Applied Radiation and Isotopes 136 (2018) 143–149

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Forensic microanalysis of Manhattan Project legacy radioactive wastes in St. Louis, MO

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HIGHLIGHTS

- Scanning Electron Microscopy with Energy Dispersive X-ray analysis is used for the forensic examination of radioactive wastes.
- Radioactive microparticles containing either uranium or thorium as primary radioactive constituents are identified.
- Fugitive emissions of radioactive legacy Manhattan Project wastes are tracked from sources into area homes via dusts.

ARTICLE INFO

Keywords:
Radioactivity
Electron microscopy
Environmental analysis
Uranium
Thorium

ABSTRACT

Radioactive particulate matter (RPM) in St Louis, MO, area surface soils, house dusts and sediments was examined by scanning electron microscopy with energy dispersive X-ray analysis. Analyses found RPM containing ^{238}U and decay products (up to 46 wt%), and a distinct second form of RPM containing ^{230}Th and decay products (up to 15.6 wt%). The SEM-EDS analyses found similar RPM in Manhattan Project-era radioactive wastes and indoor dusts in surrounding homes.

1. Introduction

During the course of processing uranium between 1942 and 1957 for the production of nuclear weapons for the United States, the Mallinckrodt Chemical Works exceeded its storage space and transported approximately 133,000 metric tons (MT) of wastes and scrap by truck to the St. Louis, MO, Airport site (SLAPS), where it sat for decades in large piles exposed to the elements. The great majority of the wastes resulted from the extraction of uranium and other elements from high purity uranium ore obtained from a mine in the Belgian Congo (Kaltofen, 2015). In 1966, 116,700 MT of the wastes were sold to a private party and transported by truck to an interim storage site on Latty Avenue in nearby Hazelwood, MO. By 1973 the remaining 8000 MT of wastes and additional contaminated soil that had not yet been removed from the Hazelwood interim storage site (HISS) were disposed in the West Lake Landfill, a municipal waste landfill in Bridgeton, MO. The spread of these wastes from open air storage and spills during transport resulted in the contamination of approximately 626,000 m^3 of soil with uranium and its daughter products at levels exceeding DOE soil cleanup guidelines (DOE, 1992).

Processing at the Mallinckrodt plant resulted in secular disequilibrium and the subsequent high concentration of uranium daughter products compared to natural uranium in the wastes. Most notably, the concentration of ^{230}Th (58.1 TBq) in wastes disposed in the West Lake Municipal Landfill was found by the Nuclear Regulatory Commission (NRC, 1988) to be approximately 300 times higher than found in a typical uranium milling tailing pile (IAEA, 2008). Approximately 80% of the radioactivity from borehole measurements ($>$ 180 kBq g^{-1} taken of the wastes dumped in the West Lake Landfill) is due to ^{230}Th (Remedial Investigation Report, 2000).

One important component of the original waste materials, which is contained in the original Belgian Congolese and Bear Valley, Idaho ores, is thorium monazite, a thoriated rare earth- phosphate mineral feedstock to the Mallinckrodt Chemical Works process in St. Louis, MO. The documented presence of relatively-insoluble thorium monazite in wastes shipped to the Latty Avenue site made this an important constituent of concern for this study. The Bear Valley ore was known to contain about 0.3 kg of monazite per m^3 of ore. Monazite particles containing ^{230}Th were not bound to soil, and that ^{230}Th levels increased as particle sizes decreased (Porter et al., 1997). This waste material was

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https://doi.org/10.1016/j.apradiso.2018.02.020
Received 14 April 2017; Accepted 5 February 2018
Available online 22 February 2018
0969-8043/

also estimated to contain stable metals including 800 metric tons of cobalt, 950 metric tons of nickel, and 500 metric tons of copper (DOE, 1981) The process also used carnotite ores from Colorado (AEC, 1967).

Although the actual total waste quantity is unknown, calculations based on limited onsite soil sample analyses conducted by the NRC estimated that at least 170,000 metric tons of radioactive material in Areas 1 and 2 of the West Lake Landfill is almost entirely contaminated with naturally occurring ^{238}U and ^{230}Th and their progeny, although the daughters are not in equilibrium. The West Lake Landfill is a 81-hectare tract of land located in Bridgeton, Missouri, the near-suburbs of St. Louis, MO, (26 km northwest of downtown) and is a product of decontamination efforts at the Cotter Corporation's Latty Avenue plant, making it a potential source area for thorium-containing particulate matter, along with the Latty Avenue and Hazelwood Interim Storage Site, and the St. Louis, MO, Lambert Field Airport site.

Facilitated transport of radioactive materials occurs when radioactive contaminants are bound to soil and dust particles which are then transported through the environment. The transport of particulate matter containing uranium, thorium and radium was investigated in the Greater St. Louis, MO area, where wastes related to the 1940s-era Manhattan Project were disposed of.

2. Methods

Particulate matter in surface soils, house dusts, and sediments was examined by scanning electron microscopy with energy dispersive X-ray analysis (SEM/EDS). The study area included the Coldwater Creek watershed, a 114 km^2 area in suburban North St. Louis County, MO (Fig. 1); and homes and drainage channels leading away from the West Lake Landfill in Bridgeton, MO. Lower Coldwater Creek flows 22.4 km from Lambert Field International Airport to the Missouri River (MO-DNR, 2014). Buried radioactive wastes related to the former Manhattan atomic bomb project can be found at location on or near Coldwater Creek and the West Lake Landfill.

