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# Tracking legacy radionuclides in St. Louis, Missouri, via unsupported $^{210}Pb$

Marco P.J. Kaltofen [a, *], Robert Alvarez [b], Lucas Hixson [c]

[a] Boston Chemical Data Corp., 2 Summer Street, Suite 14, Natick, MA 01760, USA
[b] Institute for Policy Studies, 1112 16th Street, NW, Suite 600, Washington, DC 20036, USA
[c] 1515 ½, Kyle Street, Eaton Rapids, MI 48827, USA

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Analysis of 287 soil, sediment and house dust samples collected in a 200 $km^2$-zone in northern St. Louis County, Missouri, establish that offsite migration of radiological contaminants from Manhattan Project-era uranium processing wastes has occurred in this populated area. Specifically, 48% of samples (111 of a subset of 229 soils and sediments tested) had $^{210}Pb$ concentrations above the risk-based soil cleanup limits for residential farming established by the US Department of Energy at the Fernald, OH, uranium plant, which handled and stored the same concentrated Manhattan Project-era wastes; the geographical distribution of the exceedances are consistent with water and radon gas releases from a landfill and related sites used to store and dispose of legacy uranium wastes; and offsite soil and house dust samples proximal to the landfill showed distinctive secular disequilibrium among uranium and its progeny indicative of uranium ore processing wastes. The secular disequilibrium of uranium progeny in the environment provides an important method for distinguishing natural uranium from industrial uranium wastes. In this study, the detection of unsupported $^{210}Pb$ beyond expected atmospheric deposition rates is examined as a possible indicator of excessive radon emissions from buried uranium and radium-containing wastes.

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## 1. Introduction

Legacy nuclear processing wastes, many dating back to the Manhattan Project, are a serious problem at numerous sites in the United States. At least 90 legacy management and sites are situated in 29 states, many of which have required extensive cleanup efforts.

The St. Louis, MO, region legacy sites hosted war-time uranium ore processing activities that provided material for Enrico Fermi's first nuclear pile, the Manhattan Project, and the Cold War. Uranium ore processing wastes from the Mallinckrodt plant in St. Louis, were disposed of around Greater St. Louis, MO, and are being redistributed by surface waters and by winds. Public reports (NIOSH, 2010) document a convoluted history of waste generation, transportation, and temporary storage followed by commercial sales of wastes, reprocessing and disposal in a municipal landfill. No detailed waste characterization or inventory exists (Criss, 2013).

The West Lake Landfill (Bridgeton, MO) is one of the last sites in the chain of facilities used for the disposal of St. Louis Legacy wastes. Although the West Lake Landfill contains significant amounts of long-lived radiotoxic wastes such as those contained in federally licensed commercial radioactive waste landfills, it meets virtually none of the legal requirements governing shallow radioactive waste disposal to prevent offsite migration (NRC, 2015a).

## 2. Site description

The West Lake Landfill is an 81-ha tract of land located in Bridgeton, MO, the near-suburbs of St. Louis, MO, 25 km northwest

Acronyms and abbreviations: AEC, U.S. Atomic Energy Commission; CMM, Continental Mining & Milling Company; COC, Contaminant of concern; COPC, Contaminant of potential concern; CWC, Coldwater Creek; DOE, U.S. Department of Energy; EPA, U.S. Environmental Protection Agency; FSP, Field Sampling Plan; HISS, Hazelwood Interim Storage Site; MCW, Mallinckrodt Chemical Works; MDNR, Missouri Department of Natural Resources; MED, Manhattan Engineer District; NRC, U.S. Nuclear Regulatory Commission; ORAU, Oak Ridge Associated Universities; QAP, Quality Assurance Plan; RMC, Radiation Management Corporation; ROD, Record of Decision; SAP, Sampling and Analysis Plan; SLAPS, St. Louis Airport Site; SLDS, St. Louis; MO, Downtown Site; UMC, University of Missouri-Columbia; USACE, U.S. Army Corps of Engineers; VP, Vicinity Property.

* Corresponding author.
E-mail addresses: marco@bostonchemicaldata.com (M.P.J. Kaltofen), bob@ips-dc.org (R. Alvarez), lhixson@environserve.com (L. Hixson).

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of downtown. This unlined landfill is located in an alluvial floodplain 1.6 km from the Missouri River. It has an ongoing major proximal subsurface fire e the 2nd fire in the past 21 years (EPA, 1995) (Bridjes, 2013). The NRC has estimated that at least 170,000 metric tons of radioactive material in Areas 1 and 2 of the West Lake Landfill is contaminated with naturally occurring $^{238}U$ and $^{230}Th$ and their progeny, and that these progeny are in secular disequilibrium with their $^{238}U$ parent (McLaren-Hart, 1996a; NRC, 1988).

Mallinckrodt's former uranium processing plant is on the west bank of the Mississippi River, fifteen km east of the primary study area, in the northern part of Downtown St. Louis, MO (Fig. 1). Former temporary disposal sites are at Lambert Field International Airport and Latty Avenue; 18 and 19 km to the northwest of the downtown site, respectively.

The receiving water body for runoff from the West Lake Landfill is the Missouri River upstream of its confluence with the Mississippi River, via engineered drainage canals. The receiving water body for the Airport and Latty Avenue sites is Coldwater Creek. The Coldwater Creek floodplain downstream of these sites is heavily developed for residential and commercial uses.

These sites together encompass an area of approximately 200 $km^2$ in northern St. Louis County, MO, in the Midwestern United States.

