UNCOVERING CLUES ABOUT AN OLD BURIAL SITE

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ABSTRACT

An investigation is currently being conducted to resolve questions about the contents of a site in central Michigan that was used 30 years ago for the disposal of waste material from a rare earth extraction process. The waste material contained precipitated uranium and thorium compounds. A limited site characterization and a thorough review of available historical data have provided many clues about the quantity and characteristics of the buried material. Unfortunately, these clues do not lend themselves well to a finite conclusion. Challenges regarding the interpretation of these clues have included the relatively small volume and heterogeneous nature of the buried radioactive material, analytical difficulties in determining the uranium activity, and data sets that support dissimilar conclusions. Once these and other difficulties are addressed, an estimated source term will be developed to support a dose assessment. Another issue that must be addressed is how modern-day standards apply to this site.

INTRODUCTION

A 2-acre former radiological disposal site, known as the Breckenridge Disposal Site (see Figure 1) is located near St. Louis, Michigan. The disposal site is triangular in shape, bordered on the north by a public road, the east by a creek, and is generally surrounded by farmland. The grassy site is level with surrounding land and has some large trees on the southern tip of the property.

Michigan Chemical Company (MCC) used the site for the disposal of process wastes containing thorium and uranium. The wastes were a byproduct of MCC's rare earth extraction process, which used thorium-and uranium-bearing feedstocks. The rare earth extraction process primarily yielded yttrium, which was used as a phosphor for color televisions and to produce synthetic diamonds and crystals that were used in various electronic devices.

The solid waste resulting from the extraction process was a precipitate of insoluble materials known as filtercake. The filtercake contained much of the radioactivity from the process' feedstock and was buried at the disposal site between 1967 and 1970 under a U.S. Atomic Energy Commission license and in accordance with the U.S. Code of Federal Regulations (CFR), Title 10, Chapter 20.304, "Disposal by Burial in Soil."

Site evaluations conducted by Oak Ridge Associated Universities and later by the United States Nuclear Regulatory Commission (NRC) after the site's closure noted elevated levels of surface activity at a limited

number of locations at the disposal site. Since 10 CFR 20.304 required an earthen cover of no less than 4 feet, the surface radioactivity raised concerns about the condition of the burial site and its potential impact on human health. Based on the identification of the surface activity, the NRC requested that the site's current owner, NWI Land Management (NWI), conduct a site radiological evaluation. The NRC

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also directed NWI to close the site in accordance with today's regulatory framework of 10 CFR 20, Subpart E, commonly known as the License Termination Rule. NWI contracted the radiological services of SCIENTECH NES, Inc. to characterize the disposal site, conduct a historical site assessment, complete a dose assessment, and provide other technical services in support of site closure.

In order to determine the level of public risk posed by the former disposal site, SCIENTECH NES initiated a site investigation that included a site surface survey and subsurface sampling and analyses. Also key to the investigation was a review of supporting historical data.



Figure 1. Breckenridge Disposal Site Location

HISTORICAL DOCUMENT REVIEW

The historical site assessment for the disposal site generally consisted of accumulating and reviewing all available historical documents with information pertinent to the site's radiological condition. Recovered documents were reviewed to better determine the type, quantity, activity, location, burial depth, cover depth, and any other relevant data associated with the waste material. The historical documents basically fell into three categories:

- Test and evaluation data that was generated during the engineering design phase of the extraction process (1961 to 1966).
- Data stemming from the operations and disposal period from 1967 to 1970.
- Radiological evaluation data generated from an investigation of the disposal site in the early 1980s (in conjunction with the closure of the extraction plant site)

To support the historical site assessment, the NRC Public Document Room was contacted with a request for a listing of all documents pertaining to MCC's materials license. Through this request, the NRC provided copies of all related documents including license requests, radioactive materials licenses, inspection reports and other related documents.

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NWI requested the copies of all active and archived project files regarding the disposal site from its corporate predecessor. These documents included radioactive material licenses, correspondence to and from State and Federal regulatory agencies and inter-office correspondence. The former MCC waste control engineer, who is now retired but lives nearby, also was contacted and interviewed to attempt to verify information found during the document review and to attempt to fill data "gaps".

The recovered historical data was complete in regards to the operating periods of the disposal site and the general nature and amount of material that was buried at the disposal site. Specific information about the activity of the buried materials was much less abundant. A small number of samples taken during initial phases of the site's operation were used as the basis for all disposal calculations. Variations in the specific activity of both the feed material and the filtercake over the years of operations were not addressed in the historical data.