The study objective was to determine if radioactive material related to the St. Louis, MO, Mallinckrodt facility could be identified in both potential source areas (Latty, Avenue/Hazelwood, MO, Interim Storage site and St. Louis Airport site) and in receptor areas such as private homes and surface water runoff zones.

In a previous study of northern St. Louis County (Kaltofen, 2015), a representative number of samples was collected to examine potential pathways of radioactive contaminant migration at these same locations. The primary methods for detecting environmental radioactivity were NaI scintillation probes combined with HPGe confirmation, and Scanning Electron Microscopy/Energy Dispersive X-ray analysis (SEM/EDS). The last method, SEM/EDS, is not regularly used to detect radioactive materials because it can only distinguish between elements, and it does not distinguish isotopes of the same element (NERL, 2002).

In this study, SEM/EDS was used to detect and identify microscopic particulate matter (particle diameters < 150 μm) that facilitated transport of radioactive contaminants in the environment. To overcome the instrument's inability to distinguish among isotopes of the same element, SEM/EDS data was used solely for particulate matter containing elements that have only radioactive isotopes. These include thorium, radium, and uranium. These radioactive elements are all important constituents of wastes from the St. Louis, MO, Mallinckrodt facility.

The source area examined was the Latty, Avenue area in Hazelwood, MO. This area hosted the Hazelwood Interim Storage Site (HISS) and the nearby (0.75 km north) St. Louis Airport Site (SLAP). Both sites were used to store wastes from the Mallinckrodt Facility. The receptor sites were homes and other locations along Coldwater Creek in Hazelwood and Florissant, MO; and homes and drainage channels surrounding the West Lake Landfill in Bridgeton, MO. The West Lake Landfill received waste from the Mallinckrodt facility, and Coldwater Creek drains the HISS and SLAP sites. Samples of surface soils and sediments were collected from the source area sites and analyzed (n = 15 samples). These analytical results were compared to results for soils and interior house dusts (n = 23) at the Coldwater Creek/West Lake Landfill (n = 18) receptor locations.

The purpose of the SEM/EDS analysis was limited to determining the presence and form of particulate matter facilitating the transport of radioactive contaminants between source and receptor areas. All samples were analyzed by gamma spectroscopy, but only a subset of samples were analyzed by SEM/EDS. Four of fifteen source area samples with the highest total activity were elected for SEM/EDS analysis, because these would be most likely to contain significant amounts of potential source material. Four of twenty-three house dust samples were selected in the same fashion, as were four of eighteen Coldwater Creek/West lake Landfill-area soil and sediment samples. Twelve of the 56 total samples from this study were analyzed by SEM/EDS. This judgment-based sample selection was biased to maximize the probability of identifying radioactive particles that transport radioactivity from source areas to receptors. Three reference samples from a site outside of the St. Louis area and downstream on the Mississippi River were examined by SEM/EDS. These included one sample of house dust, one of river sediment and one surface soil sample. These were examined with the same instrument, analyst, and method as the twelve study area samples.

The Latty Avenue site survey was to collect and analyze surface soil samples for the presence of radioactively impacted materials, and to collect possible source material related to historic storage of radioactive materials at this site. This location was historically used to store radioactive wastes related to the Mallinckrodt site in St. Louis, MO. Any remaining waste materials still on the site would be collected and analyzed and compared to present-day samples from locations known to have accepted this radioactive waste. The Latty Avenue Site is located in northern St. Louis County in Hazelwood, Missouri. The site is not fenced or access controlled, nor there is no signage to indicate the presence of radioactive materials. Samples were selectively sampled based on field survey data collected via a Ludlum Model 2350 Data Logger calibrated with a 1′ × 1.5′ NaI probe connected with a 3″ cable. Survey personnel also carried three CsI scintillation dosimeters.

Sampling technicians surveyed areas between the Latty Avenue Site and Coldwater Creek, including areas near railroad spurs historically used for loading and shipping Mallinckrodt-related wastes. The operator held the detector approximately 1 foot above the ground surface and advanced along the areas of interest in straight lines at a rate of 1 m/s. In the area on the north side of the railroad tracks, radiation levels peaked at 117 K counts per minute, vs. an offsite background count rate of 3–5 K cpm. After clearing surface materials, counts peaked at 600–800 K cpm at about 20 cm below the surface. A composite sample was taken from 10 to 20 cm bgs. The sampled material was dark black and fine.

A control set of soil samples (n = 2) was collected from the La Sal uranium mine in southwestern Colorado using the same procedure described here. Likewise, radioactive particulate matter found in the control samples was similarly analyzed by SEM/EDS along with the study set samples. Control samples of house dust (n = 4) were collected from an area of known thorium contamination in Richland, WA; and also analyzed by SEM/EDS along with study samples.

Three samples of material from the area of elevated survey meter counts were collected. After samples were collected the removed overburden materials were back-filled into the sampled areas. The sample bags were stored in a secure location in a manner that maintained chain-of-custody requirements until they were shopped for off-site analytical laboratories. The whole samples were double bagged, and placed in a cooler and transported to Eberline Analytics of Oak Ridge, TN, a licensed and certified commercial radioisotope testing laboratory, for analysis of isotopic uranium (234, 235 and 238), ^{226}Ra, ^{210}Pb, and thorium (228, 230 and 232). Analyses proceeded by high purity germanium gamma detection and by alpha spectrometry, using