## 3. Site history

Between 1942 and 1957, Mallinckrodt Chemical Works processed uranium ore in downtown St. Louis, MO, removing radium, rare earths and other impurities. This uranium metal production was done under contract with the Manhattan Engineering District (MED) and the U.S. Atomic Energy Commission (AEC) (NIOSH, 2005). The Mallinckrodt plant processed approximately 50,000 tons of uranium ore. About 20,000 tons of this total was highly-concentrated uranium ore from the Shinkolobwe mine in Africa (WNA, 2012). Mallinckridt also produced $UO_2$, $U_3O_8$ and $UO_2F_2$ from $UF_4$; and produced $^{230}Th$ from raffinate (Hewlett and Duncan, 1972).

By 1946 the St. Louis, MO, Lambert Field airport disposal site was being used to store raffinate (dross), slag, drummed wastes and uranium scrap. In total, an estimated 133,007 tons of process waste residues and scrap were stored at the St. Louis Airport Site (Mason, 1977).

In March 1962, the AEC proposed the sale of the waste residues at the St. Louis Airport Site. The wastes were purchased by a reclaimer, who then shipped processed wastes to Canon City, Colorado in 1966 and 1967. A total 116,700 tons of the airport wastes were also transported to open-air storage at the Latty Avenue property in Hazelwood, MO (NIOSH, 2010). Much of the residual waste materials from the airport site were then removed and disposed of at an AEC-operated quarry in Weldon Spring, MO, in 1969 (Army Corps, 2002). In 1973, an estimated 8700 tons of leached barium sulfate mixed with 39,000 tons of top-soil from the Latty Avenue site were disposed of at the West Lake Landfill (NRC, 1994).

The landfill was not designed or licensed to receive radioactive waste materials. The full inventory of radioactive materials dumped has not been adequately characterized. The chemical and physical nature of the contaminated materials, along with their exact locations, is still unknown. The Nuclear Regulatory Commission and the U.S. Department of Energy determined that two areas in the landfill contained high-levels of uranium decay products e with particular concerns over the long-term hazards associated with $^{230}Th$ and $^{226}Ra$ (NRC, 1982) (NRC, 1994) (DOE, 1979). The presence of $^{226}Ra$ raises concerns about release of its gaseous decay product $^{222}Rn$ from the soil, leading to the offsite migration of $^{222}Rn$ and radon decay products such as $^{210}Pb$. The DOE concluded in 1993 that approximately 90% of the radiological risks to future residents exposed to residual processing wastes from the Mallinckrodt plant is attributable to $^{210}Pb$ (DOE, 1993).

The disposition of the uranium processing wastes in the landfill is regulated under the Uranium Mill Tailings Radiation Control Act of 1978 (EPA, 2015), although EPA notes that soil concentrations of $^{226}Ra$ and $^{230}Th$ in the West Lake Landfill are substantially greater than typical uranium mill tailing piles (EPA, 1993) (EPA, 2008). Data reported by EPA in 1996 indicates that radon emissions were in significant excess of the DOE limit of 3.7 Bq $l^{-1}$ (100 pCi $l^{-1}$) for the interim storage of residual radioactive materials (McLaren- Hart 1996b).

Analyses conducted by the NRC concluded that the levels of $^{226}Ra$ contamination are reasonably constant in the landfill materials (NRC, 1982). Any $^{226}Ra$ in soil will eventually decay to $^{210}Pb$. This is the supported $^{210}Pb$, or $^{210}Pb$ in equilibrium with its parent $^{226}Ra$. For gas-phase $^{222}Rn$ moving freely in the environment, the resulting $^{210}Pb$ is unsupported, being physically separated from its solid-phase parent $^{226}Ra$. Over time unsupported lead will decay away as there is no parent $^{226}Ra$ to maintain it. Supported lead will remain at its equilibrium concentration, as it is constantly replenished by decay of $^{226}Ra$. (Clark et al., 1987).

The NRC calculated that the $^{226}Ra$ activity at the West Lake Landfill, and thus the $^{222}Rn$ gas flux, would increase by a factor of five in 100 years, a factor of nine in 200 years, and a factor of thirty-five 1000 years from now (as cited by Criss, 2013). This increase is due to the continuing decay of parent isotopes into radium and radon. The maximum $^{222}Rn$ emission calculated was 228 Bq $l^{-1}$, about 78 times the DOE limit for the interim storage of residual radioactive materials (DOE, 1987). In 1993 a US DOE study confirmed that unsupported $^{210}Pb$ was mobilized via the Mallinckrodt wastes, finding 3.08 Bq $g^{-1}$ of $^{210}Pb$ but only 1.81 Bq $g^{-1}$ of $^{226}Ra$ (DOE, 1993).

The DOE and the COE have adopted activity limits for the remediation of similar sites in the Ohio cities of Fernald and Dayton. The Fernald, OH, plant accepted wastes generated at the St. Louis, MO, Mallinckrodt plant. Significant amounts of radon were released from these wastes at Fernald (NAS, 1997). At Fernald a remediation level in soil of 74 Bq $kg^{-1}$ (2 pCi $g^{-1}$) for $^{210}Pb$ was established for "off property" areas, based on soil ingestion, dust inhalation, and external gamma exposure (EPA, 1996). The background level for $^{210}Pb$ at Fernald is 57 Bq $kg^{-1}$, and is set at 38.8 Bq $kg^{-1}$ for this study area (DOE, 2001).

## 4. Methods

Samples (n ¼ 287) were collected from soils and drainage channel sediments (n ¼ 266) and indoor dusts (n ¼ 21). These were collected from areas surrounding West Lake Landfill, in Bridgeton, MO, the original airport disposal site at Lambert Field, Latty Avenue, in Hazelwood, MO, the Mallinckrodt site in St. Louis, and in parks and homes along Coldwater Creek (Hazelwood and Florissant, MO). The samples from homes abutting Coldwater Creek included both soils and bulk house dust samples collected from vacuum cleaner bags (Kaltofen, 2015).

