Background Groundwater Quality, Review of 2012–14 Groundwater Data, and Potential Origin of Radium at the West Lake Landfill Site, St. Louis County, Missouri

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USGS
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Conversion Factors

Inch/Pound to International System of Units

Multiply	By	To obtain
	Length	
inch (in.)	2.54	centimeter (cm)
inch (in.)	25.4	millimeter (mm)
foot (ft)	0.3048	meter (m)
mile (mi)	1.609	kilometer (km)
mile, nautical (nmi)	1.852	kilometer (km)
yard (yd)	0.9144	meter (m)
	Area	
acre	4,047	square meter (m^2)
acre	0.4047	hectare (ha)
square mile (mi^2)	2.590	square kilometer (km^2)
	Volume	
ounce, fluid (fl. oz)	0.02957	liter (L)
pint (pt)	0.4732	liter (L)
gallon (gal)	3.785	liter (L)
gallon (gal)	0.003785	cubic meter (m^3)
cubic foot (ft^3)	0.02832	cubic meter (m^3)
cubic yard (yd^3)	0.7646	cubic meter (m^3)
	Flow rate	
foot per day (ft/d)	0.3048	meter per day (m/d)
gallon per minute (gal/min)	0.06309	liter per second (L/s)
	Mass	
pound, avoirdupois (lb)	0.4536	kilogram (kg)
ton, short (2,000 lb)	0.9072	megagram (Mg)
	Radioactivity	
picocurie per liter (pCi/L)	0.037	becquerel per liter (Bq/L)
Picocuries per gram (pCi/g)	0.037	becquerel per liter (Bq/g)
	Hydraulic conductivity	
foot per day (ft/d)	0.3048	meter per day (m/d)
	Hydraulic gradient	
foot per mile (ft/mi)	0.1894	meter per kilometer (m/km)

Vertical coordinate information is referenced to the insert datum name (and abbreviation) here for instance, "North American Vertical Datum of 1988 (NAVD 88)."

Horizontal coordinate information is referenced to the insert datum name (and abbreviation) here for instance, "North American Datum of 1983 (NAD 83)."

List of Acronyms and Abbreviations

CSU	Combined standard uncertainty
DGLS	Missouri Department of Natural Resources Division of Geology and Land Survey
DOE	U.S. Department of Energy
EPA	U.S. Environmental Protection Agency
ESRI	Environmental Sciences Research Institute
L_score	Leachate score
MCL	Maximum contaminant level
MCOO	Mid-continent Cambro-Ordovician dolostone
MDA	Minimum detectable activity
MDNR	Missouri Department of Natural Resources
MSD	St. Louis Metropolitan Sewer District
NAD83	North American Projection 1983
NGVD29	National Geodetic Vertical Datum of 1929
NPL	National Priorities List
OU1	Operable Unit 1
OU2	Operable Unit 2
p	Probability value
rho	Spearman rank-order correlation coefficient
RIM	Radiologically-impacted material
ROD	Record of Decision
RP	Responsible parties
RPD	Relative percent difference
SI	Saturation index
UO_2	Urananite
USGS	U.S. Geological Survey
UTM	Universal Transverse Mercator National Geodetic Vertical Datum 1929
WLL	West Lake Landfill

List of Chemical Abbreviations

Al	Aluminum
As	Arsenic
B	Boron
Ba	Barium
Br	Bromide
Ca	Calcium
Cl	Chloride
Co	Cobalt
Cr	Chromium
Fe	Iron

Fe_t	Total iron
HCO_3	Bicarbonate
Mn_t	Total manganese
I	Iodide
K	Potassium
Mg	Magnesium
Mn	Manganese
Na	Sodium
Ni	Nickel
pE	Electron potential
pH	In standard units
Ra	Radium
Ra-226	Radium-226 isotope
Ra-228	Radium-228 isotope
Ra228/226	Ratio of radium-228 to radium-226
SO_4	Sulfate
Sr	Strontium
Th-228	Thorium-228 isotope
Th	Thorium
Th-230	Thorium-230 isotope
Th-232	Thorium-232 isotope
U	Uranium
U-234	Uranium-234 isotope
U-238	Uranium-238 isotope
VOC	volatile organic compounds

List of Unit Abbreviations

ft	feet
ft/mi	feet per mile
g	gram
gal/min	gallons per minute
mi	mile
mg/L	milligrams per liter
µg/L	micrograms per liter
pCi/L	picocuries per liter
pCi/g	picocuries per gram

Executive Summary

During the spring of 2014, the U.S. Environmental Protection Agency (EPA) Region 7 requested that the U.S. Geological Survey (USGS) review data from the recent (2012–14) comprehensive groundwater sampling done at the West Lake Landfill (WLL) site, St. Louis County, Missouri. Of primary concern to the EPA is the presence of combined radium above the maximum contaminant level (MCL) of 5 picocuries per liter (pCi/L) in samples from several monitoring wells at the WLL site and that these detections could be the result of migration from radiologically-impacted material (RIM) placed in two areas at the WLL site during 1973. To provide a framework of groundwater-quality conditions for comparison to data from the 2012–14 comprehensive sampling, a review and compilation of historical data was done from various publications on the geohydrology and groundwater quality of the St. Louis region, the occurrence and geochemistry of radionuclides in various aquifer systems throughout the midcontinent, the geochemistry of municipal solid-waste landfills, and previous studies of the WLL site. The data compilation included groundwater samples collected during 2013 from 12 existing water-supply wells within 5 miles of the WLL site. The review of historical data and 2013 sampling data provided the range of background concentrations of chemical and radiological constituents in groundwater within the Missouri River alluvium and Mississippian-age bedrock near the WLL site.

Historical data indicate that bedrock groundwater quality in St. Louis County and St. Louis City is variable ranging from less than 150 milligrams per liter (mg/L) to more than 17,000 mg/L dissolved solids, and only about 50 percent of the wells completed in Mississippian-age rocks during the 1900s yielded potable water. Within 2 miles of the WLL site, background data indicate that at a localized scale of a few hundred to perhaps a thousand feet, there can be large fluctuations in concentrations of constituents such as sodium (110–490 mg/L), sulfate (32–1,400 mg/L), and boron (63–2,600 micrograms per liter [μg/L]) within the St. Louis Limestone or Salem Formation. Groundwater quality in the Missouri River alluvium is less variable. Concentrations of dissolved or total combined radium in background groundwater samples collected from wells within 5 miles of the WLL site open to the Missouri River alluvium or Mississippian-age bedrock were less than 4 pCi/L, and ratios of radium -228 to radium-226 (Ra228/226) ranged from 1.0 to 4.98 (alluvium) and 0.09 to 2.11 (bedrock); however, an important source of uncertainty in the background concentrations of radium in groundwater is the small set of background data that includes only 11 samples from 6 bedrock wells and 17 samples from 14 alluvial wells.

Based on the frequency of chloride, bromide, and iodide concentrations above background in groundwater samples from the WLL site, 47 of the 83 monitoring wells (37 alluvial wells and 10 bedrock wells) at the WLL site are affected by landfill leachate. Wells with the largest leachate effects also tended to have among the largest concentrations of dissolved iron, the smallest concentrations of dissolved sulfate and uranium, and produce anoxic groundwater that is iron and possibly sulfate reducing. Concentrations of dissolved combined radium were significantly larger (*p* value less than 0.0001) in samples from alluvial or bedrock monitoring wells affected by leachate compared to samples from monitoring wells at the site that do not have leachate effects.

Overall, concentrations of dissolved and total combined radium in groundwater samples from the alluvial or bedrock aquifers at the WLL site were significantly larger than background concentrations (*p* value less than 0.01). Concentrations of dissolved combined radium exceeded the MCL of 5 pCi/L in 64 of 314 (20 percent) groundwater samples collected from the WLL site between January 2012 and March 2014 where dissolved radium was analyzed. The 64 detections above the MCL were from 31 monitoring wells. The frequency of total combined radium exceeding the MCL was slightly larger with concentrations in 91 of 315 (29 percent) groundwater samples analyzed exceeding the MCL representing 38 monitoring wells. Many of the reported combined dissolved radium results only slightly exceeded the MCL such that the reported value was above the MCL, but the lower confidence interval about the reported value did not; however, 13 monitoring wells had average dissolved combined radium above the MCL. Five of these 13 wells were deep alluvial wells that also had landfill leachate effects, and 6 of the 8 bedrock wells with average dissolved combined radium above the MCL also had landfill leachate effects. Although some of these 13 wells are generally downgradient from areas containing RIM, other wells are not. There are a number of monitoring wells at the WLL site that are downgradient from RIM areas with landfill leachate effects that do not have samples containing dissolved or total combined radium above the MCL. There also are 8 shallow alluvial wells within or adjacent to RIM areas that do not have increased radium concentrations, which indicates that there is not a strong spatial association of wells surrounding or downgradient from RIM areas with elevated radium concentrations as might be expected if RIM areas were releasing substantial quantities of radium to the groundwater. In addition, radium above the MCL can occur in samples from locations with no hydrologic or physical connection to RIM areas.

The origin and transport of radium at the WLL site is complicated by its natural occurrence in groundwater and aquifer materials in the region, and the tendency of radium to be associated with mineral surfaces such as iron-oxides that are sensitive to changes in redox conditions. Changes in redox could occur with migration of landfill leachate. There is no singular mechanism, geochemical condition, or phase association that can reliably account for all occurrences of radium above the MCL in groundwater at the WLL site. Rather, there likely is a combination of mechanisms occurring across the site.

There are four general hypotheses for the origin of dissolved combined radium above the MCL in groundwater at the WLL site that include (1) leaching of radium from RIM placed at the site during the 1970s, (2) the radium values detected are within the range of natural groundwater, (3) leaching of radium from non-RIM wastes disposed at the site, and (4) mobilization of naturally occurring radium from aquifer solids by some component of landfill leachate. Except for the radium in groundwater samples from the site being within natural variation in groundwater, no single hypothesis can be invoked to explain all occurrences of radium above the MCL at the site. There is considerable uncertainty in determining the origin of above-MCL concentrations of combined radium at the WLL site. This uncertainty is predominately caused by a small set of data to establish background concentrations in groundwater, unknown concentrations and phase associates of radium and ratios of radium 228/226 within naturally occurring aquifer materials, uncertainty of the phase associations of radium and its parents in the RIM, and unknown amounts of radium contained in leachate from more traditional sanitary and demolition landfills in the region that do not contain RIM.

Using results from soil samples collected during 1996 from RIM Areas 1 and 2 that contained more than 30 picocuries per gram (pCi/g) of radium-226 (Ra-226) as representative of RIM, the average Ra228/226 in RIM of 0.006 is substantially smaller than Ra228/226 ratios in groundwater samples from the alluvium (1.0 to 1.98) or bedrock (0.09 to 2.11) indicating that radium leached from RIM is likely not the predominant source of radium concentrations above the MCL in groundwater at the WLL site. Mass-balance models of dissolved Ra228/226 ratios and chloride concentrations were used in conjunction with the location and depth of wells with respect to groundwater flow and RIM areas to determine the likelihood of a RIM origin for radium in the 13 wells that have average dissolved combined radium above the MCL. Six of the 13 wells seem to have no hydrologic or possible physical connection to RIM areas (PZ-100-SS, PZ-102-SS, and PZ-107-SS) or have either ratios of Ra228/226 inconsistent with a RIM source (D-3 and PZ-113-AD) or chloride concentrations inconsistent with RIM/leachate source (D-83), and radium above the MCL in these wells probably is not the result of leaching from RIM. A RIM contribution to radium in the remaining seven wells cannot be ruled out with the available data. Two wells (MW-1204 and PZ-104-SD) have Ra228/226 ratios and chloride concentrations that could be consistent with a RIM origin, but have no hydrologic connection to RIM areas; however, these two wells may have had a physical connection to RIM Area 1 through hypothesized runoff when RIM in Area 1 was exposed at the land surface during the early 1970s.

Monitoring well PZ-101-SS has the largest dissolved combined radium (18.4 to 32.0 pCi/L) and total combined radium (15.7 and 27.14 pCi/L) concentrations, and the smallest dissolved and total Ra228/226 ratios (0.098 to 0.108 and 0.097 to 0.294) of any monitoring well at the WLL site; has landfill leachate effects; and has dissolved radium 228/226 ratios consistent with a possible RIM source. Of any well at the WLL site, radium above the MCL in samples from this well may have the greatest potential to be related to RIM. Runoff from RIM placed in Area 1 could have physically transported RIM into the shallow North quarry between PZ-101-SS and Area 1, or even into lower areas of the South quarry possibly affecting wells PZ-104-SD and MW-1204 that also have average dissolved combined radium above the MCL.

Leaching of radium from more "traditional" non-RIM refuse landfilled at the WLL site also may contribute to radium above the MCL in some groundwater samples from the site, particularly wells with dissolved combined radium less than about 10 pCi/L. Concentrations of total combined radium in samples collected from the Bridgeton Landfill leachate discharged to the St. Louis Metropolitan Sewer District (MSD) collection system (0.6 to 23.7, average of 7.26 pCi/L) were similar to those reported in a 2005 study of radionuclides in leachate samples from Pennsylvania sanitary and demolition landfills (1.04 to 66.2, average 9 pCi/L), but larger than background groundwater samples from the Missouri River alluvium or bedrock near the WLL site (less than 4 pCi/L).

Based on the available data, mobilization of naturally occurring radium contained in aquifer materials by chemical interaction with landfill leachate is probably an important mechanism resulting in the occurrence of radium above the MCL in groundwater at the site. The presence of increased concentrations of inorganic constituents in landfill leachate combined with anoxic conditions (iron and possibly sulfate-reducing, and methanogenesis) provides the conditions described by Szabo and others (2012) as conducive for increased radium mobility. The positive correlations between dissolved combined radium in alluvial or bedrock groundwater samples and constituents such as dissolved calcium, sodium, chloride, iron, alkalinity, and significantly larger dissolved combined radium in samples from wells with landfill leachate effects are consistent with either radium being leached from "traditional" refuse materials landfilled at the site, mobilized from naturally occurring aquifer solids by landfill leachate, or both. There is insufficient data available to distinguish between mobilization of naturally occurring radium on aquifer solids by landfill leachate or leaching of radium from non-RIM refuse because the radium, uranium, and thorium isotope ratios in the aquifer materials and coatings are unknown, as are radium concentrations and ratios in leachate from other landfills in the region that do not have RIM that might provide an estimate of radium in leachate for "typical" municipal refuse from the area.

Introduction

In the spring of 2014, the U.S. Environmental Protection Agency (EPA) Region 7 requested that the U.S. Geological Survey (USGS) review data from the recent (2012–14) comprehensive groundwater sampling conducted at the West Lake Landfill (WLL) site, St. Louis County, Missouri (fig. 1, at the back of this report). Of primary concern to the EPA is the detection of combined radium above the maximum contaminant level (MCL) of 5 pCi/L (picocuries per liter) in samples from several monitoring wells at the WLL site and that these detections could be the result of migration from radioactive impacted material (RIM) placed in two areas at the site during 1973. The WLL site includes the West Lake Landfill and adjacent Bridgeton Landfill. The USGS was asked to review the analytical data and provide a technical opinion of what monitoring wells are affected by landfill leachate and the potential origin of the above-MCL combined radium detections in groundwater at the site. In addition, the USGS was asked to provide information on background groundwater quality in the region.

This administrative report presents a preliminary evaluation of groundwater-quality data from the WLL site (fig. 1) focusing on data collected during 2012–14 from four rounds of comprehensive groundwater sampling at the site (July 2012, April 2012, July 2013, and Oct 2013) and an additional round of sampling of eight newer wells (installed during 2013) during early 2014. The evaluation is based upon comparisons of general water-quality constituents. A detailed examination of the geochemistry of individual samples or temporal trends in individual wells was not done. A review and compilation of historical groundwater data from the region was first done to determine the general nature of groundwater quality in the region, and provide background concentrations for various constituents within the Missouri River alluvial aquifer and the Mississippian-age bedrock aquifer. This compilation included a review of various publications on the geohydrology and groundwater quality of the St. Louis region, the occurrence and geochemistry of radionuclides in various aquifer systems throughout the midcontinent, the geochemistry of municipal solid-waste landfills, and previous studies of the WLL site. A detailed examination of historical groundwater data for the region was done to determine the range of background concentrations of constituents in groundwater within the Missouri River alluvium and bedrock near the WLL site. To provide additional background groundwater data, the USGS conducted a field inventory and, in collaboration with the EPA, sampled 12 existing water wells in the vicinity of the WLL site during 2013. The sampling was done by the EPA (alluvial wells north of the WLL site) or the USGS and Responsible Parties (RP) for the WLL site. Groundwater data from the 2012–14 sampling at the WLL site were then compared to background concentrations determined for selected constituents and published literature on the chemical composition of leachate from municipal solid-waste landfills to identify samples from monitoring wells at the WLL site likely affected by landfill leachate.