The historical documents show that the project health physicist in the 1960's was unable to explain radioactivity imbalances that existed between the extraction process feedstock radioactivity and the combined radioactivity of the product and waste materials. These should have been equivalent but they were not. The documents show that, of the thorium activity in the feedstock, 85% showed up in the filtercake (solid waste), about 400% showed up in the liquid waste, and almost 200% showed up in the extracted rare-earth product. For uranium, virtually no activity was noted in the filtercake, about 15% of the feedstock's activity was seen in liquid waste, and another 17% was seen in the product. Details of the analytical processes employed at the time were not provided in the historical data so the cause of the discrepancies has not been determined. The health physicist that noted the errors did recommend that samples be sent to the "National Reactor Testing Station" in Idaho where scientists had a lithium-drifted germanium detector. It is not known whether his recommendations were acted on.

SITE CHARACTERIZATION

To support the site's radiological evaluation, SCIENTECH NES implemented a characterization of the Breckenridge Disposal Site in 1997 in accordance with NUREG/CR-5849, "Manual for Conducting Radiological Surveys in Support of License Termination." The objective of the characterization was to determine the content and extent of radiological material at the site. To this end, several types of radiological measurement processes were employed including:

- Dose rate measurements at 1 meter above the surface
- Activity measurements at the surface
- Downhole activity measurements
- Soil sample collection and analysis (see Figure 2)

The soil sampling program called for a single, multi-depth sampling borehole located within every 10meter by 10-meter grid location on the site. The characterization activities verified that uranium and thorium did indeed exist at varying levels at the surface of and within the disposal site. The results of the 1-meter dose rate measurements showed that there was only a single location found to be reading more than 10 microroentgen per hour (microR/hr) above background. This location was reading 25 microR/hr greater than background.

Readings from the downhole sodium iodide crystal (2"x2") measurements in the affected area of the site ranged from background up to 30,000 counts per minute (cpm). Background was about 2,000 cpm. Elevated downhole readings were generally apparent where soil sample analysis also indicated elevated activity; however, since the distribution of the radioactive materials was not uniform, there was no consistent mathematical correlation between the two sets of data.



Figure 2. Conducting subsurface sampling at Breckenridge

Since the material was known to have been buried in trenches and the locations of the trenches were unknown, the soil sample collection and analyses resulted in highly variable data. Most of the 139 sample results from the affected area showed no activity above background levels. Thirty-two of the samples showed thorium activity significantly above background and 24 samples contained uranium activity significantly above background. Except for one outlier, the samples that were above background were in the range of 2-33 picocuries per gram (pCi/g) Th-232 and in the range of 3-83 pCi/g U-238. The one borehole location that was the outlier (in regards to activity) had elevated activity at all measured depths with a maximum at the 4'-6' level. At this depth, the borehole measured 1210 pCi/g Th-232 and 1218 pCi/g U-238.

As part of a previous characterization effort to locate the burial trenches, the disposal site was subjected to a ground-penetrating radar examination. The results of this effort were inconclusive. A deep disposal well was installed for non-radiological substances in 1967. The bore tailings created many anomalies in the ground-penetrating radar data, which provided no assistance in locating the buried radioactive material. In an additional effort to locate the burial trenches, aerial photos were examined and former employees were interviewed. The results of this effort also did not help draw substantial conclusions about the exact location and orientation of the burial trenches.

ANALYTICAL CHALLENGES

Modern-day sampling and analysis faced some of the same analytical challenges that were present more than 30 years ago. Initial sampling results from the modern-day characterization effort at the disposal site showed a very limited U-238 presence. A standard quality assurance protocol uncovered an analytical discrepancy and revealed that most of the uranium was initially missed. The low-energy and low-abundance peaks from U-238 daughters, Th-234 and Pa-234m, were masked by and rendered "unrecognizable" by the Compton continuum seen in samples, which contained substantial radioactivity. The original findings led to the belief that the uranium activity was indeed small as reported by the historical data. Reanalysis, however, indicated that there is about the same amount of uranium at the disposal site as there is thorium.

BURIED MATERIAL VOLUME FOUND TO BE SMALL

The historical data is believed to provide some accurate information on the quantity of material deposited in the disposal site. This data showed that the volume of filtercake deposited was very small, about 60 cubic meters. The modern-day sampling campaign defined the affected area of the site as a soil volume of approximately 7500 cubic meters. Consequently, less than 1% of the disposal site's volume contains the radioactive filtercake material. Since only a small fraction of the site's volume contains the radioactive filtercake, the regularly-spaced sampling locations in the characterization survey provided data that did not lend itself well to deriving a conclusion about the site's radioactive material inventory. As further explanation, the modern-day characterization effort including 139 analytical samples from the affected area revealed the following statistical data about total thorium: mean-13.54 pCi/g; median-0.35 pCi/g; and a standard deviation of 107.9 pCi/g. Statistical data for the uranium was very similar to the thorium.