Areas sampled were predominantly drainage channels and engineered waterways, along with surface soils in areas subject to flooding and rainfall runoff. Control sites were more than 3 km from the landfill or disposal sites. Geologic background samples of loess deposits provided a record of natural activity in regional subsurface samples (Lippmann, 2010).

A visual survey of each area was conducted to facilitate collecting native soils and sediments, and avoiding anthropogenic fill materials. Screening analyses were performed after sampling and

Fig. 1. Map of St. Louis County, Missouri with locator inset map showing the State of Missouri in the US. Highlighted areas of concern are the Downtown St. Louis, MO, Mallinckrodt uranium-processing site, West Lake Landfill, Airport Storage Site and Latty Avenue, in Hazelwood, MO.

prior to shipping using a GammaPal® gamma analysis system with a 2″x2″ NaI crystal and 4K channel MCA. Calibrations were by NIST-traceable $^{40}$K, natural uranium and $^{137}$Cs sources.

Bulk dust and soil samples were air dried at ambient temperatures prior to analyses. Samples containing macroscopic objects, gravel or excessive biological materials were sieved to pass a 150-micron brass ASTM #100 screen. Samples were placed in Marinelli® counting containers or equivalent plastic wells for counting with standardized geometry.

For gross alpha and gross beta analyses, air dried samples were screened to <150 microns and were tested for gross alpha activity on a Ludlum Model 3030 counter. Thin layer (2 mm) samples were counted for 1 h on a known-area well-type planchette. Counts are reported per unit soil area as self-absorption prevents reporting on a mass basis. This semi-quantitative analysis provided only empirical comparative data between samples in this specific study.

Gamma photon analyses used Ortech® NaI well and flat cylinder type detectors. Count efficiency @ 662 keV was 0.30 for the Ortech® NaI well detector with a Spectech® 2K MCA. Samples were standardized against identically-prepared soils of known activities, as well as known commercial $^{226}$Ra and $^{137}$Cs standards. All samples and standards were normalized to 8.0 g dry wgt.

In situ and jar-test radon soil vapor samples were performed using Accustar® alpha track AT-100 long term radon sampling canisters. The canisters use dosimetry grade CR-39 resin-based alpha track targets. Analyses were performed by Accustar®, Inc. of Medway, MA. For jar tests, dry high activity soils and background soils (approximately 30 g weighed to ± 0.01 g) were sealed for 90 days in a 500 ml polyethylene jars along with an alpha-tracking canister. These were reported as $^{222}$Rn in Bq g $^{1}$ soil. In-situ field testers buried around the West Lake Landfill equilibrated for up to 180 days. The in-situ canisters measured soil gas radon in Bq of $^{222}$Rn per equivalent m$^{3}$ of soil gas or indoor air.

Two West Lake Landfill soils and one Latty Avenue soil were analyzed by scanning electron microscopy with energy dispersive X-ray detection (SEM/EDS) at Microvision Laboratories of N. Billerica, MA. Analyses proceeded with a LEO/Brucher SEM/EDS system, using a lithium drifted silicon semiconductor X-ray detector.

Analyses for isotopic uranium (234, 235 and 238), thorium (228, 230 and 232), $^{210}$Pb and $^{226}$Ra in duplicate soil samples (n ¼ 25) and house dusts (n ¼ 21) were performed by a certified and licensed commercial laboratory, Eberline Laboratories of Oak Ridge, TN. Analyses proceeded by high purity germanium gamma detection, using methods EML U-02, EML Th-01, EPA 903.0 Ra and LANL ER-130.

For individual counts, the value of s (standard deviation) was calculated as the square root of the total counts, with n ¼ 1 for all samples. For sets of samples s is given as the square root of the variance.

## 5. Results

Of the 287 samples collected, 229 soils and sediments were analyzed for $^{210}$Pb. Some samples were not analyzed for $^{210}$Pb due to sample mass or laboratory capacity constraints. Of the 229 samples, 48% (111 samples) contained levels of $^{210}$Pb in excess of the 74 Bq kg $^{1}$ DOE (Fernald, OH) soil guideline (Fig. 2). A total of 22% (51 samples) had no detectable $^{210}$Pb with a detection limit of 37 Bq kg $^{1}$. The mean $^{210}$Pb activity for study area soils was 54 Bq kg $^{1}$ (s ¼ 39.5 Bq kg $^{1}$, averaging non detects as 0), and the

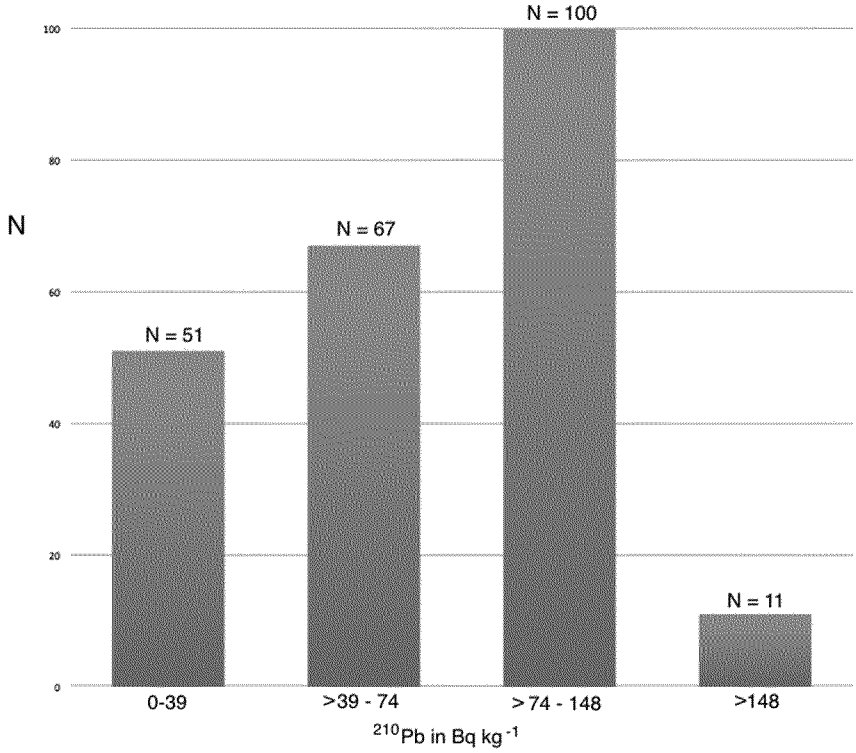


Fig. 2. Number of soils with $^{210}$Pb activities exceeding background (39 Bq kg $^{1}$) or exceeding DOE Soil cleanup guideline (74 Bq kg $^{1}$) (EPA, 1996).