Background of the West Lake Landfill Site

The 200-acre WLL site is in northwestern St. Louis County and lies on the edge of the Missouri River alluvium (fig. 1). The site was agricultural land until 1939 when a limestone quarry known as the West Lake Quarry and Materials Company began operations at the site (Herst and Associates, Inc., 2005). The primary limestone quarry was located in the southeast part of the site along the edge of the Missouri River alluvium, and limestone was mined until about the mid-1980s. The southern part of the quarry (locally known as the "South quarry;" fig. 2, at the back of this report) eventually reached about 200 feet (ft) deep with a floor elevation of about 245 ft compared to the surface elevation of about 445 ft for the adjacent Missouri River alluvium. Beginning sometime in the 1950s, parts of the quarried areas and other areas of the site were used for landfilling of municipal trash, industrial solid wastes, and construction and demolition debris (Herst and Associates, Inc., 2005). Liquid wastes and sludge also may have been disposed at the site, especially in western areas (Finkbeiner, 1989). During 1974, a permit was issued by the newly formed Missouri Department of Natural Resources (MDNR) for a landfill in the northern part of the main quarry (locally known as the "North quarry") or what is currently (2014) the Bridgeton Landfill. Also during 1974, the MDNR closed landfill areas in western parts of the site (fig. 2). The landfill in the North quarry was operated until 1985 when the landfill operation expanded into the much deeper South quarry of the Bridgeton Landfill (Herst and Associates, Inc., 2005). The Bridgeton Landfill was designed with several leachate collection risers inside the former South and North quarries, which are continuously pumped to create an inward hydrologic gradient and minimize leachate effects on surrounding groundwater (fig. 2).

During the summer of 1973, about 47,000 tons of soil containing about 8,700 tons of what is described as leached barium sulfate residues were apparently used as daily and intermediate cover for landfill operations at the WLL site (fig. 1; Herst and Associates, Inc., 2005). This material was placed in landfill areas in the northern and western parts of the site designated by the EPA as Area 1 and Area 2 (fig. 2). At this time, the North quarry was an open pit and limestone quarry operations continued in the South quarry. The soils containing the barium sulfate residues were from the cleanup of a nearby radiologically contaminated site known as the "Latty Avenue site." Wastes from 1940s uranium (U) processing for the Manhattan project from a facility in downtown St. Louis were stored at the Latty Avenue site. Apparently, the landfill operators were informed that the soils were "clean fill" (Herst and Associates, Inc., 2005). During 1990, the WLL site was placed on the National Priorities List (NPL) based on the presence of radiologic material and the potential for groundwater contamination. The soils from the Latty Avenue site and materials they may have mixed with at the WLL site are referred to as "Radiologically Impacted Material" or RIM. The RIM has been determined to be present in two areas (Area 1 and Area 2) of what is defined as operable unit 1 (OU1) of the WLL site. A second operable unit, OU2, includes other areas that were landfilled (including the Bridgeton Landfill), but where RIM was not known to be placed or occur (fig. 2).

In 2004, the landfill operation in the South quarry (fig. 2) ceased receiving wastes, but pumping of leachate risers continued. Since the 1980s, a number of site investigations have been conducted culminating with remedial investigations and feasibility studies of OU1 and OU2. During 2008, the EPA issued a Record of Decision (ROD) for the WLL site that described the selected remedy as a modified solid-waste cover for the RIM areas. In 2010, based on continued concern from local citizens and parties, the EPA determined that a supplemental feasibility study was warranted. As part of renewed efforts to examine the site, the EPA requested the RP of the WLL site to conduct four rounds of comprehensive sampling of all groundwater wells at the WLL site, which includes monitoring wells around the adjacent Bridgeton Landfill. The four rounds of comprehensive sampling began in 2012 and were completed in 2013; however, sampling of several additional monitoring wells installed during 2013 continued into early 2014. Although the EPA is investigating the site because of the radiological hazard, the MDNR oversees solid-waste related operations at the adjacent Bridgeton Landfill.

Regional Geohydrology

St. Louis County is between two major geologic structural features, the Ozark dome to the southwest and the Illinois basin to the north and east (fig. 1). Rocks in the region generally dip about 55 ft per mile (ft/mi) to the northeast; however, dips can vary because of local geologic structures (McCracken, 1966). The major structural feature in the vicinity of the site is a complex monoclinal fold and associated faults referred to as the Cap au Gres structure (Harrison, 1997) that trends west-northwest near Grafton, Illinois (not shown), then bends southward through Florissant, Mo. about 9 mi (miles) northeast of the WLL site (fig. 1). South and southwest of the Cap au Gres structure is a shallow syncline that preserves rocks of Pennsylvanian and Mississippian-age referred to as the Florissant basin by Lutzen and Rockaway (1971). The WLL site is situated within this syncline area. During the 1950s, exploration for a natural reservoir for storage of natural gas for the St. Louis region along the Florissant dome about 9 mi northeast of the

WLL site resulted in the discovery of the largest oil field in Missouri about 1,000 ft beneath the surface in the Ordovician-age Kimmswick Limestone (Rueff, 1987).

Ordovician through Pennsylvanian-age rocks are exposed in St. Louis County with older rocks exposed to the southwest and progressively younger rocks at the surface to the northeast (fig. 1). Except at lower elevations along stream valleys where they have been eroded, the uppermost bedrock in most of northern St. Louis County and St. Louis City is Pennsylvanian-age. Bedrock beneath the WLL site consists of Mississippian-age rocks that are stratigraphically equivalent to rocks of the Springfield Plateau aquifer in southwestern Missouri (Imes, 1990); however, Mississippian rocks in the St. Louis area are considered to be an independent, water-yielding unit on the northeast edge of the Ozark Plateaus because they are not in direct hydraulic communication with Springfield Plateau aquifer in western Missouri. The Mississippian-age bedrock units beneath and in the immediate vicinity of the WLL site are, in decreasing age, the Chesterian Series Ste. Genevieve Limestone; the Meramecian Series St. Louis Limestone, Salem Formation, and Warsaw Formation; and the Osagean Series Keokuk Limestone and Burlington Limestone (table 1, at the back of this report). The St. Louis Limestone and Salem Formation are extensively quarried in the region for road aggregate, and were quarried from the North and South quarries, which are now (2014) the Bridgeton Landfill. These formations also are quarried and exposed in the active Champ quarry about 1.5 mi south of the WLL site (fig. 1). The upper part of the underlying Warsaw Formation, often locally referred to as the "Warsaw Shale," has moderately thick shale beds, and is exposed in the floor of the Champ quarry to the south and likely forms the floor of the South quarry area of the Bridgeton Landfill (fig. 2). The Keokuk Limestone lies beneath the Warsaw Formation, but is not exposed in the WLL site area and often is undifferentiated from the underlying Burlington Limestone, which together are often called the Burlington-Keokuk Limestone.

The St. Louis Limestone, Salem Formation, and overlying Ste. Genevieve Limestone (table 1) generally yield little water to wells. A review of historical geologic logs for water wells in the region identified 84 wells completed in these units with yields ranging from less than 0.5 to 80 gallons per minute (gal/min) with a median yield of 2.5 gal/min, and 75 percent of the wells having yields of 5 gal/min or less. Yields of wells completed in the underlying Keokuk or Burlington Limestones and Fern Glen Formation (41 wells) were slightly larger ranging from 1 to 53 gal/min with a median yield of 5 gal/min. Most of the bedrock monitoring wells at the WLL site (fig. 1) are completed within the St. Louis Limestone (locally referred to as "SS" series wells) or the Salem Formation (referred to as "SD" series wells); however, there are four deeper monitoring wells at the WLL site completed in the Keokuk Limestone ("KS" series wells

PZ-100-KS, PZ-104-KS, PZ-106-KS, and PZ-111-KS). No monitoring wells are completed within the Warsaw Formation.

Groundwater quality in St. Louis County and St. Louis City is variable with dissolved solids ranging from less than 150 mg/L (milligrams per liter) to more than 17,000 mg/L, and only about 50 percent of the wells sampled that were completed in Mississippian-age rocks yielded potable water (Miller and others, 1974). Groundwater within synclinal areas of the bedrock is generally of poorer water quality (more saline) than surrounding areas (less saline), and the synclines were hypothesized to act as traps for mineralized water and prevent flushing by freshwater recharge (Miller and others, 1974). Most wells yielding potable water from the Mississippian-age bedrock examined by Miller and others (1974) were near the outcrop line of the Mississippian Meramecian Series rocks or to the south where these rocks are exposed at the surface (fig. 3, at the back of this report). Except for small areas of freshwater within the low-yielding Mississippian and overlying Pennsylvanian rocks, groundwater in the bedrock in northern St. Louis County and St. Louis City generally is saline (greater than 1,000 mg/L dissolved solids) with salinity generally increasing in deeper formations (Miller and others, 1974; Imes, 1990). Using historical data from 72 water-quality samples from wells open to Mississippian-age bedrock obtained from the Missouri Geological Survey (MGS), Imes (1990) mapped a generalized freshwater-saltwater transition zone in the Mississippian rocks defined by the 1,000 mg/L dissolved solids concentration line extending northwestward through St. Louis County (fig. 3). The zone was interpreted to be a mixing zone between the freshwater flow system to the southwest and a saline water flow system to the northeast previously discussed by Gleason (1935). Concentrations of chloride (Cl) in these 72 wells were variable ranging from 1 to 3,420 mg/L. At the regional 1:750,000 scale mapped by Imes (1990), the 1,000 mg/L line lies just south of the WLL site (fig. 3). Miller and others (1974) describe a more complex distribution of higher salinity groundwater in the Mississippian-age bedrock with some areas of lower chloride concentrations north of the 1,000 mg/L dissolved solids line (fig. 3) mapped by Imes (1990). Rather than gradually transitioning from freshwater to mineralized water, the boundary between freshwater and mineralized water in the Mississippian-age bedrock in St. Louis County can be abrupt (Owens, 1960). The abrupt boundary was thought by Owens (1960) to be related to structural features and changes in lithofacies within individual formations. Both Owens (1960) and Miller and others (1974) noted the common presence of mineralized water within and beneath the Warsaw Formation. In addition, thin stringers of gypsum have been noted in a zone at the base of the St. Louis Limestone (Owens, 1960), and gypsum beds several feet thick have been mapped in the St. Louis Formation in western Illinois just east of St. Louis, Mo. by the Illinois Geological Survey (Saxby and Lamar, 1957). Dissolution of gypsum or oxidation of pyrite, in addition to upwelling of high sulfate (SO_4) groundwater from lower stratigraphic horizons hypothesized by Miller and others (1974), could contribute SO_4 to groundwater. Pyrite is commonly reported on drill logs from the region and the WLL site.

Although the MGS records indicate that there were some domestic and industrial water-supply wells drilled in central and northern St. Louis County during the early and middle 20th century, by the 1970s, nearly all of these wells had been abandoned in favor of municipal supplies developed from the Missouri and Mississippi Rivers as the area transitioned from rural to urban land use. In the southwest part of St. Louis County, Cambrian and Ordovician-age rocks of the underlying Ozark aquifer yield freshwater and are widely used for domestic and public supply (Miller and others, 1974). The MGS records contain geologic logs for 28 water wells completed in the Mississippian-age bedrock within a 3-mi radius of the WLL site (fig. 4, at the back of this report). Two of these 28 logs have comments indicating the presence of saltwater (MO 005322 and MO 011387), one log (MO 006635) indicates freshwater in the Keokuk Limestone, two logs from wells drilled in the 1920s were not available for review (MO 002118 and MO 003039), and the remaining 23 logs have no water-quality comments. Log M0 011387 is for a well drilled about 2.5 mi east of the WLL site and indicates that the well was initially drilled 585 ft deep into the Warsaw formation but encountered "mineralized water" and was plugged back to 450 ft deep within the Salem Formation with a final yield of 0.75 gal/min. Log MO 005322 is for a well about 1.5 miles south of the WLL site that encountered saline water in the Keokuk Limestone at a depth of 515 ft. The assumption was made that the 23 logs for wells completed in Mississippian-age bedrock within 3 mi of the WLL site with no water-quality comment produced potable water. One of the logs with no water-quality comments was from the West Lake Quarry and Materials Company well (MO 010022) that was drilled at the WLL site in 1948 to 325 ft deep into the Warsaw Formation. Background well USGS-D1 sampled during 2013 is about 3.5 mi south of the WLL site (fig. 5, at the back of this report), is 305 ft deep (open to the St. Louis Limestone and Salem Formation), and produced about 0.3 gal/min of freshwater (Cl of 45 mg/L); however, a former 405-ft deep well across the street (MGS log MO 025279; fig. 4) that was open to the St. Louis Limestone down to the top of the Burlington Limestone produced about 4 gal/min of salty water. Attempts to locate well MO 025279 during 2013 were unsuccessful, and the well seems to have been destroyed during the early 2000s. Well log MO 003806 is about 6 miles northeast of the WLL site and is the closest well with a historical log to the WLL site that mentions brackish or saline water within the St. Louis Limestone or Salem Formation.

Background Groundwater Quality

Before effects of landfill leachate on groundwater quality at the WLL site can be determined, an understanding of background water quality was needed, especially the upper ranges of constituent concentrations in groundwater. A review of literature for the region was done, and groundwater data for St. Louis and St. Charles Counties were obtained from various agencies (USGS, MDNR, and U.S. Department of Energy [DOE]) or various publications. Included in the review were data for 2013 groundwater samples collected by the EPA or the USGS from private wells in the vicinity of the WLL site, 2013-14 data from two monitoring wells installed about 1,200 ft east of the Bridgeton Landfill, historical (1990s) samples from seven former offsite monitoring wells about 1,200 ft south and presumably upgradient from the WLL site, and monitoring well data collected as part of permit expansion of a sanitary landfill in a bedrock quarry about 1 mi south of the WLL site known as the "Champ Landfill" (fig. 1). Other than data from the WLL site, there were no existing radionuclide data from the region, and the 2013 sampling effort was an attempt to fill that data gap. Summary statistics were computed for various groupings of data (for example, by agency and by proximity to the WLL site) for the Missouri River alluvium and the Mississippian-age bedrock aquifer.

Because of the variable water quality within the region (especially within the Mississippian-age bedrock), the upper limit of background for various chemical constituents for the WLL site was selected as the 95th percentile of data from groundwater samples collected from within 5 miles of the site (table 2, at the back of this report; fig. 5). Of particular concern was the potential for the presence of brackish or mineralized water within Mississippian-age rocks in the immediate vicinity of the WLL site. Large concentrations of Cl were not present in groundwater samples from bedrock wells within 5 mi of the WLL site (maximum of 91 mg/L); however, large concentrations of sodium (Na; 110 to 490 mg/L), SO_4 (32 to 1,400 mg/L), and boron (B; greater than 2,200 µg/L [micrograms per liter]) were present in several monitoring wells completed within the St. Louis Limestone or Salem Formation at the Champ Landfill expansion area (fig. 5). Other monitoring wells at the Champ Landfill expansion area and nearby domestic bedrock well USGS-D1 open to these same units had smaller concentrations of Na (less than 99 mg/L) and SO_4 (less than 165 mg/L), indicating naturally variable concentrations of these constituents in groundwater near the WLL site. Groundwater quality in the Missouri River alluvial aquifer in the region was more consistent; however, a cluster of former offsite alluvial monitoring wells installed about 1,200 ft south of the Bridgeton Landfill and sampled during the 1990s had large concentrations of Cl (130 to 250 mg/L) compared to samples from other alluvial wells in the region (Cl less than 81 mg/L). This cluster of alluvial monitoring wells was adjacent to a small tributary downstream from a small 1960s-era subdivision, and the large Cl in the water they produced may be the result of anthropogenic effects, such as infiltration of road saltwater and wastewater leachate, and may not be representative of background conditions. Chloride concentrations from these alluvial monitoring wells were not used to derive background concentrations for the alluvial aquifer.

