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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.

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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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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

Fig. 1. Coldwater creek sample locations.

methods EML U-02, EML Th-01, EPA 903.0 Ra and LANL ER- 130. An original chain-of-custody form was placed in the cooler for shipment. Prior to shipment the cooler was surveyed with a Ludlum Model 2350 data logger and 1′ × 1.5′ NaI and pancake probes to confirm compliance with 449 CFR part 173 for the transportation of exempt quantities of radioactive materials.

The eighteen Coldwater Creek/West Lake Landfill area surface soils and sediments were collected without the use of survey meters from the top 2–3 cm at each location. The twenty-three house dust samples (also unsurveyed) were collected from vacuum cleaner bags at each residence, along with one (per house) tape lift area sample (2 cm × 2.5 cm) collected via Zefon® Biotape® adhesive microscope slides. All samples collected were screened for total gamma activity using an Ortec® NaI scintillation detector and 1K MCA.

Subsamples were sent to Microvision Laboratories of Chelmsford, MA, for SEM/EDS analysis of non-radiological materials. These were prepared by subsampling approximately 100 mg of fine materials onto Zefon® Biotape® adhesive microscope slides. Analyses proceeded with a LEO/Brucher SEM/EDS system, using a lithium drifted silicon semiconductor X-ray detector. These subsamples were taken from four samples each from the potential source areas, Coldwater Creek, and house dust sample locations; all with the highest overall activity as measured by NaI gamma spectroscopy (twelve samples total).

3. Results

Gross radiochemical analyses: Gamma and alpha spectral analysis of two surface material samples from Latty Avenue yielded radioisotope activities that were multiple orders of magnitude above prior results (Kaltofen, 2015). The gross analyses of radionuclides for the two Latty Avenue samples was very similar to historical data showing the original waste material from the Mallinckrodt process, as sampled and tested at the Hazelwood Interim Storage Site (Levine, 1987).

The ^{230}Th activity in the 2016 samples at Latty Avenue is very similar in activity to the reported 1987 averages for the Latty Avenue waste pile. The 0 to − 18 cm bgs composite soil sample collected in 2016 had activities of 404,100 Bq kg^{-1} ^{230}Th, 15,390 Bq kg^{-1} ^{238}U, 8770 Bq kg^{-1} ^{226}Ra, and 2170 Bq kg^{-1} ^{232}Th. Background activities for these isotopes are on the order of 37–74 Bq kg^{-1} (DOE, 2001). By comparison, the 1987 Levine report noted that the average activities in the Latty Avenue waste pile were: 325,600 ± 7030 Bq kg^{-1} ^{230}Th, 2660 ± 333 Bq kg^{-1} ^{238}U, 2100 ± 37 Bq kg^{-1} ^{226}Ra, and 70 ± 15 Bq kg^{-1} ^{232}Th (Table 1).

In order to remove railroad ballast stones and other coarse materials from the sample, the second surficial sample from Latty Avenue was sieved to pass a #100 ASTM 150 µm opening standard brass sieve. Given that ballast stones would not be expected to contribute significantly to gross activities in the sample, sieving should typically yield a higher result for this sample than an in-situ sample. The sample activities were 891,000 ± 320,000 Bq kg^{-1} ^{230}Th, 19,100 ± 8900 Bq kg^{-1} ^{238}U, 17,400 ± 2430 Bq kg^{-1} ^{226}Ra, and 3650 ± 160 Bq kg^{-1} ^{232}Th (Table 1). The R^2 value between the 2016 Latty Avenue whole-sample isotopic data and the Latty Avenue radioactive waste as described in Levine (1987) is 0.9998. The 2016 Latty Avenue sample likely is a remnant of the original uranium processing waste stored at the HISS before 1987, and described in Levine (1987). Given the close match in isotopic thorium composition and in activities between the 2016 Latty Avenue samples and the original HISS waste pile material, the two Latty Avenue samples were used as source materials for the next study phase, the forensic analyses of soils, dusts and sediments.

An indoor dust sample was collected from the home closest (< 700 m) to the West Lake Landfill. The house dust had the following gross analytical results: 37,300 ± 28 Bq kg^{-1} ^{230}Th, 514 ± 14 Bq kg^{-1} ^{238}U, 1780 ± 816 Bq kg^{-1} ^{226}Ra, and 970 ± 112 Bq kg^{-1} ^{210}Pb. The R^2 value between the house dust sample isotopic data and the Latty Avenue uranium-processing waste as described in Levine (1987) is 0.9990. The dust sample was collected from basement dusts found above structural beams that would normally be inaccessible to residents. It was assumed that this dust represented a long term accumulation, rather than current living-space dusts. It is reasonable to conclude that the house dust and the material detected in 2016 at Latty Avenue are both contaminated with material similar to the uranium processing wastes described in Levine (1987).