Table 1
Bq kg $^{1}$ of $^{210}$Pb and $^{226}$Ra in ten highest activity soil samples for each isotope. The number of s (standard deviations) above the mean values are noted. For $^{210}$Pb the mean activity was 54 (s ¼ 39.5) Bq kg $^{1}$. For $^{226}$Ra the mean activity was 14.05 (s ¼ 7.02) Bq kg $^{1}$.

| Area (ID) | $^{210}$Pb | s above mean | Area (ID) | $^{226}$Ra | s above mean |
|---|---|---|---|---|---|
| West Lake (BT8) | 350 | 7 | STL Airport (2) | 67.0 | 8 |
| West Lake (BMAC4) | 299 | 6 | Latty Ave. (2) | 59.7 | 7 |
| West Lake (BT4) | 267 | 5 | Coldwater Cr. (F1) | 43.5 | 4 |
| Coldwater Cr. (F1) | 204 | 4 | West Lake (O5) | 37.0 | 3 |
| Mallinckrodt (1) | 195 | 4 | West Lake (BT12) | 35.1 | 3 |
| Coldwater Cr. (C6) | 179 | 3 | West Lake (BT3-3) | 34.0 | 3 |
| West Lake (BT9) | 178 | 3 | West Lake (EC9) | 33.8 | 3 |
| Latty Ave. (2) | 163 | 3 | West lake (O2) | 33.8 | 3 |
| West Lake (N4) | 132 | 2 | West lake (O1) | 33.0 | 3 |
| West lake (O5) | 117 | 2 | Coldwater Cr. (F5) | 32.4 | 3 |

Table 2
The $^{226}$Ra activity (Bq kg $^{1}$) and the percent unsupported $^{210}$Pb are given for the ten study area soils with the highest total $^{210}$Pb activities (Bq kg $^{1}$). For $^{226}$Ra, s ¼ 7.02 Bq kg $^{1}$. For $^{210}$Pb, s ¼ 39.5 Bq kg $^{1}$. Sample Coldwater Cr. (C6) is calculated from the $^{226}$Ra detection limit.

| Area (ID) | $^{210}$Pb | $^{226}$Ra | % unsupported $^{210}$Pb |
|---|---|---|---|
| West Lake (BT8) | 350 | 15.9 | 95 |
| West Lake (BMAC4) | 299 | 21.4 | 92 |
| West Lake (BT4) | 267 | 20.3 | 93 |
| Coldwater Cr. (F1) | 204 | 43.5 | 79 |
| Mallinckrodt (1) | 195 | 17.6 | 91 |
| Coldwater Cr. (C6) | 179 | <13 | >93 |
| West Lake (BT9) | 178 | 16.2 | 91 |
| Latty Ave. (2) | 163 | 59.7 | 63 |
| West Lake (N4) | 132 | 24.1 | 82 |
| West lake (O5) | 117 | 37.0 | 57 |

median value was 72 Bq kg $^{1}$. The background activities in study area surface soils for $^{226}$Ra and $^{210}$Pb have been previously reported as 39 Bq kg $^{1}$ ± 11 Bq kg $^{1}$ (DOE, 2001).

The ten highest soil or sediment $^{226}$Ra and $^{210}$Pb activities were all found among samples from the known disposal sites, at levels as much as 7s above the mean value of all studied samples (Table 1). For the ten soil samples with the highest total $^{210}$Pb activity, most of that activity came from unsupported $^{210}$Pb. (Table 2). The $^{226}$Ra in present in these soils is not sufficient to form the $^{210}$Pb in these samples, meaning that the parent radon came from an outside source. Eight of ten soil samples exhibiting the highest activities of unsupported $^{210}$Pb in the sample set are clustered around the West Lake Landfill and Coldwater Creek. West Lake Landfill is a known source of radon, as noted in NRC and EPA reports (NRC, 1982) (McLaren Hart 1996b). Investigators have also documented significant contamination by $^{226}$Ra in Coldwater Creek sediments (ORNL, 1987). A histogram showing the number of study area samples with detectable $^{210}$Pb that are above or below the soil background and DOE soil cleanup guideline is shown in Fig. 2.