Determining background concentrations of radionuclides such as radium in groundwater for the WLL site is difficult because there were no groundwater samples with radionuclide data within 5 mi of the site, except for eight samples collected during the 1990s from seven former offsite monitoring wells south of the WLL site (S-80, I-50, MW-107, PZ-300-AD, PZ-300-AS, PZ-300-SS, and PZ-301-SS), and four samples from two monitoring wells installed east of the site during 2013 (PZ-212-SD and PZ-212-SS) (fig. 5). Because of the small amount of historical radionuclide data, a review of data from public-supply wells within about 30 miles of the WLL site was done in addition to the 2013 well inventory and sampling of selected water-supply wells in the area. Radionuclide data from 31 public drinking water systems in St. Charles County and Lincoln County (north of St. Charles County, fig. 1) that use groundwater for primary supply indicate average total combined radium concentrations ranging from 1.3 to 12.6 pCi/L (average of 4.5 pCi/L) and ratios of radium-228 to radium-226 (Ra228/226) in samples where both isotopes were above reporting limits ranging from 0.07 to 7.50 (average of 0.78). All of the systems except one (O'Fallon, St. Charles County, Mo.) pump water exclusively from bedrock aquifers with most having wells open to Ordovician-age or Cambrian-age formations, which are known to have high radium (Szabo and others, 2012), and open to either the St. Peter Sandstone or the Roubidoux Formation (primarily a sandstone). Miller and Vandike (1997) indicated localized occurrences of elevated radionuclides in public-supply wells near the eastern extent of freshwater within the St. Peter Sandstone. None of the public-supply wells pump water from the St. Louis Limestone or Salem Formation, and the one system that withdrew water mostly from Mississippian-age bedrock had average combined radium of 1.3 pCi/L and an average Ra228/226 ratio of 3.33.

The 2013 well inventory and sampling identified 15 private water-supply wells within about a 5-mi radius of the WLL site, and 11 of these wells were sampled (fig. 5). The wells sampled were completed within the Missouri River alluvium (9 wells) or bedrock (2 wells) with at least part of the uncased interval of the bedrock wells open to Mississippian-age rocks. Six alluvial wells north of the WLL site were sampled by the EPA, and three alluvial wells and two bedrock wells south or southwest of the site were sampled by the USGS or split sampled with the USGS and the RP. Samples from the wells were analyzed for total and dissolved radionuclides, volatile organic compounds (VOCs), and total and dissolved major and trace inorganic constituents. Sample results identified as "dissolved" were obtained by analysis of water samples that had been filtered through a 0.45-µm (micrometer) pore-size filter before analysis. The term "dissolved" is an operational definition because often radionuclides in groundwater are transported on or within small particulates such as colloids that can pass through a 0.45-um pore-size filter (Reynolds and others, 2003). Samples collected by the EPA were only analyzed for VOCs and total radionuclides, and one of the wells sampled by the USGS had a water softener that was not identified until after analytical results had been received (data were discarded). Also, sample data were used from two background bedrock monitoring wells (PZ-212-SS and PZ-212-SD) that were installed by the RP during 2013 about 1,200 ft east of the Bridgeton Landfill to provide additional background water quality for that facility (fig. 5). The maximum concentrations of dissolved and total combined radium in the first two rounds of samples collected by the RP from these monitoring wells were 0.48 and 1.05 pCi/L.

Using data from the 2013 sampling effort, data from the two bedrock wells installed east of the WLL site during 2013, and 1990s samples from former offsite monitoring well clusters south of the WLL site (fig. 5), the upper limit of background (95th percentile) for dissolved and total combined radium were 1.98 and 2.81 pCi/L for the alluvium, and 3.56 and 3.34 pCi/L for the Mississippian-age bedrock (table 2). Ratios of total and dissolved Ra228/226 ranged from 1.0 to 4.98 (95th percentile of 4.28 for dissolved and 4.98 for total) for the alluvium and 0.09 to 2.11 (95th percentile of 0.74 for dissolved and 2.11 for total) for the bedrock. The background dataset is small consisting of only 17 alluvial groundwater and 11 bedrock groundwater samples. Even including the 2013 supply-well sample data, the small amount of background groundwater data available, especially radionuclides in the bedrock units of concern at the WLL site (St. Louis Limestone and Salem Formation), is a limitation to understanding the occurrence of radium above the MCL at the WLL site.

Review of 2012–14 Groundwater Data from the West Lake Landfill Site

Between various investigations of radionuclides at the WLL site and groundwater monitoring required by solid-waste regulations at the Bridgeton Landfill (fig. 2), at least 90 monitoring wells have been installed at the site. Groundwater data examined included data from 83 monitoring wells sampled at least once during the 2012–14 comprehensive site-wide groundwater monitoring. Historical data (Herst and Associates, Inc., 2005) from seven background monitoring wells installed about 1,200 ft south of the site during the 1990s (S-80, I-50, MW-107, PZ-300-AD, PZ-300-AS, PZ-300-SS, and PZ-301-SS) and two background monitoring wells installed east of the site during 2013 (PZ-212-SS and PZ-212-SD) were included in the background dataset previously discussed and are not part of the following discussion. Monitoring wells completed in the Missouri River alluvium were designated in the well number as either shallow "AS" or "S" wells (15 wells), intermediate "AI" or "I wells (12 wells), or deep "AD" or "D" wells (11 wells). The designation of shallow, intermediate, or deep, is relative to the thickness of the alluvium at the particular location such that a "D" well could have a depth of more than 80 ft in deeper alluvial deposits while another "D" well might be only 35 ft deep in areas of thinner alluvium. Monitoring wells completed in the Mississippian-age bedrock at the site are designated as St. Louis Limestone "SS" wells (27 wells), Salem Formation "SD" wells (7 wells), or Keokuk Limestone "KS" wells (4 wells). In addition, there are three shallow (less than 25 ft deep) monitoring wells (MW-102, MW-103, and MW-104) designated as "US" wells and completed in what is referred to as "unsaturated zone" deposits that most likely are fill material above the former alluvial surface. There also are four shallow (less than 40 ft deep) leachate risers, or "LR" wells, in the south central part of the site (LR-100, LR-103, LR-104, and LR-105). To simplify the following discussion, the seven shallow unsaturated zone wells or leachate risers are considered to be within the group of alluvial wells.

The presence of censored data is problematic in reviewing any water-quality data, especially left-censored data commonly seen as less than the reporting level ("<") or non-detection ("U" or "ND") values. The effect of censored values also is problematic on statistical comparisons. The primary objective of groundwater monitoring at sanitary landfills is compliance with existing regulations for the facility. Laboratory methods used are optimized for meeting the compliance objective for samples that may have complex matrix interferences often with large concentrations of target constituents and are not necessarily optimized to resolve extremely low constituent concentrations in natural groundwater. A result is that method reporting limits can be larger than natural background concentrations in groundwater, especially for some dissolved trace elements such as copper (Cu), cobalt (Co), nickel (Ni), and others, which often are present at single part per billion concentrations or smaller. The WLL site dataset that was examined included major and trace elements, VOCs, and radionuclides. Values below the reporting level for non-radiological constituents were reported as not detected or "ND". There were few (less than 10) left censored (not detected) results for major inorganic constituents; however, more than one-half of iodide (I) values and values for dissolved trace elements such as aluminum (Al), Arsenic (As), Co, Cu, chromium (Cr), and Ni were left censored with reporting limits for some (such as Al [less than 200 to 1,000 µg/L], As [less than10 to 50 µg/L], barium [Ba; less than 110 to less than 250 µg/L], Cu [less than 25 to 130 µg/L], and Ni [less than 40 to 200 µg/L]) much larger than concentrations in natural groundwater in the region. For the left censored major ion and trace element concentrations with large reporting limits, a simplistic approach of removing the values from the dataset was used. More complex statistical techniques are available for use (Helsel and Hirsh, 2002), but probably are not warranted because the analytical techniques used, especially those for the inorganic constituents, were not optimized for determining low-level concentrations and reporting limits generally are large compared to most naturally occurring concentrations in background groundwater. Radionuclide results present a different challenge because the results are based on a probability of a result exceeding a detector background value that can vary through time, and thus, zero and negative values are possible (U.S. Environmental Protection Agency, 2004). For radionuclide data, left-censored values were replaced with the actual reported value such that 0.14 U becomes 0.14 even if the sample specific minimum detectable activity (MDA) was 0.62, unless the result was negative, in which case the negative value was replaced with a zero. For the calculation of Ra228/226 ratios (dissolved or total), if either isotope result was left censored ("U" code) the ratio was not calculated. Estimated values ("J" coded) for constituents were treated as true (non-estimated) values for statistical purposes.

Determination of Monitoring Wells affected by Landfill Leachate

Groundwater data from the 2012–14 sampling effort were compared to the upper limit of background (95th percentile of background values) for selected constituents with a focus on constituents commonly associated with municipal landfill leachate and concentrations of radionuclides, in particular Ra-226 and radium-228 (Ra-228). Elevated concentrations of Na, SO_4, and B have been used with other constituents as indicators of sanitary landfill leachate (Mirecki and Parks, 1994; Baedecker and Back, 1979; Clark and Piskin, 1977); however, because of the naturally occurring variable concentrations of Na, SO_4, and B in several monitoring wells at the nearby Champ Landfill expansion area, these constituents were not used as primary indicators of landfill leachate at the WLL site (fig. 1). At the WLL site, the primary indicators of landfill leachate considered were Cl and the minor anions bromide (Br) and iodide (I). Detections of anthropogenic VOCs commonly associated with fuels and oils (fuel-VOCs), and common chlorinated solvents or their degradation products (Cl-VOCs) also are common in landfill leachate; however, these compounds may have been present in the alleged disposal of industrial liquid wastes at the site (Finkbeiner, 1989), and their detection may not necessarily indicate landfill leachate. These compounds were tabulated, and their detection with increased Cl, B, and I concentrations provides additional confidence in identification of leachate effects; whereas, their detection in the absence of increased Cl, Br, and iodide concentrations may be equivocal. The specific fuel VOCs and Cl-VOCs are listed table A–1 in appendix A. Ratios and concentrations of Cl and minor anions Br and I were used by Panno and others (2006), Davis and others (1998), and Mutch and Carbonaro (2009) to discriminate between sources of Na and Cl in groundwater including landfill leachate. Concentrations of trace elements such as Cu, strontium (Sr), and Ni have also been used as indicators of landfill leachate (Mirecki and Parks, 1994; Schumacher and Hockanson, 1996), but there were insufficient data for these constituents (only one round of samples analyzed for Sr), and reporting levels for concentrations of several trace elements were not sufficiently low enough for the purpose of use as a tracer.

Based on the frequency of Cl, Br, and I above background, a "leachate score" or "L_score" was assigned to each monitoring well; "0" indicates no measurable effect from landfill leachate, 0.5 indicates a possible effect, and 1 indicates obvious effects from landfill leachate. As an additional constraint, at least one-half of the samples from a well must have Cl greater than background to be considered as having effects from landfill leachate (leachate score greater than 0). The reporting limit for I of 0.1 mg/L in the dataset provided was used to determine leachate effects even though this value was substantially larger than the upper limit in background groundwater samples collected during 2013 from the Missouri River alluvium (0.0218 mg/L) or bedrock (0.0074 mg/L) listed in table 2. The upper limits of background concentrations of Cl and Br were 79.5 mg/L and 0.09 mg/L for the alluvium (using no offsite RP wells) and were 91 mg/L and 0.12 mg/L for the bedrock. Background values for the alluvium did not include major ion data from the 1990s RP offsite alluvial monitoring wells because of the potential for local anthropogenic effects. Most wells at the WLL site were sampled four times during 2012–14; however, some wells were sampled fewer times because of insufficient water or because they had not been installed until midway through the comprehensive sampling program. The average Cl concentration in samples from each well was used to weight the leachate score with wells having average Cl more than three times background assigned a weight of three and wells having average Cl more than twice background assigned a weight of two (table 3, at the back of this report).

The result of the scoring and weighting indicates that of the 83 monitoring wells sampled on the WLL site during 2012–14, 47 wells (37 alluvial wells and 10 bedrock wells) are affected by landfill leachate with L_score greater than 0 (table 3; fig. 6, at the back of this report). A total of 10 of these monitoring wells (8 alluvial wells and 2 bedrock wells) had slight possible effects from landfill leachate with leachate scores of 0.5 and are considered as leachate affected for this discussion. There are 17 alluvial wells and 3 bedrock wells that had average Cl concentrations more than 3 times the background concentration of 79.5 mg/L for the alluvium or 91 mg/L for the bedrock, and 29 wells (23 alluvial wells and 6 bedrock wells) had average Cl concentrations more than twice the background (table 3). Wells with leachate effects are scattered across the site reflecting the widespread distribution of landfill materials. The eight alluvial wells that did not have measurable leachate effects were all less than 45 ft deep (designated as shallow or intermediate depth wells) in the western part of the WLL site (fig. 6). Alluvial wells with Cl concentrations more than three times background tended to be in the central and southwest parts of the site; and were a mixture of shallow, intermediate, and deep wells. A total of five alluvial wells (D-3, D-14, I-68, I-73, and PZ-113-AD) and a leachate riser (LR-105) had average Cl concentrations more than five times the background of 79.5 mg/L; and, except for well I-73, these wells were adjacent to Area 1. Bedrock wells with leachate effects were dispersed around the North and South quarries with 7 of the 10 wells being shallow "SS" series wells.

In general, concentrations of several constituents had moderate to strong correlations with landfill leachate effects (table 4, at the back of this report). A ranked pairwise correlation using all 2012–14 groundwater sample data from the site indicates moderate to strong positive correlations (Spearman's rho values greater than 0.4) between L_score and fuel VOCs (0.58), dissolved concentrations of calcium (Ca, 0.60), Ba (0.76), iron (Fe, 0.72), magnesium (Mg, 0.67), Na (0.73), potassium (K, 0.65), manganese (Mn, 0.48), and total alkalinity (0.72). Moderate to strong negative correlations were noted between L_score and concentrations of SO_4 (-0.41), uranium-234 (U-234, -0.44), and uranium-238 (U-238, -0.42) indicating a possible decrease in SO_4 and uranium (U) concentrations in anoxic leachate effected groundwater. No relations were seen between concentrations of dissolved thorium isotopes (thorium-228 [Th-228], thorium-230 [Th-230], and thorium-232 [Th-232]) and landfill leachate effects (not shown). Concentrations of several other constituents such as total alkalinity, Ba, and Fe were substantially larger than background in leachate-affected wells and tended to be largest in leachate-affected alluvial wells (fig. 7, at the back of this report). The larger concentrations of these constituents in wells identified as leachate affected is consistent with published literature documenting large concentrations of these constituents in landfill leachate (Mirecki and Parks, 1994; Clark and Piskin, 1977), indicating that the use of Cl, I, and Br to identify landfill leachate effects on groundwater has merit at the WLL site. Concentrations of SO_4 were substantially smaller in leachate-affected wells, especially alluvial wells with an L_score of 3, which had SO_4 concentrations generally less than 10 mg/L (probably resulting from SO_4 reduction) compared to the more typical 35 to 100 mg/L in alluvial wells with no leachate effects (fig. 7). Because of abundant electron donors in the form of reduced organic compounds, landfill leachate often is strongly reducing (Fe to SO_4 reducing or even methanogenic), and the smaller SO_4 concentrations in leachate-affected alluvial wells is consistent with landfill leachate effects, which can reduce SO_4 to sulfide (Mirecki and Parks, 1994). The negative correlation between L_score and U-234 and U-238 (table 4) is consistent with landfill leachate effects lowering the electron activity (pE) of groundwater leading to reduction of U. In reducing groundwater, U is immobile with the end product generally being urananite (UO_2; Morrow, 2001). Concentrations of dissolved oxygen and field measurements of pH were not provided in the WLL site data reviewed; however, the large dissolved concentrations of Fe, Mn, and in some cases small SO_4 concentrations, especially in alluvial wells with L_score greater than 0, indicate anoxic conditions that are at least Fe and Mn reducing and are probably typical of groundwater at the site. The small SO_4 and large dissolved Fe and Mn concentrations in samples from some wells may indicate groundwater in some areas is SO_4 reducing.