The isotopic analyses for ^{230}Th, ^{238}U, ^{226}Ra, and ^{210}Pb were compared for the 2016 Latty Avenue source materials and offsite uranium-milling related dust emissions (Table 1) (Rood et al., 2008). The R^2 values for the Latty Avenue material and milling emissions were: yellow cake dusts $R^2 = 0.07$, tailing pile dusts $R^2 = 0.06$, ore haul-road dusts $R^2 = 0.74$, and milling dusts $R^2 = 0.10$. The correlation coefficients among the St. Louis area materials are significantly greater than between these materials and generic uranium processing dusts.

Forensic SEM/EDS analyses of the source area samples showed that the high-activity Latty Avenue samples contained particles composed of percent levels of uranium as the primary radioactive element (Fig. 2). The uranium-type particles detected contained from 20.2% to 46% by weight uranium, along with vanadium, oxygen, cobalt, copper and

Table 1
Isotopic activities in soils and dusts.

Data in Bq kg^{-1}	^{238}U	^{230}Th	^{226}Ra	^{210}Pb
Latty Avenue (HISS) @ Coldwater Creek	15,390	404,100	8770	962
Latty Avenue (HISS) @ Coldwater Creek (fines)	19,100	891,000	17,400	No data
Airport (SLAPS) @ Coldwater Creek	40.7	511	No data	No data
Airport (SLAPS) @ Coldwater Creek	52	265	93	ND
House dust (Bridgeton, MO)	514	37,300	1780	970
House dust (Bridgeton, MO)	ND	15	96	4475
House dust (Bridgeton, MO)	ND	512	ND	144
Indoor dust (Bridgeton, MO)	10	ND	ND	636
Background data				
Regional soil background (DOE, 2001)	37	37	37	74
Regional soil background (Kaltofen, 2015)	16	23	25	< 80
Mallinckrodt waste data				
Average of 5 DOE Latty Ave. soils (1986)	No data	31,850	1180	No data
Latty Avenue waste (Levine, 1987)	2660	325,600	2100	No data
SLAPS (Levine, 1987)	1220	33,300	376	No data
Average of Cotter Site (Levine, 1987)	2260	155,400	4070	No data
Reference data as relative activity in Bq[a]				
Yellow cake uranium (fugitive emissions)	41,600	3830	66	2960
Tailing pile fugitive dusts	229	13,400	15,830	55,210
Truck drag-out (ore haul-road dusts)	616	804	562	689
Milling dust emissions	3080	3110	3110	3110

[a] Rood et al. (2008).

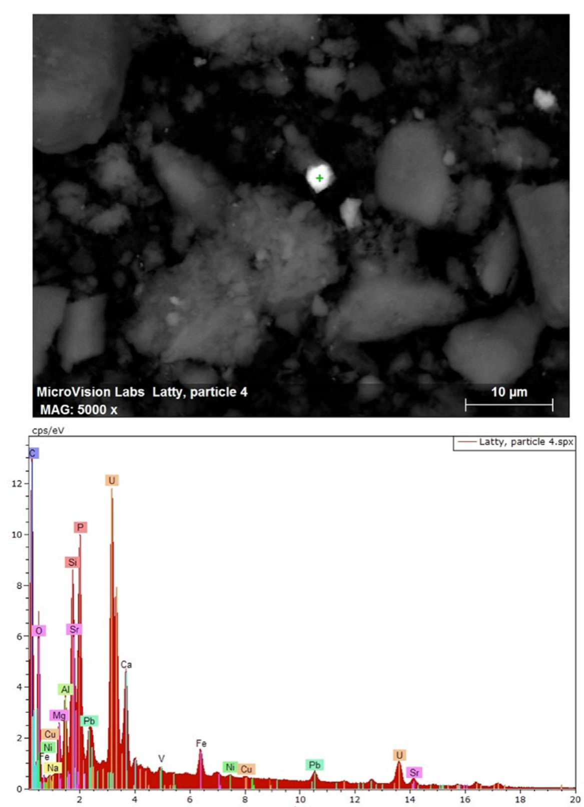

Fig. 2. Mallinckrodt uranium processing waste particle from Latty Avenue railroad siding, Hazelwood, MO. (Top) photomicrograph, (Bottom) SEM/EDS spectrum.

nickel. The presence of copper, cobalt and nickel in the uranium particles is consistent with studies showing that the Mallinckrodt waste at Latty Avenue contained nearly 2000 metric tons of copper, cobalt and nickel metals. Vanadium is a common component of uranium ore (DOE, 1981). The presence of these metals known to be in the original Mallinckrodt material strengthens the conclusion that these particles collected in 2016 are related to the original Shinklobwe mine material.