Fig. 3 plots $^{226}$Ra vs. $^{210}$Pb activity for each soil sample with detectable $^{210}$Pb. The linear fit between $^{226}$Ra and $^{210}$Pb is poor ($R^2$ ¼ 0.068). If the ten highest activity $^{210}$Pb soil samples are

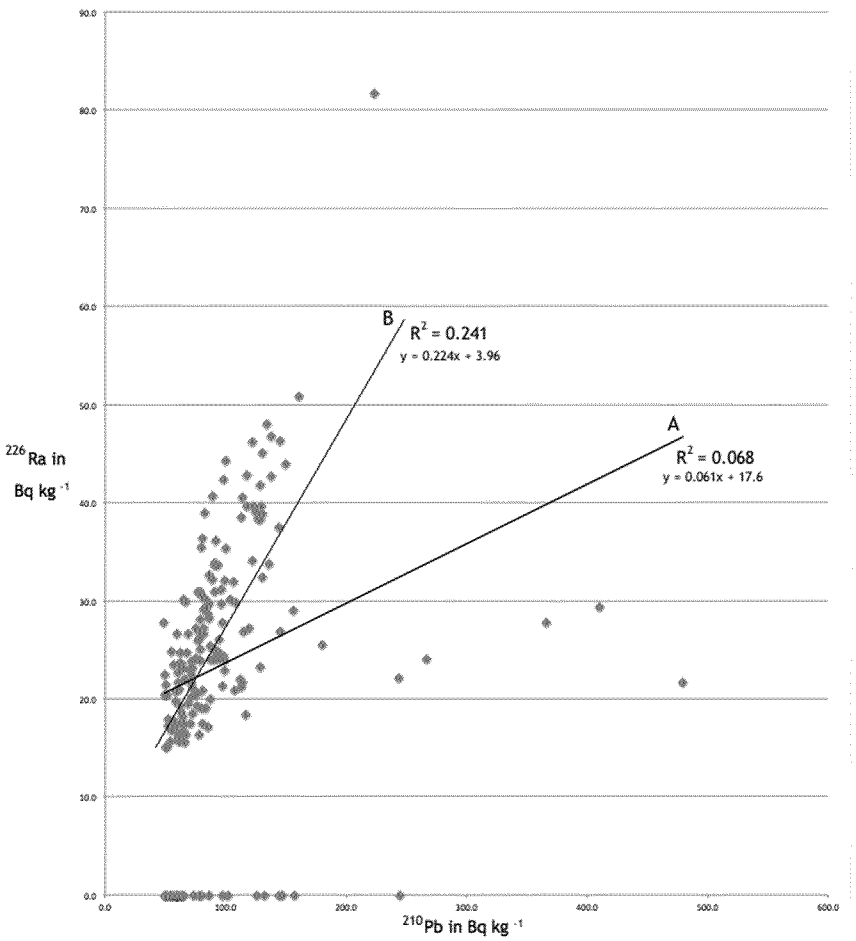


Fig. 3. Plot of $^{226}$Ra vs. $^{210}$Pb (Bq kg $^{1}$) in soil samples with detectable $^{210}$Pb. $R^2$ ¼ 0.068 for the set of soils with detectable $^{210}$Pb (line A). $R^2$ ¼ 0.241 for the set of soil with detectable $^{210}$Pb, neglecting the ten highest $^{210}$Pb activity soil samples (line B).

neglected, the linear fit improves somewhat ($R^2$ ¼ 0.241). This poor fit adds to the evidence that a portion of the $^{210}$Pb in soils is not related to in-situ $^{226}$Ra, but to an external radon source.

Mean gross alpha count rate was 6.58 counts hr $^{1}$ cm $^{2}$ with s ¼ 2.77 counts hr $^{1}$ cm $^{2}$. Mean gross beta count rates for all samples were 50.2 CPM g $^{1}$ with s ¼ 2.03 CPM g $^{1}$. None of the West Lake or "impacted" sample sets were 2s or more above the mean gross alpha or gross beta values. The mean alpha count rate for the bedrock and background loess samples was higher than for the study area samples at 8.82 counts hr $^{1}$ cm $^{2}$ with s ¼ 3.53 counts hr $^{1}$ cm $^{2}$. Gross alpha and beta activity were not indicators of impact from uranium processing wastes in this sample set.

Among the in-situ radon in soil gas tests (n ¼ 4), a Spanish Village sample collected adjacent to West Lake Landfill had $^{222}$Rn at >1330 Bq m $^{3}$of soil gas. The remaining West Lake Landfill in-situ soil tests were nondetect for $^{222}$Rn at <15 Bq m $^{3}$. Soil gas $^{222}$Rn activity was 4030 Bq m $^{3}$ at a public athletic field 0.6 km from the West Lake Landfill.

Among the soil jar test samples (n ¼ 7), the mean $^{222}$Rn activity in Bq per kg soil tested was 4.8 with s ¼ 5.1. The soil sample from the Downtown St. Louis Mallinckrodt site had the highest head-space radon activity at 15.0 Bq kg $^{1}$ soil. The jar test results were well below the levels detected in-situ around the West Lake Landfill, and do not reproduce the high in-situ levels detected. It is important to note, however, that given the highly variable soil conditions and radon emission rates encountered, firm conclusions can not be drawn from this small number of samples.

For house dust samples, both $^{210}$Pb and $^{226}$Ra were detected in nine of 13 samples tested (Table 3). The small mass of some dust samples caused increased detection limits, and eight of 21 were too small to test at all. The range of $^{210}$Pb activities in dusts (<74 to 3790 Bq kg $^{1}$, n ¼ 13) was higher than the range in soil samples (<37 e 350 Bq kg $^{1}$). More than 90% of the $^{210}$Pb in seven of 13 house dust samples tested was unsupported. The high percentage of unsupported $^{210}$Pb found in house dusts sampled near Coldwater Creek implies that there is an external source of $^{222}$Rn that is impacting the indoor environments at these homes.