The major ions Na, K, Cl, and occasionally SO_4 generally make up a larger proportion of the dissolved ions in samples from wells identified as having landfill leachate effects, which is a relation readily observable as a shift in position on an ion-composition trilinear diagram (fig. 8, at the back of this report). Samples from most wells having no leachate effects trend toward the Ca+Mg-HCO_3 vertex, whereas samples from leachate-affected wells trend toward the Na+K and Cl or SO_4 vertex. Notable exceptions are samples from wells PZ-100-KS and PZ-111-KS that are completed in the Keokuk Limestone and a sample from recently installed well PZ-211-SD. Samples from these bedrock wells have Na concentrations greater than 200 mg/L and greater SO_4 relative to Cl or HCO_3 concentrations, and are similar to samples from several background wells at the Champ Landfill expansion area about 1.5 mi south of the WLL site. The trilinear diagram also shows a slight difference in major ion composition between the alluvium and bedrock in wells not affected by landfill leachate with alluvial wells tending to have a slightly larger proportion of Ca and Na relative to Mg, and Cl+SO_4 relative to HCO_3.

Radium in Groundwater at the West Lake Landfill Site

A review of quality assurance data summaries in the quarterly reports for the 2012-14 comprehensive sampling at the WLL site was done in addition to a comparison of relative percent differences (RPDs) from 32 field replicate samples to the relative analytical 2-standard deviation precision estimate for dissolved and total Ra-226 and Ra-228 of the individual samples. The median RPDs for dissolved and total Ra-226 and Ra-228 (19.2 to 37.9 percent) were smaller than the median values of the relative analytical precision estimates indicating differences in the field replicate samples generally were within the analytical precision. Comparing individual sample replicates indicates RPDs for five dissolved Ra-226 and seven total Ra-226 results were larger than the analytical precision. In contrast, RPDs for 10 dissolved Ra-228 and 12 total Ra-228 results were larger than the analytical precision. Most occurrences of RPDs exceeding the analytical precision were from values estimated below the laboratory reporting limit ("J") flagged data; however, RPDs for four dissolved Ra-228 and five total Ra-228 results exceeded the analytical precision even in samples that were not "J" flagged. Summaries of split sample results between the RP, and EPA or MDNR during the four rounds of comprehensive sampling generally indicate results were comparable within the combined standard uncertainty (CSU) of the respective values. Additional discussion of the usability of the radionuclide data is provided in appendix B.

Concentrations of dissolved and total combined radium concentrations in groundwater samples from the alluvium (including unsaturated zone/leachate risers) or bedrock at the WLL site were significantly larger (Kruskal-Wallis test p value less than 0.001) than in background groundwater samples from the alluvium or bedrock (fig. 9, at the back of this report). No differences were detected between concentrations of thorium and uranium isotopes in groundwater samples from the WLL site and background. Concentrations of dissolved combined radium exceeded the MCL of 5.0 pCi/L in 64 of 314 (20 percent) of 2012–14 groundwater samples collected from the WLL site that had dissolved radium analyzed. The 64 detections above the MCL were from 31 different monitoring wells; 41 of the 64 exceedances were from a group of 13 monitoring wells with average dissolved combined radium above the MCL (table 3). The frequency of total combined radium exceeding the MCL was slightly larger than dissolved combined radium with concentrations in 91 of 315 (29 percent) groundwater samples analyzed exceeding the MCL for total combined radium from 38 different monitoring wells (48 of the exceedances were from the group of 13 wells with average dissolved combined radium above the MCL; table 3). A total of 103 samples had either total or dissolved combined radium above the MCL. Of the 103 samples, total and dissolved combined radium exceeded the MCL in 52 samples; whereas in 39 samples, only total combined radium exceeded the MCL, and only 12 samples had only dissolved combined radium exceeding the MCL.

Analytical uncertainty should be considered when making one-sided comparisons such as a value exceeding a standard such as the MCL. Concentrations of radionuclides are reported as a result plus or minus the CSU, which is taken as the standard deviation of the analytical result. The CSU for radionuclide values reported for the WLL site groundwater data are at the 2-sigma level (two standard deviations) meaning that, assuming a normal distribution, the reported result has a 95 percent probability of being within the range of the results plus or minus the CSU. The reported value is the most likely "true" value within the range of the possible results; for example, a sample collected from leachate riser LR-103 during October 2013 contained dissolved concentrations of Ra-226 at 1.1 plus or minus 0.48 pCi/L and Ra-228 at 4.33 plus or minus 1.34 pCi/L. The Ra-226 has a 95 percent probability of being between 0.62 and 1.58 pCi/L, and the Ra-228 result has a 95 percent probability of being within 2.99 and 5.67 pCi/L. The sum of the reported results less than their respective CSU would provide a 97.5 percent confidence that the dissolved combined radium in the sample exceeded the MCL. In this case, although the sum of the reported Ra-226 and Ra-228 values (5.43 pCi/L) exceeded the MCL, the sum of the results less than the CSU of 3.61 pCi/L did not. Whereas 64 samples from 31 wells had dissolved combined radium exceeding the MCL, only 26 samples from 12 wells (8.4 percent) had the combined lower confidence interval that exceeded the MCL, indicating that a large fraction of exceedances of the MCL at the WLL site are within the CSU of the analytical results. Only 3 of these 12 wells (I-73, S-82, and PZ-302-AS) were not from the group of 13 wells with average dissolved combined radium above the MCL (table 3, at the back of this report), indicating the average dissolved combined radium value used is a reliable value for comparison to the MCL in contrast to the individual sample results. The small number of samples with the lower confidence interval with dissolved combined radium above the MCL does not imply that the reported values are clustered near the lower confidence interval, but only that 26 of the 64 samples have the highest confidence of an exceedance of the MCL. The reported laboratory values are the "statistically most likely" values among all possible values within the range of the confidence interval for an individual sample; in fact, there are 44 additional samples that have a reported dissolved combined radium less than the MCL, but have the upper confidence interval above the MCL.

Concentrations of dissolved or total combined radium in groundwater from the WLL site varied somewhat with geologic material with wells in the alluvium or bedrock having slightly larger concentrations than those in the three unsaturated zone wells and shallow leachate risers (fig. 9). A comparison of specific units indicates that the smallest radium concentrations generally were in monitoring wells open to the Keokuk Limestone, which are also the deepest (more than 350 ft deep) wells at the site, and the largest concentrations generally were in the deep alluvial wells (fig. 9). As expected, concentrations of total combined radium generally were larger than dissolved combined radium; however, probably as a result of analytical error and small sample size (or perhaps sampling methods and processing), dissolved concentrations tended to be slightly larger than total concentrations in samples from the leachate risers ("LR" wells) and Keokuk Limestone ("KS") wells.

A simple ranked correlation matrix indicates moderate correlations (rho values greater than 0.40 or less than -0.40) between concentrations of dissolved or total radium and Ra228/226 ratios and some inorganic constituents in groundwater samples from the alluvium or bedrock at the WLL site (table A–2 and A–3 in appendix A). The strongest positive correlations tended to be between dissolved or total combined radium and dissolved or total Ra-226; and dissolved concentrations of Ba, Ca, Fe, total iron (Fe_t), Mn, total manganese (Mn_t), and I. Dissolved combined radium concentrations in samples from the alluvium and bedrock had strong positive correlations with Ba (rho of 0.67 and 0.57) and weaker negative correlations with SO_4 (-0.38 and -0.20). Concentrations of total combined radium and total or dissolved Ra-226 had weak to moderate positive correlations with total Fe in samples from alluvial wells (0.18 to 0.34) and bedrock wells (0.38 to 0.51). Radium can coprecipitate with barite (Langmuir and Melchoir, 1985; Martin and Akber, 1999), but adsorption onto clay minerals and Fe oxides on aquifer minerals may be a more important source/sink of radium in groundwater (Szabo and others, 2012). Korner and Rose (1977) indicated the Fe-oxides were greatly enriched in radium compared to the surrounding rock matrix, and dissolution of these oxides in anoxic conditions can release radium to the groundwater (Landa and others, 1991). Krishnaswami and others (1982) showed that interaction of radium with amorphous Fe and Mn oxides can be rapid with radium adsorbing within a matter of minutes; similarly, desorption can be rapid.

Concentrations of dissolved combined radium were positively correlated with landfill leachate effects (rho of 0.30; table 4). Of the 13 wells having average dissolved combined radium above the MCL, 11 were from wells having leachate effects (table 3). Kruskal-Wallis tests indicate significantly larger (*p* less than 0.0001) dissolved combined radium samples from alluvial or bedrock wells affected by leachate (L_score greater than 0) compared to samples from wells that do not have leachate effects. The significantly larger dissolved combined radium concentrations in wells with landfill leachate effects, however, do not necessarily indicate that the source of the radium is from leaching of RIM at the WLL site.

The pH and oxidation-reduction potential of the groundwater is important in understanding and interpreting radium concentrations in groundwater. Because field measurements of temperature, pH, dissolved oxygen, and turbidity were not available, only a cursory examination of possible geochemical controls of groundwater quality and on radium concentrations in groundwater at the WLL site could be completed. The geochemical reaction modeling code PhreeqeC (Parkhurst and Appelo, 2013) was used to calculate saturation indices (SI) for barite. Graphs of barite SI as a function of Ba and SO_4 concentration indicate that groundwater samples from the site span a range of barite SI values from about -2.0 to 1.5, and exhibit a strong increasing trend with increasing SO_4 concentration (fig. 10, at the back of this report). Although a number of samples are supersaturated with respect to barite (SI greater than or equal to 0), the fraction of samples with dissolved combined radium above the MCL and barite SI greater than or equal to 0 (17 percent) was similar to the fraction of samples having dissolved combined radium above the MCL and barite SI less than 0 (25 percent) and not significantly different (*p* value of 0.54), indicating that barite may not be an important control in groundwater at the WLL site. Although Ba concentrations were significantly larger in wells with leachate effects (*p* value less than 0.001), many barite SI values in leachate-affected wells were smaller because of the much smaller SO_4 concentrations in leachate-affected wells (fig. 10). Barite is unlikely to precipitate where SO_4 reduction is ongoing, but rather is likely to dissolve.

Reduction of Fe in Fe-oxides on aquifer solids can be important because chemical reduction of these oxides could release sorbed metals and radionuclides into the groundwater, or prevent their adsorption because the surfaces are coated with reduced divalent Fe and Mn cations (Szabo and others, 2012). In groundwater from principal aquifers from across the United State containing SO_4 or HCO_3 as the predominate anion, Szabo and others (2012) noted a correlation between radium and Fe (rho of 0.52 and 0.40, respectively), and found that most radium concentrations that exceeded the MCL were from anoxic groundwater that generally had high concentrations of Fe or Mn. They also noted that in some areas there was an association between radium and high concentrations of Ca, Mg, Ba, and Sr, which can compete with radium for sorption sites on aquifer solids. In groundwater with SO_4 as the predominate anion, Szabo and others (2012) noted that most radium above the MCL was from Fe-reducing groundwater characterized by low (less than 1 mg/L) dissolved oxygen, Fe concentrations greater than about 100 µg/L, and SO_4 concentrations less than 10 mg/L. Although groundwater samples from the WLL site are mostly Ca-Mg-HCO_3 water type, a similar pattern is observed in the 303 samples from the WLL site dataset that had reported results for dissolved Fe, dissolved combined radium, and SO_4. Dissolved combined radium exceeded the MCL in 56 of 248 (23 percent) samples from presumably anoxic groundwater (in the absence of field dissolved oxygen measurements anoxic is defined here as having dissolved Fe greater than 100 µg/L) compared to exceeding the MCL in only 6 of 60 (10 percent) samples from presumably oxic groundwater (fig. 11, at the back of this report). The 28 percent fraction of samples with radium exceeding the MCL from anoxic groundwater samples that probably were SO_4-reducing (defined here as SO_4 concentration less than 10 mg/L and dissolved Fe concentration greater than 100 µg/L) was slightly larger compared to the 19 percent fraction of samples exceeding the MCL from presumably Fe-reducing groundwater (SO_4 greater than 10 mg/L and dissolved Fe greater than 100 µg/L; fig. 11). When limited to only samples from alluvial wells, the fraction of samples having dissolved combined radium above the MCL in presumably SO_4-reducing samples (29 percent, 20 of 69 samples) was considerably larger than the fraction of samples having dissolved combined radium above the MCL in presumably Fe-reducing samples (10 percent, 7 of 67 samples). In bedrock groundwater samples, 42 percent (5 of 12) of samples from presumably SO_4-reducing groundwater had dissolved combined radium above the MCL compared to 29 percent (23 of 78) exceeding the MCL in presumably Fe-reducing groundwater. The tendency for dissolved combined radium to exceed the MCL in anoxic groundwater samples, especially those that likely are Fe or SO_4-reducing (large dissolved Fe and small SO_4 concentrations), indicates that adsorption onto Fe-Mn oxides and redox conditions within the groundwater that affects the stability of these oxides probably are important controls on radium in groundwater at the WLL site.

Radium, like other metals, can adhere or sorb onto small particulates such as clay or iron-oxides resulting in larger concentrations in unfiltered or "total" samples than filtered samples. Often total radium concentrations will be correlated with sample turbidity (Tuckerfield and others, 2005). Many monitoring wells at the WLL site yield small quantities of water and samples often can be turbid (Ward Herst, Herst and Associates, Inc., written commun., 2013). Because field measurements made during sample collection such as water temperature, pH, dissolved oxygen content, and turbidity were not available in the database provided, the effect of sample turbidity on total concentrations of radionuclides such as total combined radium cannot be evaluated. Because of analytical variability associated with any one sample and uncertainty introduced in unfiltered samples that can have large turbidly, the focus of subsequent discussion on the distribution and possible origin of radium in groundwater will be mostly on dissolved radionuclide concentrations in monitoring wells at the site.

The 31 monitoring wells having at least 1 sample with dissolved combined radium concentration above the MCL do not show obvious clustering around RIM areas. The largest concentrations of dissolved combined radium were detected generally in the central and eastern parts of the site near the North and South quarry areas (fig. 12, at the back of this report). An example is shallow leachate riser (LR-103) in the central part of the site that had a dissolved combined radium concentration (5.43 pCi/L) above the MCL in one sample that is distant from RIM areas, and the geologic log indicates no fill material was encountered during its installation. Except for a detection of 6.97 pCi/L in a sample from shallow (22 ft deep) alluvial well PZ-302-AS, none of the samples from the 11 wells along the south part of the site west of the South quarry had detections of dissolved combined radium above the MCL; however, several of these wells have landfill leachate effects. What is unique about these wells is that, with the exception of well D-81 that is 60 ft deep, all of the wells in this area are shallow alluvial wells less than 35 ft deep. Within the alluvium at the WLL site, the occurrence of samples with dissolved combined radium above the MCL was greatest in deep alluvial wells (7 of 11 wells) compared to intermediate wells (3 of 12 wells), shallow wells (3 of 15 wells), or unsaturated zone and shallow leachate risers (1 of 7; table 3).

The 13 wells at the WLL site with average dissolved combined radium above the MCL are along the west edge of Area 2 and scattered around Area 1 and the North and South quarry areas (fig. 13, at the back of this report). All five of the alluvial wells having average dissolved combined radium above the MCL are deep alluvial wells that have landfill leachate effects. Based on water-level measurements made during 2012–13 (potentiometric maps in appendix C), deep alluvial wells D-6, D-83, and D-93 in the west part of the site are generally downgradient from Area 2 in addition to other landfilled areas of the site. Deep alluvial wells D-3 and PZ-113-AD are near the southwest corner of Area 1 and presumably generally downgradient from the southern part of Area 1 and possibly parts of the North quarry (fig. 14, at the back of this report). The presence of average dissolved combined radium above the MCL only in deep alluvial monitoring wells with landfill leachate effects may be coincidental, but it is likely related to more anoxic conditions naturally present near the base of the alluvium and in leachate-affected groundwater, and the fact that the deeper alluvial sediments generally are coarser-grained and more permeable allowing for more rapid migration of landfill leachate through them. Smith and Richards (2008) noted greater lateral migration of treated sewage effluent in deeper, higher permeability parts of the Missouri River alluvium in the McBaine Bottoms near Columbia, Mo. (not shown). The increased average dissolved combined radium in deep alluvial wells D-6, D-83, and D-93 is consistent with geochemical accumulation of dissolved radium along anoxic and leachate-affected flow paths in the deeper more permeable alluvial deposits.