For uranium-type particles, copper composition ranged from 0.11% to 1.4%, cobalt ranged from not detected to 2.7%, nickel ranged from 0.1% to 1.7% and vanadium ranged from 0.10% to 2.1%. One uranium-type particle also contained 0.66% polonium. In the control samples collected from the La Sal uranium mine in southwestern, CO, radioactive particles identified by SEM/EDS contained a composition that was distinct from that found at Latty Avenue. Vanadium in control particles ranged from 2% to 44%. Copper was not detectable in 75% of control particles, and copper did not exceed 0.2% in any control particle. Cobalt and nickel were absent from all radioactive particles in the control set from the La Sal mine.

A set of particles containing thorium as the primary radioactive element was also detected in the Latty Avenue samples. These particles had the form of thorium-monazites, with traces of copper, cobalt and nickel (Fig. 3). The thorium-type particles contained 2.5–9.8% thorium by weight. Radium and polonium were not detected in the thorium-type particles. For these particles, copper composition ranged from not detected to 0.14%, cobalt ranged from not detected to 0.21% and nickel ranged from not detected to 0.21%. Thorium monazite is the primary radioactive material found in the Congolese ore raffinate. Raffinate is the type of industrial waste originally stored at Latty Avenue, and now known to be in the West Lake Landfill (NRC, 1988).

Radium was not detected in the uranium or thorium-type particles, nor was it seen in any other particle or sample examined by SEM/EDS. Although gross radiochemical analyses of Latty Avenue samples found significant radium, its activity was only a small fraction of the thorium detected. Further SEM/EDS investigations with larger sample sizes may be needed to detect radium (if present) in Latty Avenue particulate matter.

Samples of Coldwater Creek sediments collected from the point where the creek passes under a Lambert Field Airport access road (McDonnell Blvd.) also contained thorium monazite particles detectable by SEM/EDS. This location is 1.25 km upstream of the Latty Avenue sample site. The gross radiochemical analyses of the Airport site samples found ^{230}Th at $511\,Bq\,kg^{-1} \pm 95\,Bq\,kg^{-1}$, vs. ^{232}Th at $38\,Bq\,kg^{-1} \pm 14\,Bq\,kg^{-1}$. This suggests that the thorium at this site is likely due to the presence of Mallinckrodt wastes, as the 230 isotope of thorium predominates in the wastes compared to natural abundances, where the 232 isotope is more common (Table 1) (Levine, 1987).

All isotopes of uranium and thorium are radioactive. SEM/EDS, which does not distinguish between isotopes of the same element, can be used to determine if a given particle of is radioactive based on detection of uranium, thorium, radium or other entirely radioactive elements. SEM/EDS data for the Latty Avenue samples found uranium and thorium-bearing particles which also contained strontium (up to 2.1%) and cesium (up to 0.32%). Although these elements are often associated with fission wastes, the SEM/EDS analysis does not provide any evidence that the cesium or strontium detected was radioactive. Gross radiochemical analyses of these samples did not detect the common radioactive isotopes of cesium (^{134}Cs and ^{137}Cs). The activity of ^{90}Sr detected was $36\,Bq\,kg^{-1}$ vs. a typical background level at this latitude of $9\,Bq\,kg^{-1}$ (calculated from Wallo, 1994). The activity of ^{90}Sr was significantly below the activities detected for ^{230}Th and ^{226}Ra.

No particles containing SEM/EDS-detectable uranium, thorium, radium or other definitively-radioactive elements were detected in the three reference samples.

The aliquot of Latty Avenue sample used for the SEM/EDS analysis had a mass of 25 mg, and contained eight apparently-radioactive particles ranging in size from 2 to 14 µm. Given the small sample size, the SEM/EDS data are assumed to be examples of Mallinckrodt waste material, but are not necessarily representative of the waste as a whole. These data from Latty Avenue provide a semi-quantitative standard material allowing for further studies of radioactive particles in waste-impacted areas, such as homes located near known waste sites in the St. Louis area.

SEM/EDS data for house dust samples: SEM/EDS analyses showed that samples of house dusts and soils in the St. Louis area contained the same uranium-type and thorium-type particles that were detected within the Latty Avenue and St. Louis Airport Site source material samples of Mallinckrodt-related wastes.