Table 3
The $^{210}$Pb and $^{226}$Ra activities in Bq kg $^{1}$ with percent unsupported $^{210}$Pb in house dusts are given for residences fronting on Coldwater Creek. Only homes with detectable $^{210}$Pb are tabulated. Sample "House E" is calculated from the $^{226}$Ra detection limit.

| Sample ID | $^{210}$Pb | $^{226}$Ra | % unsupported $^{210}$Pb |
|---|---|---|---|
| House A | 3790 | 286 | 93 |
| House B | 445 | 23.2 | 95 |
| House C | 360 | 16.8 | 95 |
| House D | 347 | 17.3 | 95 |
| House E | 271 | <5 | >96 |
| House F | 263 | 25.3 | 90 |
| House G | 225 | 11.2 | 95 |
| House H | 95 | 43.4 | 54 |
| House I | 74 | 38 | 49 |

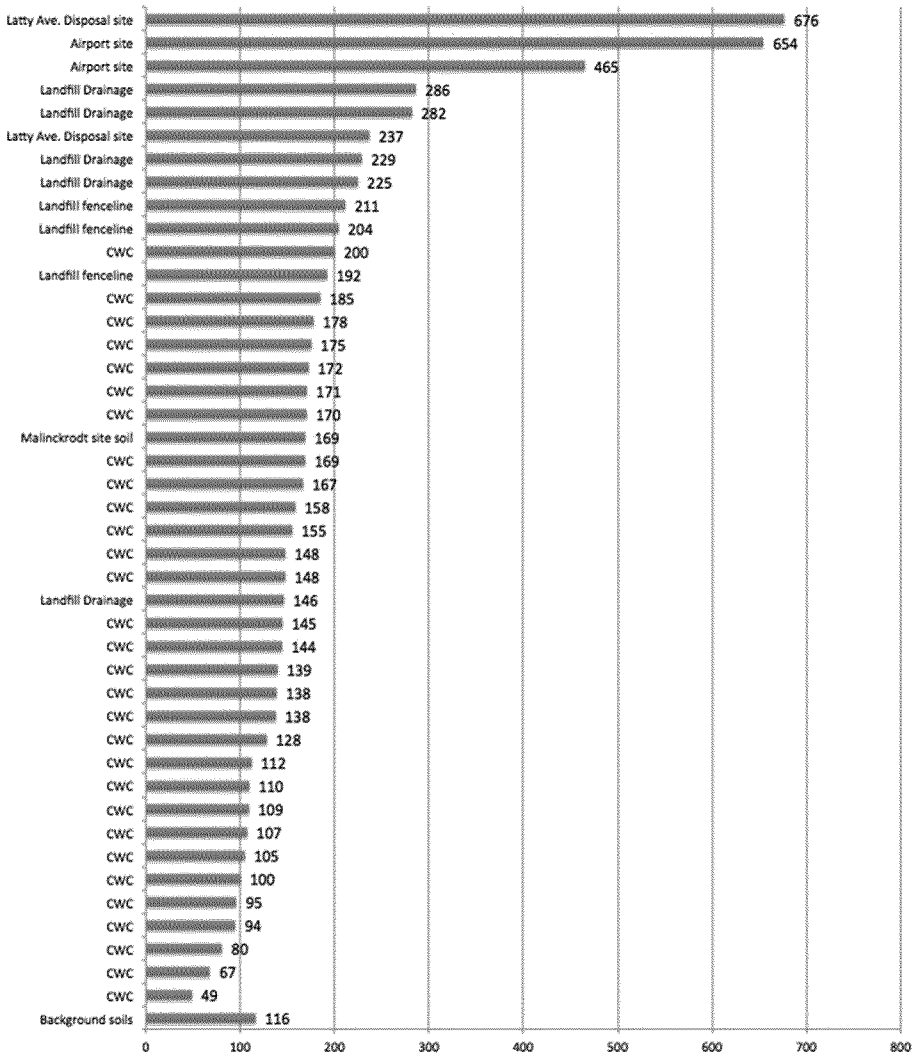


Fig. 4. Total uranium (234, 235 & 238) and thorium (228, 230 & 232) isotopes (Bq kg $^{1}$) in soils analyzed by the commercial laboratory. Soil samples are from Coldwater Creek (CWC) sediments and floodplain soils, and known disposal sites. The mean of background soils (n ¼ 5) tested at the commercial laboratory is also included.

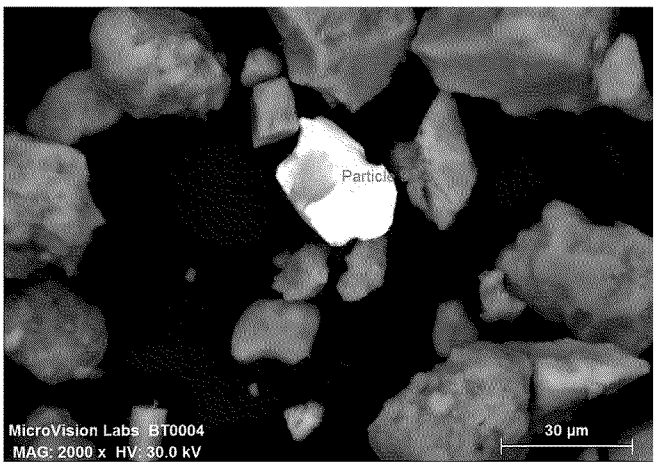


Fig. 5. SEM/EDS photomicrograph for typical thorium monazite mineral particle found at Riverwoods Park & Trail.

Fig. 4 is a histogram showing the total combined uranium and thorium activity for 43 soil and sediment samples tested by Eberline Laboratory. The value for the mean of background soils (n ¼ 5) are also shown. The entire subset of soil samples from Latty Avenue, the airport site, and the West Lake Landfill surface water drainage system exceeded the mean background uranium plus thorium soil activity of 116 ± 41 Bq kg $^{1}$. The combined uranium and thorium activities of the full set of 43 samples ranged from 49 ± 13 Bq kg $^{1}$ to 676 ± 194 Bq kg $^{1}$. Both uranium and thorium are found naturally in soils. Nevertheless, some amount of the original uranium, and most of the original thorium would be expected to remain in the Mallinckrodt waste materials, potentially contributing to the activities detected around the legacy disposal sites.