Eight of the nine bedrock monitoring wells having average dissolved combined radium above the MCL are located around and within about 200 ft of the North and South quarry areas of the Bridgeton Landfill, and one well (PZ-115-SS) is on the north side of Area 1 (fig. 14). This distribution is in part an artifact of well placement because there are few bedrock monitoring wells in the central part of the site and no bedrock monitoring wells in the west part of the site; however, there are some general relations between the locations and depths of bedrock wells with average dissolved combined radium above the MCL. The two bedrock wells with average dissolved combined radium above the MCL along the east and south side of the South quarry (PZ-104-SD and MW-1204) are all deep wells (bottom of screen elevations 213–260 ft) completed near or below the floor of South quarry (about elevation 240 ft), and, assuming generally inward radial flow into the South quarry because of pumping from leachate risers in the South quarry, have no landfill areas upgradient or to the east or south from them (fig. 14). Although there are no landfilled areas east or south of these wells, two of the three wells (PZ-104-SD and MW-1204) have measurable effects from landfill leachate. The remaining five bedrock wells with dissolved combined radium above the MCL are all shallow "SS" wells with the base of screens at elevations between 348–392 ft that are 100 ft or more higher than the floor of the South quarry. Two shallow bedrock wells on the east side of the North and South quarries (PZ-100-SS and PZ-102-SS) have no measurable landfill leachate effects although samples from PZ-100-SS have contained less than 5 µg/L fuel-related VOCs. Assuming generally inward flow to the South quarry from leachate pumping, the absence of leachate effects in PZ-100-SS and PZ-102-SS is consistent with no landfill areas upgradient or east or northeast from these wells (fig. 14).

Well PZ-101-SS has the largest dissolved and total combined radium concentrations at the WLL site (maximum of 32.0 and 27.2 pCi/L) and has landfill leachate effects. This well is unique because of several factors: (1) it is at a corner near the boundary between the North and South quarry and essentially has landfill on three sides, (2) it is near the buried high wall between the North quarry (floor elevation about 430 ft) and South quarry (floor elevation about 235 ft), and (3) a pumping leachate riser LCS-5A is about 200 ft to the southwest (fig. 14). Pumping from leachate riser LCS-5A and from other risers within the South quarry may place PZ-101-SS along a downward and southerly flow path between both the North quarry and Area 1, and the South quarry. The elevations of groundwater measured in alluvial well D-14 on the south side of Area 1 and leachate riser LCS-6B in the North quarry during the four rounds of comprehensive sampling were consistently 10 to 33 ft higher than in PZ-101-SS (fig. 14). The base of leachate riser LCS-6B is at elevation 430 ft, and water-level elevations of about 448 ft measured in the riser during 2012–13 indicate a minimum of 18 ft of saturated fill within the North quarry. Groundwater elevations in D-14 were higher than in LCS-6B indicating the potential for groundwater to move from the south part of Area 1 through the North quarry and the vicinity of PZ-101-SS toward the South quarry (fig. 14).

Bedrock wells PZ-107-SS and PZ-110-SS on the west side of the South quarry have average dissolved combined radium above the MCL (fig. 14). These wells have screen elevations (348 to 371 ft) more than 100 ft above the floor of the South quarry and have landfill leachate effects (fig. 15, at the back of this report). Assuming radial inward flow induced by pumping leachate from leachate risers in the South quarry, both of these wells are presumably downgradient of landfill areas to the west and northwest (fig. 14). Concentrations of Cl in well PZ-107-SS (260–340 mg/L) and deep well MW-1204 (9.1–1,400 mg/L) on the south side of the South quarry are the largest detected in bedrock wells at the site. About 600 ft southwest of PZ-107-SS and just south of Old St. Charles Rock Road was a former leachate equalization basin where leachate from the North and South quarry areas of the Bridgeton Landfill was pumped for aeration before offsite disposal (Paul Rosasco, Environmental Management Support, Inc., written commun., 2014). Based on aerial photography (Google Earth, 2011), the leachate equalization basin was in existence in 1990 and contained water as late as 2002. The large Cl concentrations in well PZ-107-SS and MW-1204 may be, in part, related to this basin. A former shallow (less than 21 ft deep) alluvial monitoring well S-53 on the west berm of the former leachate basin had Cl concentrations less than 35 mg/L that would seem to indicate no leakage from this basin; however, well S-53 may not have been of sufficient depth to intersect leachate migrating from the bottom of the lagoon.

Shallow bedrock well PZ-115-SS is on the northeast side of Area 1, and has average dissolved combined radium above the MCL and landfill leachate effects (fig. 14). Boring logs indicate that the land surface at the well is about elevation 450 ft, fill (containing concrete and other rubble) extends 12 ft deep to about elevation 438 ft, and the base of the alluvium and top of underlying bedrock is at an elevation of about 416 ft (Golder and Associates, 1996). The thickness of the alluvium increases to the west and south of PZ-115-SS and decreases to the east with no alluvium present at PZ-100-SS or PZ-101-SS (fig. 14). The screen in well PZ-115-SS is set at about elevation 376 to 366 ft, which is about 55 ft lower than the base of the North quarry (fig. 15). The presence of landfill leachate in alluvial well PZ-114-AS and adjacent bedrock well PZ-115-SS is consistent with the landfill materials immediately south of these wells. The hydraulic gradient between PZ-114-AS and adjacent well PZ-115-SS varied from downward in July 2012 to upward for the remaining three rounds of comprehensive sampling during April, July, and October of 2013; however, water-level elevations in well D-14 (about 452 ft) on the south side of Area 1 and in leachate riser LCS-6B (about 448 ft; see figs. C-1 to C-4) within the North quarry were more than 10 to 20 ft higher than in PZ-115-SS, PZ-114-AS, and other nearby alluvial monitoring wells that indicated a groundwater mound within the south part of Area 1 and possibly parts of the North quarry (fig. 14). It is likely that wells PZ-114-AS and PZ-115-SS are downgradient of this groundwater high. Down-hole gamma readings indicate RIM in Area 1 is probably above saturated fill (estimated from water levels in D-14) and in a layer varying about 1 to 5 ft thick from about elevation 456 ft to just below the land surface (figs. 3–20 and 3–21, McLaren and Hart, Inc. 1996). There are no shallow monitoring wells within the 15-20 ft of fill that lies between much of the RIM in Area 1 and the underlying alluvium that might allow for collection of leachate samples directly beneath RIM to assess radionuclide concentrations within the fill.

Possible Origin of Radium above Maximum Contaminant Level (MCL) in Monitoring Wells

While there is a strong positive relation between the occurrence of average combined radium above the MCL and effects from landfill leachate in wells at the WLL site, this relation does not necessarily indicate that the source of the radium is the result of leaching from RIM within the landfill. Tuckerfield and others (2005) summarized the origin of excess radium concentrations in groundwater associated with a landfill in Georgia into three general mechanisms:

1. Leaching of radium from the waste materials itself (at the WLL site this would be leaching from the RIM). Determining this would require information on the nature of radium in the RIM such as solid-phase associations and radium isotope ratios.
2. Natural variations in groundwater (at the WLL site, determining this would require a dataset of sufficient size to capture the natural variability in groundwater from wells near the site, but not affected by current or historical human activities).
3. Mobilization of radium contained within or on aquifer solids by some component of landfill leachate from wastes at the site. An example of this could be mobilization of natural radium from aquifer solids by sanitary landfill leachate reducing the stability of Fe-oxides in the aquifer or ions in higher ionic strength landfill leachate competing for radium on sorption sites on aquifer solids.

At the WLL site there is a fourth possible origin of radium in groundwater:

4. Radium within demolition, sanitary, or other wastes disposed at the site that are not associated with RIM materials from the Latty Avenue site.

The origin and transport of radium at the WLL site is complicated by its natural occurrence in groundwater within bedrock aquifers in the region, unknown variability at the local scale within these aquifers to some degree, and also by its natural occurrence within aquifer materials and the tendency of radium to be associated with mineral surfaces such as iron-oxides that are sensitive to changes in redox conditions that could occur with migration of landfill leachate. In addition, little is known about radium contents in other materials (municipal refuse and construction and demolition debris) disposed at the WLL site. The possible origins of radium mentioned above can be formulated as four general hypotheses to account for concentrations of radium above the MCL in groundwater at the WLL site. Any one of the four, or combinations of them, is feasible, and the following will discuss observations from the dataset in relation to each.

The ratio of Ra228/226 may provide some insight into the origin of radium in groundwater at the WLL site. Vinson and others (2012) concluded that the Ra228/226 ratio was an important indicator of the key mineral phases (silicate or carbonate) that were the probable origin of radium in the groundwater. They indicated that because of its highly reactive nature with mineral surfaces (such as iron oxides and clays) and alpha recoil (physical expulsion of radium from a solid into groundwater by alpha decay of its thorium parent), radium concentrations in groundwater generally are attributable to local water-rock interaction and the local geochemical environment, and not the result of transport from remote sources through the groundwater system. Vinson and others (2012) further concluded that the primary mechanism for increased radium in the Jordan aquifer in parts of Minnesota (not shown) was from alpha recoil of radium from the decay of parents in carbonate cements in the aquifer noting that Ra228/226 ratios in the groundwater were less than 1 and consistent with the carbonate aquifer solids that are enriched in U relative to Th (thorium; thus enriched in Ra-226 relative to Ra-228). Sturchio and others (2001) noted that the Ra228/226 ratio in groundwater from carbonate aquifers in the Midwestern United States (average of 0.30) reflects the low Th:U ratio in the carbonate rock matrix. Therefore, the Ra228/226 ratio may provide some insight into the origin of radium in groundwater at the WLL site.

Potential for leaching from radiologically impacted materials (RIM)

The 1996 subsurface investigation report for RIM Areas 1 and 2 at the WLL site listed results for radioisotopes in the U-238 and Th-232 decay series for 165 samples (Engineering Management Support, Inc., 1997). Data from samples containing greater than 30 pCi/g Ra-226 (n=12) were assumed to represent RIM. Ra-226 values in these samples ranged from 38 to 3,720 pCi/g and had Ra228/226 ratios ranging from 0.004 to 0.039. For this report, the average Ra228/226 of 0.006 was assumed to represent the average ratio in the RIM, and the assumption is made that the Ra-228 activity in the RIM is supported by the presence of its parent Th-232; and thus, Ra-228 activity is not decreasing with time. If this assumption was not true, then roughly 3 half-lives of Ra-228 decay occurred between 1973 when the RIM was placed and when the 1996 soil borings were collected, and that the average Ra228/226 in the RIM in 1973 could have been as large as about 0.06. In addition, if Ra-228 in the RIM was unsupported, then roughly 3 half-lives of decay have occurred since the 1996 soil borings were made meaning that the average Ra228/226 ratio in the RIM in 2014 could be as low as about 0.001.

Ratios of Ra228/226 in groundwater samples from the WLL site are substantially larger than the average ratio of 0.006 in RIM and generally within the range of ratios in background groundwater samples (fig. 16, at the back of this report). The large Ra228/226 ratios in groundwater samples from the site indicate that it is unlikely that leaching of RIM is a major origin of radium in groundwater across the WLL site; however, the slight negative correlations between dissolved or total combined radium and dissolved or total Ra228/226 ratios in groundwater samples from alluvial wells (-0.14 and -0.23, table A–2) and stronger negative correlations in bedrock wells (-0.53 and -0.60; table A–3) seem to indicate that the source of increased radium in some groundwater samples has a lower Ra228/226 ratio than background groundwater. The ratio of Ra228/226 in groundwater may not be static if there is not a continual input of Ra-228 along the flow path, in which case, as groundwater migrates, the Ra228/226 ratio will decrease by roughly one-half every 5.8 years because of Ra-228 decay (U.S. Environmental Protection Agency, 2014). Simple mixing of various proportions of a RIM end member with either a background alluvial or bedrock groundwater end member could result in small values of Ra228/226 ratios that are similar to a number of groundwater samples from the site; however, while simple mixing could generate Ra228/226 ratios that could be similar to some groundwater samples from the site, the simple mixing may not generate both Ra228/226 ratios and radium concentrations measured in the groundwater samples, especially those of interest that have dissolved combined radium above the MCL.

To determine if it is possible for leaching of radium from the RIM to contribute to above-MCL dissolved combined radium detected in WLL site monitoring wells, a simple radium isotope mass-balance model was computed for the 13 monitoring wells having average dissolved combined radium concentrations above the MCL. Because of analytical uncertainty and variability inherent in individual radionuclide analysis, only average values in samples from monitoring wells were examined in the model. Random errors in individual radionuclide analyses can be minimized by averaging. Model assumptions include that the Ra228/226 ratio in the RIM is 0.006, the background range of radium concentrations and Ra228/226 ratios in groundwater is known, and after leaching from the RIM and mixing with natural groundwater the system is closed with respect to radium and there is no interaction with aquifer solids or change in the Ra228/226 ratio resulting from continued decay of Ra-228. In the model, the difference between the average combined radium concentration in samples from a well and the upper limit of background is referred to as "excess radium" and is assumed to be exclusively from leaching of the RIM. Dividing the "excess radium" by the average combined radium in the well samples indicates the fraction of radium hypothetically derived from RIM leaching. This hypothetical fraction of combined radium from RIM was placed in an isotope balance equation for Ra228/226 according to the following:

$$F_{(\mathrm{RIM})} * \mathrm{Ra228/226}_{(\mathrm{RIM})} + F_{\mathrm{GW}} * \mathrm{Ra228/226}_{(\mathrm{GW})} = \mathrm{Ra228/226}_{(\mathrm{well})}$$

where

$F_{(\mathrm{RIM})}$	is the fraction of combined radium in the well above background assumed to be derived from RIM,
$\mathrm{Ra228/226}_{(\mathrm{RIM})}$	is the average Ra228/226 ratio of the RIM (0.006),
F_{GW}	is the fraction of combined radium in the well from natural groundwater sources (1- $F_{(\mathrm{RIM})}$),

$Ra228/226_{(GW)}$	is the Ra228/226 ratio of background groundwater (either dissolved or total samples), and
$Ra228/226_{(well)}$	is the average measured Ra228/226 ratio in samples from the well.

The range of dissolved combined radium concentrations in each of the 13 monitoring wells and range of dissolved or total Ra228/226 ratios in background alluvial or bedrock wells were used to provide a bracket of predicted dissolved Ra228/226 ratios in the monitoring well (table 5). Total background Ra228/226 ratios were included because of the small number of dissolved Ra228/226 ratios available (2 samples) for bedrock wells. If the average dissolved Ra228/226 from the well does not lie within the model predicted range, then is it not feasible for the RIM to be the source of the radium in the well. As an example, the average combined dissolved radium and average dissolved Ra228/226 ratio in samples from alluvial monitoring well D-3 are 7.40 pCi/L and 1.63. The upper limit for dissolved combined radium in background alluvial wells is 1.98 pCi/L (table 2) indicating that 5.42 pCi/L of radium in well D-3 or 73 percent (5.42/7.40*100) is excess above background and assumed to be from the leaching of RIM. Substituting in 0.73 for the fraction of radium from RIM, 0.006 for the Ra228/226 ratio in the RIM, 0.27 (1.0-0.73) for the fraction of background alluvial groundwater, and 1.0 to 4.98 for range of the Ra228/226 ratios in background alluvial groundwater (total or dissolved samples) into the above equation predicts the average Ra228/226 ratio in well D-3 samples to be between 0.27 and 1.34. The average measured Ra228/226 ratio of 1.63 in samples from well D-3 lies outside the predicted range indicating that using the assumptions in the model, the excess radium above background measured in the well cannot be exclusively from leaching of RIM.