ESTIMATING THE SITE'S RADIOACTIVE MATERIAL INVENTORY

Characterization of the disposal site revealed that about 7500 cubic meters (in an area of 3000 square meters) of the property contained elevated levels of radioactivity. Most of the measurements, however, were just slightly above background. Only a limited number of samples displayed significant radioactivity. This was because, as revealed by the historical site assessment, less than one percent of the material within this area was the actual filtercake material.

The recent characterization involved a multiple-depth sampling at regular intervals across the site. The results of this effort, if averaged over the affected area of the site, result in a derived thorium inventory that is about 3 times that noted in the historical data. The same approach indicates that the historical data underestimates the uranium inventory of the disposal site by several orders of magnitude. Historical values were taken directly from archived documents.

Another approach in deriving a source term is to assume that the one outlier in the sampling process was a "direct hit" of the filtercake and to further assume that the activity is generally consistent throughout the entire inventory of the 60 cubic meters of filtercake buried at the site. This approach may prove to be the most suitable since the historical analytical data for thorium compares reasonably well to the thorium value in the outlier borehole. A comparison of the approaches is provided below in Table I.

	From Historical Data	Average Values from the Characterization Data	Using Outlier as Average Activity of Filtercake
Thorium (µCi)	60,830	172,000	105,000
Uranium (µCi)	523	187,000	106,000

Table I.Radioactive Material Inventory Values using Different Approachesin microcuries (µCi)

DOSE ASSESSMENT

Since the area around the disposal site is used primarily for agriculture, the resident farmer scenario is appropriate in determining the potential future dose consequences of the site. The relatively insoluble filtercake material is fairly innocuous under a significant cover since crop uptake and the direct exposure pathways are insignificant. If substantial quantities of the material are at or near the surface, dose from these two pathways increases dramatically. Because of the filtercake's insoluble nature (did not dissolve

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in the extraction process' acidic solution), groundwater is impacted only slightly regardless of the depth of the contaminants. As noted previously, a limited amount of surface activity currently exists at the disposal site. This material is believed to have originated from spillage that occurred during site operations in the late 1960's. The pervasiveness of the surface activity will require further evaluation since it will largely determine the outcome of the dose assessment.

The heterogeneous nature of the material buried at Breckenridge also presents some challenges in conducting a dose assessment. Standard modeling codes traditionally use a homogenous mix of contaminants in a volume of soil. Since this is not the case at Breckenridge, alternative approaches for dose modeling are being evaluated. One approach is to consider spacing requirements in 10 CFR 20.304, which was used to control site burial activities. By combining the spacing requirements of the regulation and a postulated excavation volume for a home basement and foundation, an estimate can be made of the material that is likely to be brought to the surface.

MODERN STANDARDS FOR AN OLD SITE

Another unique issue associated with the Breckenridge Disposal Site revolves around the application of the current-day regulatory framework to a site that has been closed for 30 years. This site and others like it operated and closed under much more liberal requirements than those put forth in today's regulations. The NRC's new License Termination Rule (LTR) requires, for an unrestricted release, no more than 25 millirem per year of dose to the average member of the critical receptor group associated with a decommissioned site. This translates to only a few pCi/g of natural thorium at or near the surface using the resident farmer scenario.

Since the disposal site was previously closed and is no longer associated with a materials license, the applicability of the LTR is being carefully examined. Formerly closed disposal sites are under no obligation to automatically meet the LTR free-release criteria of 25 millirem per year. In fact, many of the formerly closed burial sites clearly would not meet the LTR's base criteria. However, if a formerly closed site is found to present an undue public risk or, in some other manner, is re-opened, the LTR does apply. It should be noted that the site is currently fenced and access is controlled. There is no immediate threat to any members of the public.

CONCLUSIONS

Deriving conclusions about the radioactive material buried at the Breckenridge Disposal Site has been a challenge. Many clues have surfaced about the site from historical data and site characterization efforts. The historical data provides necessary facts about operating dates and the general contents of the burial site. The same data leaves questions about the activity of the material and the accuracy of the analytical processes employed at the time. Current-day characterization data provides relatively accurate information about the specific activity of certain boreholes; however, it is difficult to gauge how well the boreholes represent the entire site. Additional subsurface characterization data is expected to be gathered shortly. Instead of random sampling, however, attempts will be made to retrieve direct samples of the filtercake material. Additional information about the specific activity of the specific activity of the filtercake, coupled with the volume information provided by the historical data, can be used to develop a defensible radioactive material inventory. Once a final determination is made on the radioactive material inventory, a dose assessment will be conducted. This will lead to a discussion regarding public risk and the site's disposition. The applicability of the License Termination Rule and what constitutes "acceptable" public risk are other issues that will also require resolution.