The combined uranium and thorium activities for the 18 of 21 house dust samples with sufficient mass to analyze ranged from 15 ± 6 Bq kg $^{1}$ to 1305 ± 324 Bq kg $^{1}$ with a median value of 54 ± 26 Bq kg $^{1}$, versus 34 ± 5.7 Bq kg $^{1}$ in the control house dust. This range was higher than that of the soil samples. No information is available about whether these higher levels reflect potential workplace-related contamination. The indoor dust samples were collected from Coldwater Creek floodplain homes, plus one control house 20 km distant from the study area.

Individual soil particles from known disposal sites were

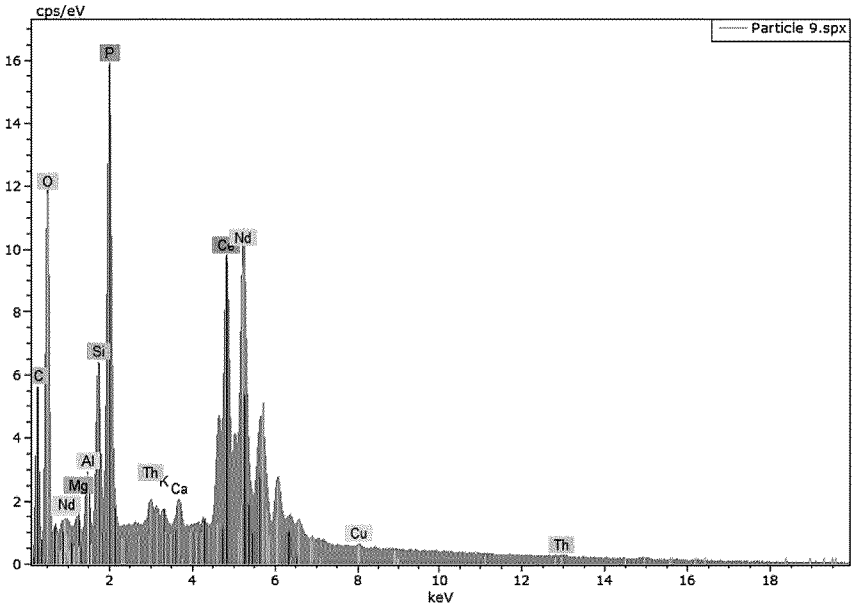


Fig. 6. SEM/EDS data for typical thorium monazite mineral particle found at Riverwoods Park & Trail.

examined using scanning electron microscopy/energy-dispersive X-ray. Results were generally unremarkable and typical for soil materials. Multiple metal particles in the 5 to 25 um size-range were detected. Most were typical industrial metal particles such as copper, zinc, lead, and other nonradioactive metal particles or paint pigments. One area had unusual results. Two soil samples came from a park adjacent to the West Lake Landfill. The site is locally known as the Riverwoods Park & Trail, and is a pathway for surface water runoff from the landfill. The samples had individual mineral particles (Fig. 5) with the characteristic spectra of thorium monazite minerals. (Fig. 6) This naturally-occurring radioactive mineral is native to some soils and is also part of the dross removed from uranium ores during processing into uranium metal. While this mineral can be native to soils, it is also among the materials processed by the Mallinckrodt firm in St. Louis, MO. This sample with thorium monazite had $^{210}$Pb activity ¼ 366 Bq kg $^{1}$ ± 19 compared to the study set mean of 54 Bq kg $^{1}$ (s ¼ 39.5).

## 6. Conclusions

This paper reports radionuclide analyses of the 287 surface soil, dust and sediment samples, collected to test whether significant, off-site dispersal of radionuclides has occurred from the West Lake Landfill site in Bridgeton, MO. Levels of $^{210}$Pb in key samples were well above background activities, and were significantly out of secular equilibrium with other members of the uranium decay chain. This is strong evidence that the $^{210}$Pb originated by decay of short-lived, fugitive radon gas that escaped the landfill. The use of the unsupported $^{210}$Pb marker was an important element of our analysis, allowing the identification of waste-impacted areas.

$^{210}$Pb activities were highest in areas known to be contaminated with wastes from the Mallinckrodt uranium processing wastes.

Radon soil headspace test and in-situ pore-volume radon activities for soil samples were widely variable, with too few samples available to directly relate these activities to the presence of uranium or uranium processing wastes in soils and sediments. Some individual samples had very high ratios of radon in headspace to soil masses. Given the importance of radon releases from soils to air as a vector for public exposure to radioactivity, increasing the density and frequency of radon measurements around the West Lake Landfill should be an important priority. If the West Lake Landfill fire were to intrude upon areas with buried uranium-processing wastes, radon emissions may increase further.

Isotopes of uranium and thorium reach high levels in sediments around Coldwater Creek. More disturbingly, indoor dusts in homes adjacent to Coldwater Creek have potentially higher levels of uranium and thorium than those found in sediments at known disposal sites.

After reviewing the 287 environmental sample results, the most effective method for tracking uranium-processing wastes was to monitor unsupported $^{210}$Pb, as well as uranium and thorium in sediments and house dusts.

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## References

Army Corps of Engineers, February 2002. Working Draft-the North Country Uranium Residues Historical Synopsis, St. Louis, Missouri; Army Corps of Engineers (Army Corps). St. Louis District Office, Formerly Utilized Sites Remedial Action Program.

Bridjes, O'Neil, October 23, 2013. Documents Sought on Radioactive Landfill Fire, St. Louis American. http://www.stlamerican.com/news/local_news/article_8b8dbf58-3c49-11e3-bd9b-001a4bcf887a.html .

Clark, J., Gregory, Kenneth John, Gurnell, Angela M., Jan 1, 1987. Horizons in Physical Geography. Rowman & Littlefield, ISBN 0-389-20752-7.