An additional constraint on the radium isotope mass balance was a secondary mass balance on chloride. The chloride mass balance was constructed in a similar manner as the radium isotope mass balance with fraction of excess radium [$F_{(RIM)}$] used to estimate the fraction of landfill leachate. Chloride concentrations ranging from 100 to 2,240 mg/L reported by Tchobanoglous and others (1977) were used to approximate the Cl concentrations in the landfill leachate source term, and the upper of limit of Cl in background alluvial or Mississippian-age bedrock wells of 79.5 and 91 mg/L (table 2) were used for the background groundwater concentrations. Results from the radium isotope and Cl mass balance were evaluated in the context of groundwater flow based on water-level measurement made in monitoring wells at the site, and the 13 wells were placed into groups (A–D) according to the mass-balance results and their likelihood of having a hydrologic or physical connection to RIM areas. Group A wells have the highest potential to have a RIM component and Group D wells that have no identifiable hydrologic or physical connection to RIM areas and have the least potential for a RIM component. The subsequent discussion will examine these 13 wells in greater detail. A positive result from the isotope and Cl mass balance calculations (radium isotope ratios and Cl concentrations consistent with a possible RIM and landfill leachate origin) while consistent with a possible exclusive RIM origin, does not exclude the other three hypotheses. Given uncertainty in available information and broad data constraints, which are discussed in more detail in appendix B, more confidence is assigned to negative results from the model calculations (measured average Ra228/226 ratio or average Cl concentrations in a well are not within the model predicted range).

Completing the above computations for all 13 monitoring wells having average dissolved combined radium above the MCL (8 bedrock wells and 5 alluvial wells) indicates that dissolved Ra228/226 ratios or Cl concentrations in five of the wells (D-3, PZ-113-AD, D-83, PZ-100-SS, and PZ-102-SS) are inconsistent with the model predicted results. In addition, wells PZ-100-SS and PZ-102-SS do not seem to have either a hydrologic or physical connection to known RIM areas. A sixth well (PZ-107-SS) also has no hydrologic or physical connection to RIM areas although its radium isotope ratios are consistent with an exclusive RIM source. Concentrations of dissolved radium above background in two wells (D-3 and PZ-113-AD) cannot be derived from leaching of RIM using the assumptions in the mixing model (table 5). In both cases the average measured Ra228/226 ratio was larger than the predicted range. It is possible for the excess combined dissolved radium in the remaining 11 wells to be generated from simple mixing between background groundwater and RIM with hypothetical fractions of radium from RIM ranging from 30 to 87 percent; however, this does not indicate that RIM is the source of the above-MCL radium in these wells, only that a RIM origin cannot be readily ruled out using Ra228/226 ratios (table 5). The secondary Cl mass balance indicated that the measured Cl concentrations in three wells (D-83, PZ-100-SS, and PZ-102-SS) were substantially smaller than would be expected assuming a fraction of landfill leachate needed to generate the radium concentrations measured in excess of background. This result is expected in two of the wells (PZ-100-SS and PZ-102-SS) that had no measurable landfill leachate effects (leachate score of zero). Important limitations in the simple mass-balance model approach are the limited amount of background data for radionuclides in groundwater and variable Cl concentrations that can occur in landfill leachate. The background radionuclide dataset for groundwater in the Mississippian-age bedrock is small (only 11 samples from 6 wells with only 2 samples with both Ra-228 and Ra-226 detected above reporting levels). The upper limit of background dissolved combined radium for the bedrock is heavily influenced by samples from domestic supply well USGS-A5 about 5 mi southwest of the site (dissolved combined radium of 2.99 to 3.59) and former offsite bedrock well PZ-301-SS (maximum dissolved combined radium of 2.0 pCi/L; however, only Ra-226 was analyzed). If the dissolved combined radium values in recently (2013) installed monitoring wells PZ-212-SS and PZ-212-

SD (less than 1.0 pCi/L) just east of the WLL site are more reflective of background conditions for the bedrock at the site and background dissolved combined radium concentrations in the bedrock aquifer are less than 1.0 pCi/L, none of the eight bedrock wells with dissolved combined radium above the MCL have Ra228/226 ratios that can be generated by hypothetical leaching of RIM. Regional studies (Sturchio and others, 2001; Szabo and others, 2012) tend to indicate that such low radium concentrations in the bedrock aquifers are not commonplace, but cannot be ruled out.

Based on the radium Ra228/226 isotope mass balance, the presence of average dissolved combined radium above the MCL in deep alluvial wells D-6, D-83, and D-93 along the west edge of Area 2 cannot be excluded as having a RIM origin (table 5). While two of these wells (D-6 and D-93) fall within Group A (table 5) and are among the most likely to have a possible RIM component, the average dissolved combined radium in these wells (5.98 pCi/L and 5.63 pCi/L, respectively) is only slightly above the MCL and, except for one sample from well D-83 (5.97 pCi/L), all samples from these wells had the lower confidence level (97.5 percent confidence) combined dissolved radium below the MCL. This is interesting because the Cl concentrations in samples from well D-83 (54 to 75 mg/L, average of 62 mg/L; table 3) were smaller than the upper limit of Cl in background alluvial wells (79.5 mg/L; table 2) and as a result smaller than predicted by the secondary Cl mass balance (94–1,594 mg/L). For the Cl concentrations measured in well D-83 to lie within the predicted range using the leachate fraction of 70 percent assumed from the excess average dissolved combined radium (table 5), the background Cl concentration would need to be less than 10 mg/L, which is an unlikely scenario indicating that the hypothesized leachate fraction of 0.70 (assuming excessive dissolved combined radium is exclusively from RIM) probably is too large. Groundwater in these three deep alluvial wells is probably Fe-reducing and possibly SO_4-reducing having dissolved Fe concentrations greater than 10,000 μg/L and SO_4 concentrations generally less than 10 mg/L, and geochemical conditions are favorable for radium to be mobile. These wells are generally downgradient from Area 2 and have landfill leachate effects, albeit the effects in well D-83 are minor (leachate score of 0.5; table 3). Based on concentrations of Cl, K, Na, and total alkalinity, the largest leachate effects are in well D-93, which has slightly lower dissolved combined radium than wells D-6 and D-83 (table 5). The average Ra228/226 ratios in these wells ranged from 1.39 to 1.62, and the radium isotope mass-balance model predicts that a RIM source would have to compose about 65 to 70 percent of the dissolved combined radium in these wells and the leachate component would have to contain about 6 to 12 pCi/L of dissolved combined radium derived from RIM. The average Ra228/226 ratios in these wells are within the range of samples from background alluvial wells (1.0 to 4.98), and this ratio combined with the inconsistent Cl mass balance in well D-83 indicates uncertainty in assigning an exclusive RIM origin for the dissolved combined radium in these wells. Alternative hypotheses consistent with the data will be discussed in subsequent sections.

In contrast to alluvial wells on the west side of Area 2, deep alluvial wells D-3 and PZ-113-AD at the southwest corner of Area 1 have average dissolved Ra228/226 ratios (1.63 and 2.14, respectively) that are inconsistent with a RIM origin. Three of four individual samples from both of these wells had dissolved combined radium above the MCL and also had the lower confidence dissolved combined radium above the MCL. Concentrations of K, Na, Cl, and total alkalinity in these wells were similar as were concentrations of dissolved Fe (above 30,000 µg/L), and SO_4 (less than 0.5 mg/L). The large dissolved Fe and small SO_4 concentrations indicate probable SO_4-reducing conditions, which favor radium mobility. Although these wells are generally downgradient of RIM Area 1, have landfill leachate effects, and have measured Cl consistent with the secondary Cl mass balance; the dissolved Ra228/226 ratios are inconsistent with a RIM origin and within the range of dissolved and total Ra228/226 ratios in background alluvial groundwater samples (1.00 to 4.98). Although containing effects from landfill leachate, a RIM origin for the radium in wells D-3 and PZ-113-AD is unlikely.

Two bedrock wells in the southeast corners of the "notches" in the South quarry area (PZ-104-SD and PZ-102-SS) have above-MCL average dissolved combined radium concentrations; however, only samples from well PZ-104-SD have the lower confidence interval of dissolved combined radium above the MCL (3 of 4 samples). Based on water-level measurements made during the four rounds of comprehensive sampling (appendix C), neither of these wells seems to be downgradient of RIM areas or along a pumping induced flow path between RIM Area 1 and the South quarry. Well PZ-104-SD is the deeper of the two wells, has the largest and most variable dissolved combined radium concentrations (5.66 to 14.4 pCi/L), and is the only well with landfill leachate effects. The secondary Cl mass balance indicates measured Cl concentrations in samples from well PZ-104-SD are consistent with the hypothesized leachate fraction of 0.68 (table 5). The average Ra228/226 ratios in wells PZ-104-SD and PZ-102-SS could be consistent with a RIM origin; however, the small Cl concentrations (less than 10 mg/L) in samples from well PZ-102-SS are not consistent with landfill leachate (table 5). It seems unlikely that the increased dissolved combined radium in these wells is the result of leaching from RIM in Area 1 or Area 2, and transport via groundwater flow.

Assuming that RIM is a possible source for above-MCL dissolved combined radium in monitoring wells in the east part of the WLL site near the North and South quarries, a hydrologic connection must exist or have existed in the past between the RIM areas and the wells. Based on the potentiometric map, which shows groundwater in the alluvium flowing generally to the northwest (fig. 14), it is unlikely that Area 2 is a possible origin for above-MCL radium in monitoring wells in the central and eastern parts of the WLL site. The more likely possible source for the above-MCL dissolved combined radium is Area 1. In the absence of pumping from the South quarry area, most monitoring wells in the vicinity of the South quarry with above-MCL dissolved combined radium would be expected to be upgradient from RIM area 1 (fig. 14); however, dewatering during active quarry operations and subsequent pumping from leachate risers as the South quarry was landfilled may have altered natural groundwater flow, reversed the natural gradients, and caused groundwater in the adjacent bedrock to move generally radially into the South quarry. Because the floor of the North quarry (elevation 430 ft) is nearly 200 ft higher than that of the South quarry (elevation 240 ft), dewatering in the South quarry may induce migration of groundwater from Area 1 to flow downward and southward through the bedrock beneath the North quarry toward the South quarry (fig. 15). If this hydraulic flow is or has occurred, then it is possible that wells PZ-101-SS and PZ-110-SS, which have screened intervals at elevations between the floor of the North and South quarries, may lie along pumping-induced flow paths between Area 1 and the South quarry. Both wells have landfill leachate effects, Ra228/226 ratios that could be consistent with mixing of RIM and background groundwater, and average Cl concentrations consistent with the secondary Cl mass balance (table 5). Besides having dissolved combined radium concentrations (18.4 to 32.0 pCi/L) substantially larger than other wells at the WLL site, samples from bedrock monitoring well PZ-101-SS also have the smallest dissolved and total Ra228/226 ratios (0.098 to 0.108 and 0.097 to 0.294, respectively; fig. 16) of any well at the WLL site. Compared to well PZ-101-SS, samples from well PZ-110-SS have smaller dissolved combined radium concentrations (4.10 to 8.63 pCi/L) and larger average dissolved and total Ra228/226 ratios (0.42 to 1.52).

Bedrock wells PZ-100-SS and PZ-115-SS in the northeast part of the site have Ra228/226 ratios that could be generated by mixing of RIM with background groundwater. It is unlikely that these two wells would lie along pumping-induced flow paths from Area 1 toward leachate risers in the South or North quarries. These two wells are among the shallowest bedrock wells (screened above elevation 360 ft), and because of the proximity of well PZ-115-SS to Area 1 and an apparent groundwater mound near well D-14 in the south part of Area 1, this well could be affected by Area 1 irrespective of induced pumping effects from the South quarry. The absence of landfill leachate effect in well PZ-100-SS seems inconsistent with a RIM component because of the fact that RIM was mixed with other wastes in the landfill.

Assuming pumping induced migration of groundwater from the vicinity of RIM Area 1 toward the South quarry, it is difficult to envision that wells PZ-102-SS, PZ-104-SD, MW-1204, and PZ-107-SS lie along pumping-induced flow paths emanating from Area 1 despite all of these wells having Ra228/226 ratios that could be generated by mixing of RIM with background groundwater (table 5). These wells are more likely to be affected by groundwater moving from the east, south, or west towards pumping leachate risers within the South quarry. The South quarry is unlined and refuse is in direct contact with the bedrock along the floor and sides of the former quarry. Low water-level elevations measured in the pumping leachate risers compared to water-level elevations in bedrock wells around the South quarry have been used to indicate the presence of a groundwater gradient into the South quarry from the surrounding bedrock. This assumption seems to be inconsistent with the detection of landfill leachate in several bedrock wells around the South quarry (fig. 6); for example, water-level elevations in leachate risers in the South quarry during July 2013 ranged from about 260 to 279 ft compared to elevations of 461 and 466 ft in wells PZ-104-SD and PZ-202-SS on the east side of the quarry that have leachate effects (appendix C). The inconsistency may be related to water-level measurements made in the pumping leachate risers not being reflective of water-levels within South quarry outside of the leachate risers that may be considerably higher, especially near the edges of the quarry far from the risers. In addition, because of the layered nature in which refuse and daily and intermediate cover is placed in landfills, infiltration and waste degradation may form perched water zones within the nearly 200 ft of refuse. Preferential lateral flow of perched groundwater or landfill leachate caused by the layered nature of the fill could drive some leachate from the refuse into the adjacent bedrock along the former quarry walls where it could migrate short distances along fractures and bedding planes laterally and vertically through the bedrock and be intercepted by monitoring wells close to the former quarry face.

It is also possible that a physical connection might have existed between Area 1 and monitoring wells around the South quarry through runoff. Historical accounts indicate that the RIM was placed at the WLL site during 1973, and was used as daily and intermediate cover material (Nuclear Regulatory Commission, 1982). A review of 1968 and 1974 aerial photographs indicates that the South quarry was a large topographic low before and shortly after the RIM was placed (fig. 17, at the back of this report). In the 1974 aerial photograph, Area 1 seems to be a topographic high that could allow runoff (and associated sediment-bound radium from RIM used as daily cover) from Area 1 to flow east and possibly into parts of the North quarry and the topographic low of the South quarry. The lowest elevations in the South quarry in the 1974 photograph seem to be in the northeast in the vicinity of wells PZ-101-SS, PZ-104-SD, and PZ-111-SS (fig. 17). If runoff from Area 1 flowed into the South quarry after the RIM was placed during 1973, then it is possible that some RIM may have been transported into and perhaps mixed with refuse in the lower parts of the South quarry landfill. This possible radium-bearing runoff source could be consistent with above-MCL dissolved combined radium in deeper wells PZ-104-SD and MW-1204 on the east and south sides of the South quarry. Changes in Ra228/226 ratios in leachate pumped from the Bridgeton Landfill (fig. 2) also may be consistent with a physical connection of RIM to the lower South quarry. A shift in total combined radium concentrations and total Ra228/226 ratios in leachate discharged from the Bridgeton facility occurred beginning in 2012 with the total Ra228/226 ratio spiking followed by total combined radium concentration spiking during 2013, and then both decreasing rapidly to levels different than the previous 6 years (fig. 18, at the back of this report). A subsurface smoldering event began within waste in the South quarry in about 2010 resulting in a complex response action (including installation of gas collection and injection wells, additional leachate risers, and a synthetic cap, among others) by the RP, especially during 2012 and 2013. This event resulted in accelerated waste decomposition and increased leachate production. During 2012–13, there was substantial variability in how leachate was removed from the facility. Deep pumping from some of the original leachate risers continued, but additional pumping from new shallower leachate extraction points was added in response to the increased leachate produced from the subsurface smoldering event. Pumping of leachate during 2012–13 was extremely dynamic as new extraction points were added, others were discontinued or temporarily disconnected, and then reactivated (Paul Rosasco, Environmental Management Support, Inc., written commun., 2014). The recent apparent increase in leachate pumping from shallower sections of the South quarry seems to correspond with the shift in Ra228/226 ratios toward larger values during and after 2012–13 and a decrease in total combined radium concentrations (following the brief spike during 2013). That change in isotope ratio could be consistent with more of the leachate pumping from deeper depths before 2012 producing lower Ra228/226 ratios and larger radium concentrations consistent with leachate in the lower parts of the South quarry containing radium originating from physical transport of RIM (low radium 228/226 ratios) in runoff from Area 1 during the 1970s. A change in analytical laboratories casts some additional uncertainty because the spike in Ra228/226 ratios during the middle of 2013 also corresponds with a change in analytical laboratories with the last three leachate samples analyzed by a different laboratory (fig. 18).

