potential exists that the elevated U under the ACP and Railroad properties is Site related. Understanding the Site waste and potential processes of its transport helped to focus the evaluation and characterization.

Potential waste transport mechanisms are:

- Wind/Air (dusts)
- Physical Placement (fill or dumping)
- Water (runoff, groundwater, flooding, erosion/sedimentation)

Screening Evaluation of potential transport mechanisms are in Table II Ex-1 below.

Table II Ex-1. Transport Mechanism Screening

Mechanism	Type	Method	Discussion	Retained
Air	Dust	Wind dispersion	Dusts would carry all Site waste Contaminants of Concern (COC) not just U. Placement of Site wastes was well after filling of railroad and ACP properties.	No
Physical Placement	Fill	Artificial fill, Burial	All Site COCs would be present. Aerial photos do not show areas of disturbances after the Railroad and ACP were present. Vegetation along Brook not disturbed and former structure on the ACP property would have prevented wastes placement.	No
Water	Surface	Runoff	Brook serves as a drainage for the Railroad and the Property, thus runoff would be carried off. All Site COCs would be present.	No
	Surface	Flooding	All Site COCs would be present. Elevated material would be in many layers and on/near the surface.	No
	Surface	Erosion/Sedimentation	All Site COCs would be present.	No
	Ground	Lateral Movement due to perched water	Uranium is generally more soluble than other Site Waste COC. Overburden water does move. Would expect to find elevated U in other layers but possibly trapped by the organic layer only.	Yes
	Ground	Dispersion via groundwater flow	The only aquifer is the bedrock aquifer. The known Site wastes are buried in the overburden. U is elevated in groundwater at the property but below action levels.	Yes

Retained Mechanisms.

The two (2) retained potential mechanisms to transport Site wastes from known fill areas to other portions of the property and adjacent properties both involve the flow of ground water. In

these theoretical scenarios, the movement of water through the contaminated overburden would dissolve U and transport it laterally. This dissolved U would then precipitate out of solution when encountering the organic Swamp mat layer. Since the Th in the Site wastes is relatively insoluble it is not expected to move with the U. It is also expected to be primarily Th-232 as in the wastes. Additional characterization (U and Th isotopic analyses) is recommended.

Example Characterization

Samples were collected and analyzed for isotopic U and Th. Results are in Table III Ex-2 below. The Th-232 results from the Swamp mat layer were at background levels which do not fit the Site waste signature.

Table III Ex-2. Isotopic Results (pCi/g) from Swamp Mat under Railroad Property.

Gamma Spectrometry Ra-226	Gamma Spectrometry Ac-228	Alpha Spectrometry U-234	Alpha Spectrometry U-235	Alpha Spectrometry U-238	Alpha Spectrometry Th-230	Alpha Spectrometry Th-232
.37	1.13	20.52	0.83	12.87	2.40R	1.29R
1.36	0.39	76.50	7.62	59.54	3.53R	2.31R
3.86	1.09	549.05R	11.64R	304.51R	NA	NA
2.63	2.14	35.92	0.78	22.14	3.51	0.48
1.73	1.25	65.34	23.92R	95.32	8.35	0.34
2.81	1.57	165.96	3.93	103.16	10.1	0.88
Mean (“R” qualified data not included)						
2.29	1.26	72.85	3.29	58.61	7.32	0.57

Note: U-234/U-238 = 1.2; U-235/U-238 = 0.05; Th-230/U-234 = 0.10; Th-232/U-238 = 0.01.
NA = Not Analyzed.
R = Rejected data based on lab quality control procedures.

Gamma Spectrometry results from the 18 borings on the ACP property also identified elevated U in the absence of elevated Th-232. Accordingly, samples of the Swamp mat were analyzed by alpha spectrometry for isotopic U and Th. Results of isotopic analysis are included in Table IV Ex-3 below.

Table IV Ex-3. Isotopic Results (pCi/g) from Swamp Mat layer (10-12 feet bgs) ACP Property.

Gamma Spectrometry Ra-226	Gamma Spectrometry Ac-228	Alpha Spectrometry U-234	Alpha Spectrometry U-235	Alpha Spectrometry U-238	Alpha Spectrometry Th-230	Alpha Spectrometry Th-232
5.98	1.54	518	16.1	317	20.3	0.77
2.61	1.31	170	5.38	104	8.62	0.40
1.94	1.38	NA	NA	NA	NA	NA
1.70	1.09	NA	NA	NA	NA	NA
3.46	1.12	NA	NA	NA	NA	NA

3.40	0.91	179	5.64	109	12	0.13
1.26	0.92	NA	NA	NA	NA	NA
Mean ("R" qualified data not included)						
2.91	1.18	289	9.04	176.67	13.64	0.43

Note: U-234/U-238 = 1.64; U-235/U-238 = 0.05; Th-230/U-234 = 0.05; Th-232/U-238 = 0.002.
NA = Not Analyzed.
R = Rejected data based on lab quality control procedures

The elevated Swamp mat U data validity was questioned at first given the appearance of Enriched Uranium (EU). The U-234 ratio to U-238 suggested an enrichment of approximately one (1) percent. When the U-235 to U-238 ratio was considered the U appears to be natural. All isotopic data was validated by an independent 3rd party. Data Qualifiers were assigned and only unqualified data was used in this evaluation. This answered the question of the data quality but not that of the U disequilibrium.