Four of the four homes tested (100%) contained thorium-type particles in dusts. One of four (25%) contained uranium-type particles. Both types of particles found in the homes had elemental compositions similar to the Latty Avenue source material samples. Microanalysis of dusts from a home 0.75 km from the West lake Landfill contained both types of radioactive particles. The uranium and thorium-bearing particles in this home also contained traces of copper, nickel, cobalt and/or vanadium (Fig. 4). The house dust particle with the highest uranium concentration (15.7% U by weight and 6 µm in avg. diameter) contained 0.49% copper, 0.30% nickel, 0.02% cobalt and 0.06% vanadium. Four of five of these metals concentration values lie approximately in the middle of the ranges found for uranium-type particles at the source area. The vanadium concentration is close to the low range found at the source area. Of the thorium-type particles in the same house dust, the values from the highest concentration thorium particle (3.1% by weight and 6 µm in diameter) were: copper 0.80%, nickel 0.30%, cobalt 0.01%

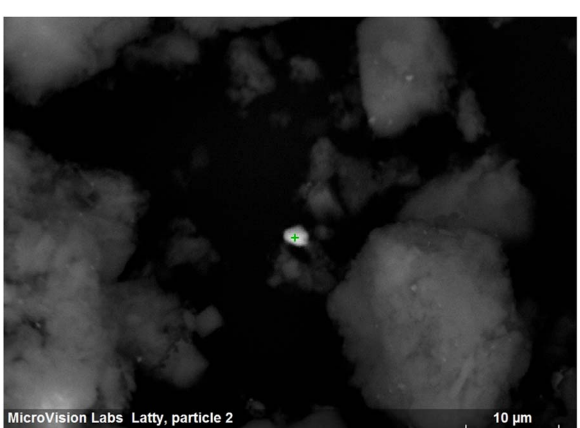

Fig. 3. Latty Avenue composite sample - Thorium monazite particle with copper, cobalt and nickel.

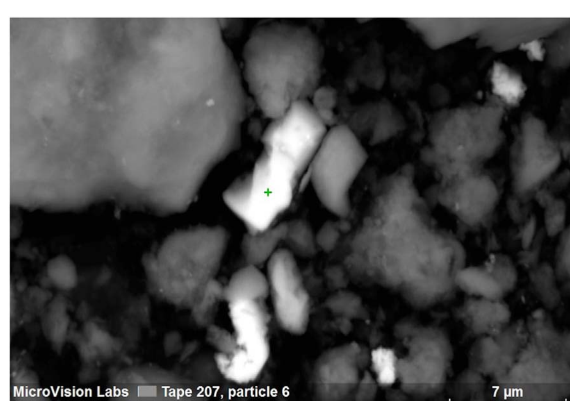

Fig. 4. Interior house dust particle, West Lake Landfill neighborhood, "House 1", Bridgeton, MO.

Table 2
Radioactively-hot particles detected.

Source Area - Uranium & Thorium RPM	U %	Th %	Co %	Cu %	Ni %	V %
Latty Avenue @ Coldwater Creek (Fig. 2)	20.2	–	–	0.1	0.1	0.2
Latty Avenue @ Coldwater Creek	36	–	1.5	1.4	1.7	0.27
Latty Avenue @ Coldwater Creek	46.4	–	–	0.34	0.11	0.10
Latty Avenue @ Coldwater Creek	4.15	–	0.37	0.24	0.13	2.12
Latty Avenue @ Coldwater Creek	1.6	–	0.85	0.96	0.42	7.20
Latty Avenue @ Coldwater Creek (Fig. 3)	–	2.5	0.21	0.14	0.21	–
Latty Avenue @ Coldwater Creek	–	–	2.7	6.3	0.69	9.6
Latty Avenue @ Coldwater Creek	–	9.8	–	–	–	–
Airport Site @ Coldwater Creek	–	2.72	–	–	–	–
Bridgeton - Uranium & Thorium RPM						
Indoor house dust (Fig. 4)	15.7	–	0.02	0.5	0.3	0.1
Indoor house dust	3.07	12.0	–	1.82	–	–
Indoor house dust	–	3.1	0.01	0.81	0.30	5.2
Indoor house dust	–	1.25	–	0.53	–	–
Indoor house dust	–	3.95	–	0.44	–	–
Indoor house dust	–	2.8	–	0.44	–	–
Indoor house dust	–	2.4	–	–	–	–
Indoor house dust	–	0.99	–	–	–	–
Indoor house dust	–	2.7	–	–	–	–
Bridgeton Trails - sediments	–	3.8	–	0.15	–	–
Coldwater Creek - Thorium RPM						
Indoor dust (home)	–	3.09	–	0.26	0.05	–
Indoor dust (home)	–	4.6	–	0.19	–	–
Indoor dust (home)	–	2.08	–	0.08	–	–
Indoor dust (home)	–	9.1	–	0.32	–	–
Coldwater Creek sediments	–	3.10	–	0.26	–	–
Coldwater Creek sediments		2.34				
Coldwater Creek sediments		2.72				
Coldwater Creek sediments		5.71				

and vanadium 5.21%. In this home near the West Lake Landfill, 10 uranium or thorium particles were detected in 50 mg of dust examined by SEM/EDS.