Potential for radium concentrations in groundwater to be within natural variability.

The dataset for characterizing background concentrations of radium in the Missouri River alluvium and bedrock aquifer in the vicinity of the WLL site is small and probably inadequate to provide a high degree of confidence that any detection of radium above the MCL in monitoring wells at the site, with the possible exception of samples from well PZ-101-SS, is outside of the natural variation in groundwater.

Given the variability in groundwater quality as evidenced by major ion data from the Champ Landfill expansion area and historical MGS well logs within a few miles of the WLL site, the 11 radionuclide samples from 6 background bedrock wells (none of which have increased Ca, Na, and SO_4 measured in some vicinity groundwater) does not capture the range of major ions known to exist in groundwater in the area and may not capture the natural range of radium concentrations or ratios of Ra228/226 in groundwater within the Mississippian-age bedrock. In addition, concentrations of radium in groundwater can be variable depending on sample turbidity and geochemistry of the groundwater, especially the presence of anoxic conditions. The absence of field measurements (such as turbidity, dissolved oxygen, temperature, and pH) from the WLL site groundwater dataset and for some offsite samples collected by others, including the alluvial wells north of the site sampled during 2013, increases uncertainty in reviewing radionuclide data from the site. None of the samples from background bedrock wells contained dissolved or total combined radium above the MCL. The maximum total combined radium in public-supply wells from the region was 12.6 pCi/L; however, radionuclide data from public-supply and domestic wells could be biased low because these wells often have long open-intervals (often tens to hundreds of feet), and water samples from these wells is a mixture of water from many geologic units compared to bedrock monitoring wells at the WLL site that extract water from a limited 10- to 20-ft thick section of the bedrock strata within the St. Louis Limestone or Salem Formation. In water-supply wells, a small amount of water produced from isolated zones or fractures with the bedrock aquifer that might have larger radionuclide contents can be diluted by water produced from other zones. Isolated fractures could be important as Bell (1963) noted that the Salem Formation locally can contain small grains of fluorite (a mineral often enriched in uranium compared to the carbonate matrix) and also noted that during wet periods the sporadic appearance of a yellow carnotite-type mineral, which contains uranium, on rock faces of quarries in the Salem Formation near St. Genevieve, Mo.; however, two samples from the bulk limestone did not have unusually large amounts of uranium (0.0004 and 0.0003 percent). Although offsite monitoring wells PZ-212-SD and PZ-212-SS had small (less than 1.0 pCi/L) combined radium, samples from well PZ-212-SD had among the largest dissolved and total U-234 (greater than 10 pCi/L) and dissolved and total U-238 concentrations (greater than 4 pCi/L). The isotope U-238 decays through several isotopes, including uranium-234, before decaying into Ra-226. The moderate to strong positive correlation between landfill leachate effects and the presence of radium above the MCL in groundwater at the WLL site seems to indicate that natural excursions of dissolved combined radium substantially above the MCL, such as detected in PZ-101-SS (which has landfill leachate effects), in groundwater at the WLL site probably are unlikely, but cannot be ruled out.

Potential leaching of radium from technically-enhanced naturally-occurring radioactive materials (TENORM) within traditional municipal and demolition waste.

During 2005, the State of Pennsylvania analyzed leachate from 54 landfills in that state to determine if radioactive materials were present in untreated landfill leachate (Civil and Environmental Consultants, Inc., 2005). Leachate samples for the Pennsylvania study were initially analyzed for gross alpha, gross beta, gamma emitting radionuclides by spectroscopy, and tritium. Leachate samples from 19 facilities had gross alpha activities exceeding 5 pCi/L that subsequently were analyzed for other radionuclides. Samples from 18 of the 19 facilities were similar and had total Ra-226 concentrations of 0.18 to 8.79 pCi/L (average of 2.31 pCi/L), total Ra-228 concentrations of 0.71 to 8.53 pCi/L (average of 3.37 pCi/L), total combined radium of 1.31 to 11.32 pCi/L (average of 5.68 pCi/L), and Ra228/226 ratios ranging from 0.25 to 6.28 (average of 2.25). Leachate from a demolition/construction waste landfill in Lancaster County, Pennsylvania, however, had much larger concentrations of total Ra-228 (41.95 pCi/L), total Ra-226 (24.2 pCi/L), and total combined radium (66.2 pCi/L) but a Ra228/226 ratio (1.73), similar to the other 18 landfills. The large radium concentrations in leachate from the Lancaster County facility were hypothesized to possibly indicate the presence of technologically-enhanced naturally-occurring radioactive material (TENORM) such as foundry sand, coal power-plant ash, bricks, stack fly ash, or stack scrubber residue (Civil and Environmental Consultants, Inc., 2005). These materials can contain elevated concentrations of radionuclides and often are enriched in Ra-226 (World Nuclear Association, 2014). In addition, Szabo and others (2008) reported septic sludges can accumulate radium, especially the longer-lived Ra-226 isotope, and these materials often are disposed of in sanitary landfills. Because landfills contain a mixture of wastes and native soils used for daily cover, any radiological constituents found in leachate likely are a mixture derived from leaching of the wastes and native soils. The Pennsylvania study did not investigate if perhaps the natural soils at the Lancaster County facility had potentially higher natural radionuclide concentrations compared to other landfills in the study. If unrelated to naturally high radium in soils, the fact that substantially larger Ra-226 and Ra-228 concentrations exist in leachate from the Lancaster County Pennsylvania demolition/construction waste landfill indicate that it is possible that similar wastes at the WLL site may also be a source of radium.

Leaching of radium from more traditional non-RIM refuse landfilled at the WLL site may contribute to radium above the MCL in some groundwater samples from the WLL site, particularly wells with dissolved combined radium less than about 10 pCi/L. Concentrations of total combined radium in samples collected from the Bridgeton Landfill leachate discharged to the St. Louis Metropolitan Sewer District (MSD) collection system (0.6 to 23.7, average of 7.26 pCi/L) were similar to those in leachate samples from Pennsylvania landfills (1.31 to 66.2, average 8.86 pCi/L), but larger than background groundwater samples from the Missouri River alluvium or Mississippian-age bedrock near the WLL site (less than 4.0 pCi/L; fig. 19, at the back of this report). The Bridgeton Landfill leachate samples tended to have similar Ra228/226 ratios for total combined radium (0.2 to 35.1, average of 2.1) to leachate from the Pennsylvania landfills (0.25 to 6.28, average of 2.22) and background groundwater samples from the Missouri River alluvium (1.00 to 4.98, average of about 3.1), but slightly larger dissolved or total Ra228/226 ratios than background bedrock groundwater samples (0.09 to 2.11, average 0.8) and substantially larger than the 0.006 average ratio in the RIM. The larger total combined radium in Bridgeton Landfill leachate samples compared to background groundwater samples seems to indicate a radium source within the landfill complex having Ra228/226 ratios between about 0.2 and 2 (fig. 19). Because the total combined radium concentrations in Bridgeton Landfill leachate were within the range found in Pennsylvania landfills that are not known to contain radiologic materials, it is a reasonable interpretation that the radium in the Bridgeton Landfill leachate may be unrelated to RIM in Area 1 or 2, and may originate from more typical municipal, demolition, or construction wastes at the site and perhaps from mobilization from native cover soils and aquifer solids. The collection of additional dissolved radionuclide samples from the Bridgeton Landfill leachate discharged to MSD and from individual leachate risers at various depths within the complex and other landfills in the region would provide more conclusive data.

Mobilization of radium from naturally occurring aquifer materials

Tuckerfield and others (2005) examined radium concentrations around a sanitary landfill at the Savannah River site in South Carolina (not shown) to determine probable sources of elevated radium concentrations in some monitoring wells. Elevated radium concentrations (above the MCL) were found in several downgradient and at least one upgradient monitoring well. Unlike the WLL site, no radioactive materials were known to have been placed in the landfill examined at the Savanah River site; however, similar to many regions of the county, elevated naturally-occurring radium concentrations exist in groundwater in the region around the Savanah River site. Tuckerfield and others (2005) concluded that elevated radium in monitoring wells was likely derived from naturally occurring radium in aquifer materials citing similar and consistent Ra228/226 ratios in monitoring wells compared to Ra228/226 ratios in offsite groundwater wells from the region. A mechanism for increased radium in monitoring wells was hypothesized to be from landfill leachate interacting with and mobilizing naturally occurring radium contained in native materials.

One explanation for the strong positive relation between detectable effects of landfill leachate and above-MCL concentrations of dissolved combined radium in monitoring wells at the WLL site is that landfill leachate is mobilizing naturally occurring radium from aquifer materials. Szabo and others (2012) indicate that in the most general sense, aquifer materials with low sorptive capacity such as limestone in combination with geochemical properties that reduce the tendency for radium to adsorb to aquifer materials (acidic, anoxic, or sub-oxic conditions; and increased mineral content of the water) lead to the highest likelihood of radium detection in groundwater. As previously discussed, the tendency for dissolved combined radium to exceed the MCL in presumably anoxic groundwater samples from the WLL site may indicate that adsorption onto Fe-Mn oxides and redox conditions within the groundwater that affects the stability or surface chemistry of these oxides are important controls on radium in groundwater at the WLL site. Additional evidence for the importance of Fe oxides on radium concentrations can be seen in the positive correlations between total Fe and total combined radium in samples from alluvial wells (0.33; table A–2) and bedrock wells (0.46; table A–3). Of the 91groundwater samples with total combined radium above the MCL, 82 had total Fe concentrations greater than 1,000 μg/L.

Landfill leachate typically contains large concentrations of Ca, Na, Cl, Fe, HCO_3, and SO_4 (Uhlman, 1991). The presence of increased concentrations of inorganic constituents combined with anoxic conditions (Fe- and possibly SO_4-reducing and methanogenesis) provides the conditions described by Szabo and others (2012) as conducive for increased radium mobility. The positive correlations between dissolved combined radium in alluvial or bedrock groundwater samples and constituents such as dissolved Ca (0.18 and 0.51; tables A–2 and A–3), Na (0.35 and 0.06), Cl (0.36 and 0.19), Fe (0.26 and 0.36), total alkalinity (0.25 and 0.38), and effects from landfill leachate (0.30; table 4) from the site are consistent with either radium being leached from refuse materials, aquifer solids, or both. There are insufficient data available to distinguish between mobilization of naturally occurring radium on aquifer solids by landfill leachate or leaching of radium from non-RIM refuse because the radium, uranium, and thorium isotope ratios in the aquifer materials and coatings are unknown (as are radium concentrations and ratios in leachate from other landfills in the region that do not have RIM that might provide an estimate of radium in leachate for "typical" municipal refuse from the area). In addition, the small amount of background radionuclide data in groundwater increases uncertainty in understanding radium occurrence at the site.

Conclusions

1- Published historical data indicate that bedrock groundwater quality in St. Louis County and St. Louis City is variable ranging from less than 150 milligrams per liter (mg/L) to more than 17,000 mg/L of dissolved solids. Only about 50 percent of the wells completed in Mississippian-age rocks during the 1900s yield potable water (Miller and others, 1974). Within 2 miles of the West Lake Landfill (WLL) site, background data indicate that at a localized scale of a few hundred to perhaps a thousand feet, there can be large natural variability in concentrations of constituents such as sodium (110–490 mg/L), sulfate (32–1,400 mg/L) and boron (63–2,600 micrograms per liter [µg/L]) in groundwater within the St. Louis Limestone or Salem Formation. Groundwater quality in the Missouri River alluvium is less variable.

2- The Missouri River is a regional groundwater drain. Natural groundwater flow in the bedrock in the vicinity of the WLL site generally is to the northwest toward the Missouri River. At normal stages of the Missouri River, gradients between the bedrock and alluvium generally are upward, and upward gradients generally exist within the bedrock units at the site. Pumping from the Bridgeton Landfill may have locally reversed natural flow gradients in the bedrock; however, an upward gradient persists from deeper bedrock units beneath the floor of the former quarry. Groundwater flow in the alluvium at the site generally is to the west-northwest toward the Missouri River.

3- Based on the frequency of chloride, bromide, and iodide concentrations above background in groundwater samples from the WLL site, 47 of the 83 monitoring wells (37 alluvial wells and 10 bedrock wells) at the WLL site are affected by landfill leachate. Wells affected by landfill leachate also have increased concentrations of dissolved calcium, magnesium, sodium, potassium, barium, iron, manganese, strontium, total alkalinity, and dissolved combined radium. Wells with the greatest leachate effects tend to have smaller concentrations of sulfate and uranium and produce anoxic groundwater. Concentrations of dissolved combined radium were significantly larger (*p* value less than 0.0001) in samples from alluvial or bedrock monitoring wells affected by landfill leachate compared to samples from monitoring wells that do not have landfill leachate effects.

4- Although pumping of leachate from refuse within the South quarry of the Bridgeton Landfill is inferred to have created an inward gradient to minimize migration of leachate into the bedrock, several monitoring wells around the North and South quarry areas of the landfill have effects from landfill leachate.

5- An important limitation on understanding the origin of radium in groundwater at the WLL site is the small amount of background data on radionuclide concentrations in groundwater near the site. In the vicinity of the site, the background dataset for radionuclides in groundwater from the specific bedrock units of interest at the site (St. Louis Limestone and Salem Formation) consists of only 7 samples from 4 wells. Only 11 samples from 6 wells exist when including radionuclide data from domestic-supply wells within 5 miles of the site. The background radionuclide dataset for the Missouri River alluvium in the vicinity of the site is slightly larger and consists of 17 samples from 14 wells; however, dissolved radionuclide data were available for only 11 samples from 8 wells. Including data collected during 2013 from water-supply wells within 5 miles of the WLL site, concentrations of dissolved or total combined radium in background groundwater samples from the alluvium or bedrock were less than 4 picocuries per liter (pCi/L), and radium 228/226 (Ra228/226) ratios ranged from 1.0 to 4.98 (alluvium) and 0.09 to 2.11 (bedrock). The 95th percentile was used as the upper limit of background for dissolved combined radium in groundwater in the alluvium (1.98 pCi/L) and Mississippian-age bedrock (3.56 pCi/L) near the WLL site.

6- Concentrations of dissolved and total combined radium in groundwater samples from the WLL site were significantly larger than background (*p* value less than 0.01). Concentrations of dissolved combined radium exceeded the maximum contamination level (MCL) of 5 pCi/L in 64 of 314 (20 percent) of groundwater samples collected from the WLL site between January 2012 and March 2014 where dissolved radium was analyzed. The 64 detections above the MCL were from 31 different monitoring wells. The frequency of total combined radium exceeding the MCL was slightly larger with concentrations in 91 of 315 (29 percent) groundwater samples exceeding the MCL and represented 38 monitoring wells. Many of the reported combined dissolved radium results only slightly exceeded the MCL such that the reported value was above the MCL, but the lower confidence interval about the reported value was not. A comparison of the lower confidence interval for sample results (reported value less the combined standard uncertainty), which represents a value at which there is 97.5 percent confidence that the reported result exceeds, indicates that only 26 samples from 12 wells (8.4 percent) had the dissolved combined radium combined lower confidence interval that exceeded the MCL.

7- There are four general hypotheses for the origin of dissolved combined radium above the MCL in groundwater at the WLL site that include (1) leaching of radium from radiologically impacted material (RIM) placed at the site in the 1970s, (2) the radium values detected are within the range of values found in natural groundwater, (3) leaching of radium from non-RIM wastes disposed at the site, and (4) mobilization of naturally occurring radium from aquifer solids by some component of landfill leachate. Except for the radium in groundwater samples from the site being within natural variation in groundwater, no single hypothesis can be invoked to explain all occurrences of radium above the MCL at the site, and the available data are not adequate to provide definitive conclusions regarding the validity of any hypotheses.