Data (gamma spec) from the Swamp Mat and the known Site Wastes on the Property are presented in Table V Ex-4 below.

Table V Ex-4. Results (pCi/g) from Swamp Mat and Site Wastes at the Property.

	Gamma Spectrometry Ra-226 (Pb-214)	Gamma Spectrometry Th-232 (Ac-228)	Gamma Spectrometry U-238 (Th-234)
Site Wastes	52.19	300.51	46.13
Swamp mat layer 1	4.79	1.43	49.16
Swamp mat layer 2	2.80	0.91	81.48
Swamp mat layer 3	2.82	0.99	64.47
Swamp mat layer 4	2.18	1.18	158.76
Mean of Mat Layer	3.15	1.13	88.47

Note: Th-232/U-238 ratio of wastes = 6.51; Th-232/U238 of mat layer = 0.013.

Uranium Disequilibrium

The U-235 to U-238 ratio is consistent at approximately 0.05 which approximates the expected natural abundance ratio of 0.046 (especially given measurement uncertainty).

The U-234 to U-238 ratio varies in water samples but the mean ratio is 1.7. Approximating the expected ratio from 1% enriched U.

U disequilibrium has been used in numerous studies to age ground water. U-234 is preferentially mobilized in ground waters. A review of U isotopic data from groundwater at the Property, sediment data from Brook, and surface water from Brook, indicates that U is elevated in each and the typical U-234 to U-238 ratios for each demonstrates disequilibrium. Results are provided in table VI Ex-5 below.

Table VI Ex-5, Data from the Property and the Brook

Media	U-234 (pCi/l)	U-238 (pCi/l)	Ratio U-234/U-238
Sediment [(a)]	2.38	1.34	1.77
Surface Water [(a)]	2.01	1.18	1.70
Surface Water [(a)]	1.85	1.37	1.35
Overburden Water [(b)]	12.57	6.36	1.98
Bedrock Water [(c)]	1.55	0.83	1.88

(a) Ground Water Remedial Investigation Data
(b) Monitoring Well Data
(c) Mean Dewatering Well Data (representative of Bedrock aquifer, treatment system data from wells only)

Uranium Accumulation (in the Swamp mat layer)

Wetlands are known to accumulate U.[5] In fact, artificial wetlands are being used to remediate some U contaminated water.[6]

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).

Given that the water at the properties and from the Brook is enriched in U-234 and the known precipitation of U in wetlands, it is likely that the original wetland surface (now the Swamp mat layer) accumulated U naturally from the Brook.

Th-230 and formation age determination.

Since Th is relatively insoluble it would not move with the U. Isotopic Th data from the Swamp mat layer on the Railroad and APC properties show background levels of Th-232 but elevated levels of Th-230. Th-230 is not a Site COC and is a decay product of the Uranium decay series. Since relatively insoluble, any ingrown Th (from the decay of U) would remain in the Swamp mat layer. Th-230 will ingrow from the decay of U-234 at a known rate. Accordingly, the Th-230 concentration can be used to date the age of the U in the Swamp mat. Using the equations for decay chain daughter calculations [8] the U in the Swamp mat layer would be much older than that potentially moving from Site wastes. Calculations using slightly different assumptions of U-234 and/or U-238 derived Th-230 and initial concentrations, indicate that the U in the Swamp mat layer would be between 4,000 and 75,000 years old. Additionally, dating of the material was conducted using equations from Wijk 1987.[9] This resulted in an age estimate of 3,000 to 5,500 years. Both estimates agree well with the age estimate of North American U bogs as reported in *Bog Dating***,** By G. L. Ziegler.[10]

Given the time required to in grow Th-230 at the levels observed in samples from the ACP and railroad properties it is likely that the U present in the Swamp mat layer is naturally occurring.

Example Conclusions

The elevated U under the ACP and railroad properties associated with the Swamp mat layer is not Site related.

It is likely that the elevated U associated with the Swamp mat layer is from natural accumulation of U from the waters of the Brook.

CONCLUSIONS

Distinguishing between Site wastes and enhanced Background material can be facilitated by establishing and applying a formal process.

Significant project cost avoidance may be realized by distinguishing Site wastes from enhanced NORM.

RECOMMENDATIONS

Collection of information on other potential sources of radioactive material and physical information related to the potential for other radioactive material sources should be gathered and reported in the Historical Site Assessment. At a minimum, locations of other such information should be recorded.

Site decision makers should approach each Site area with the expectation that non site related radioactive material may be present and have a process in place to distinguish from Site and non Site related materials.

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