Despite the percent levels of uranium and thorium found in individual particles in the West Lake Landfill-area home, the gross radiochemical activities of this home's living-space dust were within ranges found in previous studies (Kaltofen, 2015). The activity of ^{210}Pb was 171 Bq kg^{-1} ± 13 Bq kg^{-1}, ^{226}Ra was 42 Bq kg^{-1} ± 7 Bq kg^{-1}, and ^{230}Th was 26 Bq kg^{-1} ± 5 Bq kg^{-1}. This data suggests that the uranium and thorium activity in the house dust is found in the form of discrete radioactively-contaminated particles that originated with the Mallinckrodt uranium-processing wastes.

The three remaining homes tested by SEM/EDS were located in the Coldwater Creek floodplain near St. Cin Park, Wedgewood Neighborhood Park, and the Florissant Golf Club. All three samples of house dust contained thorium-type particles. The St. Cin Park home had one detectable radioactive particle (9.1% thorium by weight and 11 µm in average diameter) in a 25 mg sample. The activity of this particle can be calculated from the specific activity of thorium (ANL, 2005). At this size and composition, a particle of 9.1% ^{230}Th would have an activity of 11 Bq, while for 9.1% ^{232}Th the particle activity would be a much smaller 0.06 mBq.

The Wedgewood Neighborhood Park home had three detectable radioactive particles in a 25 mg sample (up to 4.7% thorium by weight and 3 µm in average diameter). The Florissant Golf Club home had one detectable radioactive particle in a 25 mg sample (3.1% thorium by weight and 8 µm in average diameter). No detectable particles containing uranium, thorium, vanadium, or cobalt were found in the reference samples.

SEM/EDS analysis of soil and sediment samples: Four of four (100%) samples of sediments and soils collected from the Coldwater Creek/West Lake Landfill areas each had positive SEM/EDS detections for thorium monazite particles. Two samples came from the Duchesne Park section of Coldwater Creek in Florissant, MO. Both of these were positive for thorium-type particles. Both samples had three detectable thorium-type particles per 50 mg subsample, with thorium found at 2.1–3.1% by weight. Sizes for thorium-type particles at these locations ranged from 6 to 12 µm. No uranium-type or radium-containing particles were found at this location.

Two samples came from drainage channels at the Bridgeton Recreation Area near the West Lake Landfill. Both of these were positive for thorium-type particles. The combined samples had three detectable thorium-type particles in a 100 mg subsample, with thorium found at up to 3.8% by weight. Sizes for thorium-type particles at these locations ranged from 20 to 30 µm. No uranium-type or radium-containing particles were found at this location. No cobalt or nickel was detected in the thorium-type particles from these soil and sediment samples, but copper was found at up to 0.15% in thorium-type particles in the sediments (Table 2).

4. Conclusions

Gross radiochemical analyses established a strong link between historic data on Mallinckrodt uranium processing wastes, legacy waste-contaminated soils still found near the HISS location, and dust accumulations in a home near the West Lake Landfill, a repository for wastes from the HISS location.

X-ray microanalysis of legacy wastes related to the St. Louis Mallinckrodt uranium processing facility found two distinct types of radioactive particulate matter. One type was particulate matter containing a high percentage of uranium. The second type contained thorium and rare earths associated with monazite-type minerals. Both types contained the metals cobalt, copper, nickel and vanadium. These are associated with the raw ores and radioactive dross remaining after uranium has been extracted for the production of uranium metal billets.

Microscopic radioactive particles bearing the distinctive fingerprints of raw uranium ores and of thoriated dross were found in source areas, creek sediments and drainage channel sediments, and in public parks and residences that were in the Coldwater Creek and West Lake Landfill floodplains.

The evidentiary link between the Mallinckrodt uranium-processing wastes and local house dusts was established by gross radiochemical activities and SEM/EDS data for the source and receptor area samples. SEM/EDS identification of matching radioactive particles at the source and at the receptor provided a fingerprint distinguishing natural and industrial radioactive contaminants.

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