Criss, Robert E., March 2013. Risk and Character of Radioactive Waste at the West Lake Landfill. Washington University, Dept. Of Earth and Planetary Sciences, Bridgeton, Missouri. Report. http://mediad.publicbroadcasting.net/p/kwmu/files/201311/2013-03-14_Robert_Criss_West_Lake.pdf.

Department of Energy, September 1979. Formerly Utilized MED/AEC Sites Remedial Action Program, Radiological Survey of the St. Louis Airport Storage Site. Missouri, St. Louis. DOE/EV-0005/16 .

Department of Energy, March 1987. Guidelines for Residual Radioactive Material at Formerly Utilized Sites Remedial Action Program and Remote Surplus Facilities

Management Program Sites. Revision 2, p. 9. http://energy.gov/sites/prod/files/FUSRAP_ResidualRadGuidelines.pdf .

Department of Energy, November 1993. Baseline Risk Assessment for Exposure to Contaminants at the St. Louis Site. St. Louis, MO, DOE/OR-23701e41.1.Tables 2.4 and D.6.

Department of Energy, May, 2001. Addendum to the CERCLA/RCRA Background Soil Study. Fernald Environmental Management Project, Fernald, Ohio.

Environmental Protection Agency, September 1983. Final Environmental Impact Statement for Standards for the Control of Byproduct Materials from Uranium Ore Processing (40 CFR 192) table 4.5.

Environmental Protection Agency, April 1995. Region VII, Operable Unit 2 RI/FS Work Plan, West Lake Landfill, Bridgeton, pp. p.2e39. MO, prepared for Laidlaw Waste Systems (Bridgeton) Inc., by Golder Associates, Rev. 0.

Environmental Protection Agency, January 31, 1996. Superfund Record of Decision: Feed Materials Production Center (USDOE). EPA/ROD/RO5e96/312. Table 9.3. http://www.lm.doe.gov/land/sites/oh/fernald_orig/Future/PDFs/r0596312.pdf .

Environmental Protection Agency, May 2008. Record of Decision, West Lake Landfill Site, Bridgeton Missouri. Operable Unit 1. http://www3.epa.gov/region07/cleanup/west_lake_landfill/pdf/west_lake_landfill_ROD2008.pdf.

Environmental Protection Agency, November 2015. Health and Environmental Protection Standards for Uranium and Thorium Mill Tailings (40 CFR Part 192). http://www2.epa.gov/radiation/health-and-environmental-protection-standards-uranium-and-thorium-mill-tailings-40-cfr .

Richard G. Hewlitt and Francis Duncan, Atomic Shield, A History of the United States Atomic Energy Commission, vol. II, 1947/1952.http://energy.gov/sites/prod/files/2013/04/f0/Hewlett%20and%20Duncan%20-%20Atomic%20Shield%20%28complete%29.pdf.

Kaltofen, M.P.J., April 5, 2015. Assessing human exposure to contaminants in house dust, dissertation. Worcest. Polytech. Inst. Dept. Civ. Environ. Eng.

Lippmann, J.L., Criss, R.E., Osburn, G.R., 2010. Reworked Loess and Red Clays in Missouri Caves: a Comparative Compositional Study. Missouri Speleology, 50 #2-3, pp. 10e20.

Mason, M.G., August 19, 1977. History and Background Relative to the Radiological Monitoring of Mallinckrodt by the Energy Research and Development Administration. Report prepared for Mallinckrodt, Inc.

McLaren-Hart, 1996a. Soil Boring/Surface Soil Investigation Report. West Lake Landfill Areas 1 & 2.

McLaren- Hart, November 1996b. Radon Gas, Landfill Gas and Fugitive Dust Report. West Lake Landfill Areas 1&2.

National Academy of Sciences, 1997. National Research Council, Committee on an Assessment of CDC Radiation Studies, Commission on Life Sciences, a Review of the Radiological Assessments Corporation's Fernald Dose Reconstruction Report, p. 7. http://www.nap.edu/catalog/5519/a-review-of-the-radiological-assessments-corporations-fernald-dose-reconstruction-report .

National Institutes of Occupational Safety and Health, March 10, 2005. ORAU Team Dose Reconstruction Project for NIOSH, Basis for Development of an Exposure Matrix for the Mallinckrodt Chemical Company St. Louis Downtown Site and the St. Louis Airport Site. St Louis, Missouri, Period of Operation: 1942-1958, ORAU TKBS-0005. http://www.cdc.gov/niosh/ocas/pdfs/arch/mallink1.pdf .

National Institutes of Occupational Safety and Health (NIOSH), April 12, 2010. Special Cohort Evaluation Report. Petition SEC-00150. Table 5e1. http://www.cdc.gov/niosh/ocas/pdfs/sec/slaps/slapser-150-r0.pdf .

U.S. Nuclear Regulatory Commission, May 1982. Radiological Survey of the West Lake Landfill. St. Louis County, Missouri, NUREG/CR-2722.

Nuclear Regulatory Commission, June 1988. Radioactive Material in the West Lake Landfill. Summary Report, NUREG-1308, Rev. 1. http://www.epa.gov/region07/cleanup/west_lake_landfill/pdf/3WestLake_Kay_Drey.pdf.

Nuclear Regulatory Commission, March 12-24, 1994. Region III, Inspection Report No. 999-90003/9020(DRSS).

Nuclear Regulatory Commission, 2015a. 10CFR Part 61, Licensing Requirements for Land Disposal of Radioactive Waste. http://www.nrc.gov/reading-rm/doc-collections/cfr/part061/.

Oak Ridge National Laboratory, May 1987. Radiological Survey of Properties in the Vicinity of the Former Cotter Site. Hazelwood/Berkeley. ORNL/TM-10008, MO.

World Nuclear Association, 2012. Supply Uranium. http://www.world-nuclear.org/info/Nuclear-Fuel-Cycle/Uranium-Resources/Supply-of-Uranium/ .