8- The origin and transport of radium at the WLL site is complicated by its natural presence in groundwater and aquifer materials, and the tendency of radium to be associated with mineral surfaces such as iron-oxides that are sensitive to changes in redox conditions. Changes in redox conditions in groundwater can occur with migration of landfill leachate. There is no singular mechanism, geochemical condition, or phase association that can reliably account for all occurrences of radium above the MCL in groundwater. Rather there likely is a combination of mechanisms occurring across the site.

9- Using results from soil samples collected from RIM Areas 1 and 2 during 1996 that contained more the 30 pCi/L of radium-226 (Ra-226) as representative of RIM, the average Ra228/226 in RIM of 0.006 is substantially smaller than Ra228/226 ratios in groundwater samples from monitoring wells at the WLL site completed within the alluvium (1.0 to 1.98) or bedrock (0.09 to 2.11) indicating that radium leached from RIM is likely not the predominant source of radium concentrations above the MCL in groundwater at the WLL site.

10- There are several monitoring wells downgradient from RIM areas with landfill leachate effects that do not have radium above the MCL. There also are 8 shallow alluvial monitoring wells within or adjacent to RIM areas (S-82, PZ-114-AS, S-84, PZ-112-AS, PZ-113-AS, PZ-207-AS, S-61, and S-10) that do not have increased radium concentrations such that there is not a strong spatial association of monitoring wells surrounding or downgradient of RIM areas with elevated radium concentrations as might be expected if RIM areas were releasing substantial quantities of radium to the groundwater. In addition, radium above the MCL can occur in samples from locations with no hydrologic or physical connection to RIM areas such as samples from shallow leachate riser LR-103 (dissolved combined radium of 2.14 to 5.43 pCi/L) in the south-central part of the site or three monitoring wells that have average dissolved combined radium above the MCL (PZ-100-SS, PZ-102-SS, and PZ-107-SS), but probably are not downgradient of RIM areas or have a physical connection to RIM areas.

11- Of 83 monitoring wells sampled at the WLL site during 2012–14, 13 had average dissolved combined radium above the MCL. Five of these wells were deep alluvial wells that also had landfill leachate effects, and two were bedrock wells with no landfill leachate effects (PZ-100-SS and PZ-102-SS). Six of the eight bedrock wells having average dissolved combined radium above the MCL also had landfill leachate effects. While some of these 13 wells are generally downgradient from the RIM areas (such as D-3, D-6, D-83, D-93, PZ-113-AD, PZ-101-SS, PZ-110-SS, and PZ-115-SS), other wells (PZ-100-SS, PZ-102-SS, PZ-107-SS, PZ-104-SD, and MW-1204) probably are not. Mass-balance models on dissolved Ra228/226 ratios and chloride concentrations were used in conjunction with the location and depth of wells with respect to groundwater flow and RIM areas to determine the likelihood of a RIM origin for the dissolved combined radium in the 13 wells. Of the 13 wells, 6 seem to have no hydrologic or possible physical connection to RIM areas (PZ-100-SS, PZ-102-SS, and PZ-107-SS), or have either ratios of Ra228/226 inconsistent with a RIM source (D-3 and PZ-113-AD) or chloride concentrations inconsistent with RIM/leachate source (D-83), and radium above the MCL in these wells probably is not the result of leaching from RIM. A RIM contribution to radium in the remaining seven wells cannot be ruled out with the available data. Two wells (MW-1204 and PZ-104-SD) have Ra228/226 ratios and chloride concentrations that could be consistent with a RIM origin, but have no hydrologic connection to RIM areas; however, these two wells may have had a physical connection to RIM Area 1 through hypothesized runoff when RIM in Area 1 was exposed at the land surface during the early 1970s.

12- Although 7 of the 13 wells that have dissolved combined radium above the MCL cannot be ruled out as having a RIM origin using the isotope and chloride mass balance or excluded as not having a possible hydrologic or physical connection to RIM areas, this does not indicate that RIM contributes to above-MCL radium detections in groundwater at the site, only that this origin cannot be conclusively ruled out with the available data. The limited amount of background radionuclide data in groundwater; the absence of data on the distribution of radium isotopes in aquifer solids, "typical" non-RIM wastes, and "typical" landfill leachate; and the potential for landfill leachate to mobilize naturally occurring radium from aquifer solids all limit the ability to conclusively assign an origin of radium in groundwater at the site. The potential for anoxic landfill leachate to mobilize radium, whether from non-RIM waste sources in the landfill or from aquifer solids (naturally occurring), indicates that radium concentrations above the MCL in groundwater will likely remain commonplace at the WLL site.

13- Well PZ-101-SS has the largest dissolved combined radium (18.4 to 32.0 pCi/L) and total combined radium (15.7 and 27.14 pCi/L), the smallest dissolved and total Ra228/226 ratios (0.098 to 0.108 and 0.097 to 0.294) of any monitoring well at the WLL site, and has landfill leachate effects and dissolved radium 228/226 ratios consistent with a possible RIM source. Of any wells at the WLL site, radium in this well may have the greatest potential to be related to RIM. Runoff from RIM placed in Area 1 could have physically transported RIM into the shallow North quarry between PZ-101-SS and Area 1, or even into lower areas of the South quarry possibly affecting wells PZ-104-SD and MW-1204 that also have average dissolved combined radium above the MCL. In addition, pumping from leachate riser LSC-5A about 200 ft southwest from well PZ-101-SS could result in PZ-101-SS being along a flow path between the riser and RIM Area 1.

14- Leaching of radium from more "traditional" non-RIM refuse landfilled at the WLL site also may contribute to radium above the MCL in some groundwater samples from the WLL site, particularly wells with dissolved combined radium less than about 10 pCi/L. Leachate samples from the Bridgeton Landfill (2006–13) have total combined radium concentrations (0.6 to 23.7, average of 7.26 pCi/L) larger than background groundwater samples from the Missouri River alluvium or bedrock aquifer near the WLL site (less than 4 pCi/L), but that were within the range of concentrations (1.31 to 66.2, average 8.86 pCi/L) in 19 landfill leachate samples from a 2005 Pennsylvania study. None of the Pennsylvania landfills examined were known to have received radiological materials, and the largest total combined radium concentration (66.2 pCi/L) was from a construction/demolition waste landfill. The Bridgeton Landfill leachate samples also tended to have similar Ra228/226 ratios for total combined radium (0.2 to 35.1, average of 2.1) to leachate from the Pennsylvania landfills (0.25 to 6.28, average of 2.22), but substantially larger than the average Ra228/226 ratio of 0.006 in the RIM. The similar total combined radium concentrations and Ra228/226 ratios in leachate from the Bridgeton Landfill and Pennsylvania landfills indicates that the presence of RIM at the WLL site has not resulted in increased radium in leachate pumped from the Bridgeton Landfill that could not also reasonably originate from leaching of more traditional municipal, construction, and demolition wastes at the site.

15- Based on the available data, mobilization of naturally occurring radium contained in aquifer materials by chemical interaction with landfill leachate is probably an important mechanism resulting in the occurrence of radium above the MCL in groundwater at the WLL site. The presence of increased concentrations of inorganic constituents in landfill leachate combined with anoxic conditions (Fe- and possibly SO_4-reducing and methanogenesis) provides the conditions described by Szabo and others (2012) as conducive for increased radium mobility. The positive correlations between dissolved combined radium in alluvial or bedrock groundwater samples (and constituents such as dissolved calcium, sodium, chloride, iron, alkalinity, and significantly larger dissolved combined radium in samples from wells with landfill leachate effects) are consistent with either radium being leached from "traditional" refuse materials landfilled at the site, mobilized from aquifer solids by landfill leachate, or both. There are insufficient data available to distinguish between mobilization of naturally occurring radium on aquifer solids by landfill leachate or leaching of radium from non-RIM refuse because the radium, uranium, and thorium isotope ratios in the aquifer materials and grain coatings are unknown (as are radium concentrations and ratios in leachate from other landfills in the region that do not have RIM that might provide an estimate of radium in leachate for "typical" municipal refuse from the area).

16- Absent from the groundwater dataset at the WLL site are samples from individual leachate risers from within the North and South quarry areas of the Bridgeton Landfill, and leachate collection points within RIM areas and landfilled materials between RIM and underlying alluvium that might provide more conclusive evidence of the potential for migration of radium from RIM areas compared to leachate collected from non-RIM areas. Also absent are samples from leachate risers at various locations and depths within the Bridgeton Landfill, some of which may have recently been installed, that could provide more reliable data to understand variations in radium within leachate discharged from the facility and possible differences in radium generated from various intervals within the refuse.

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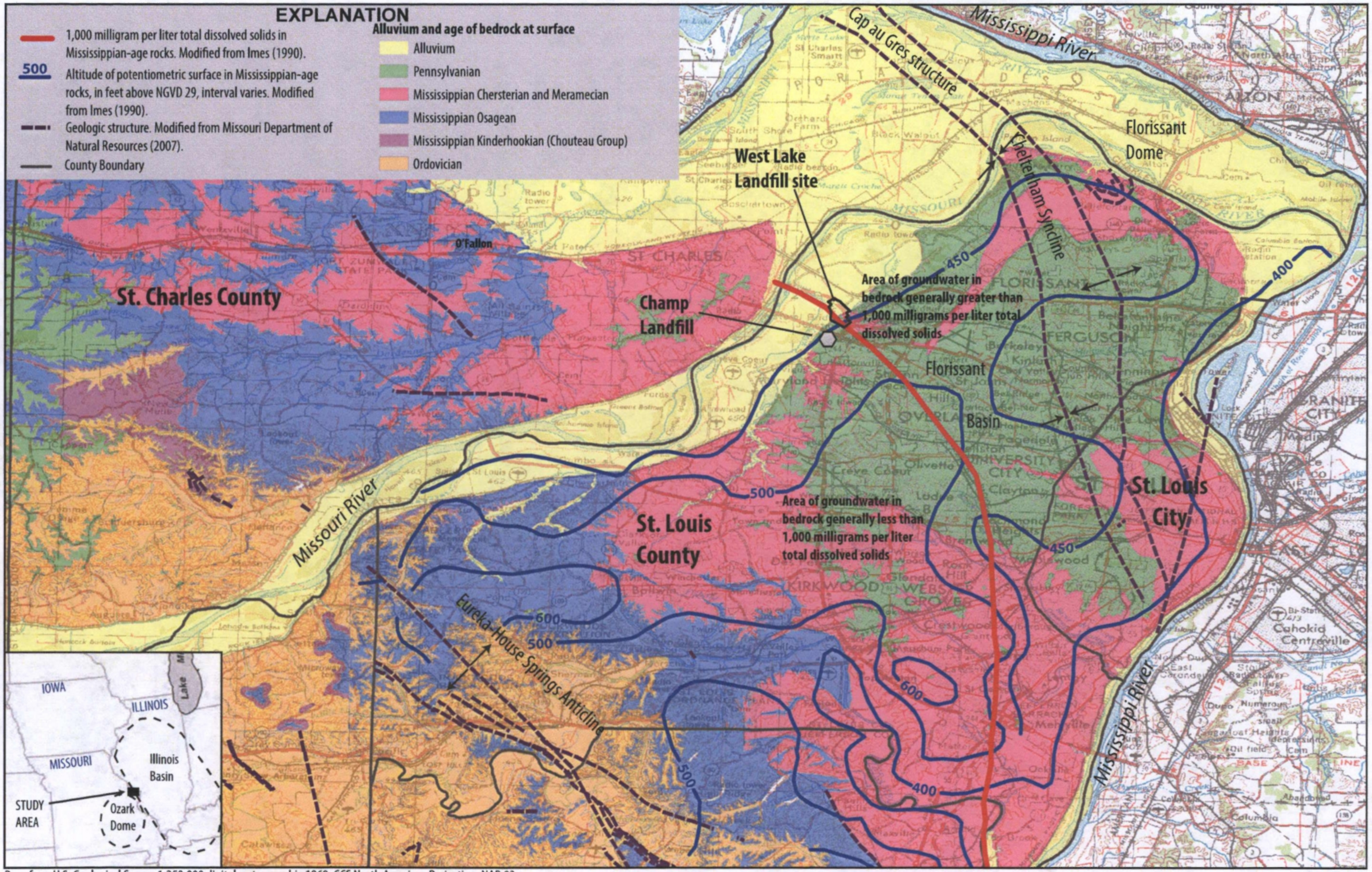
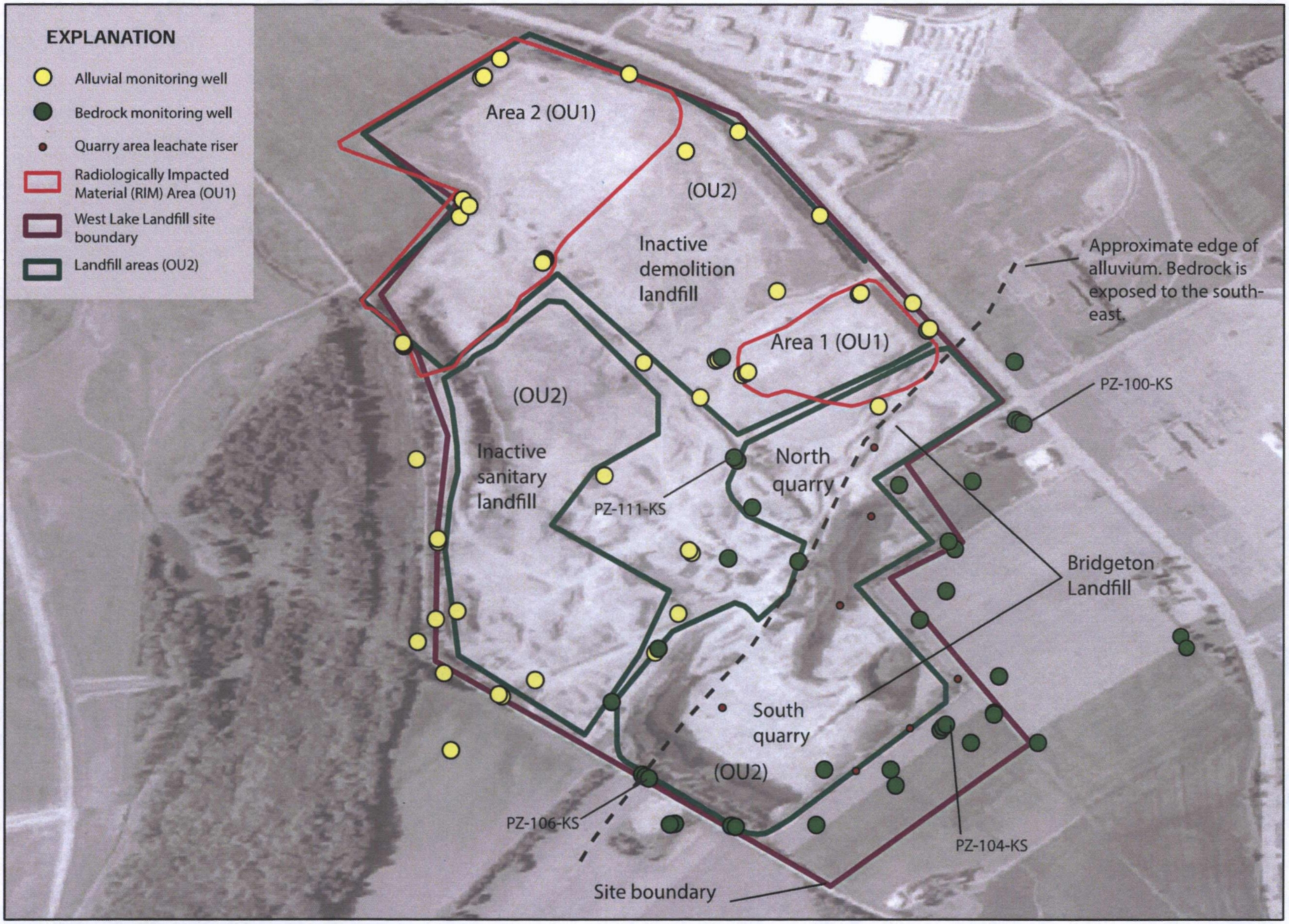


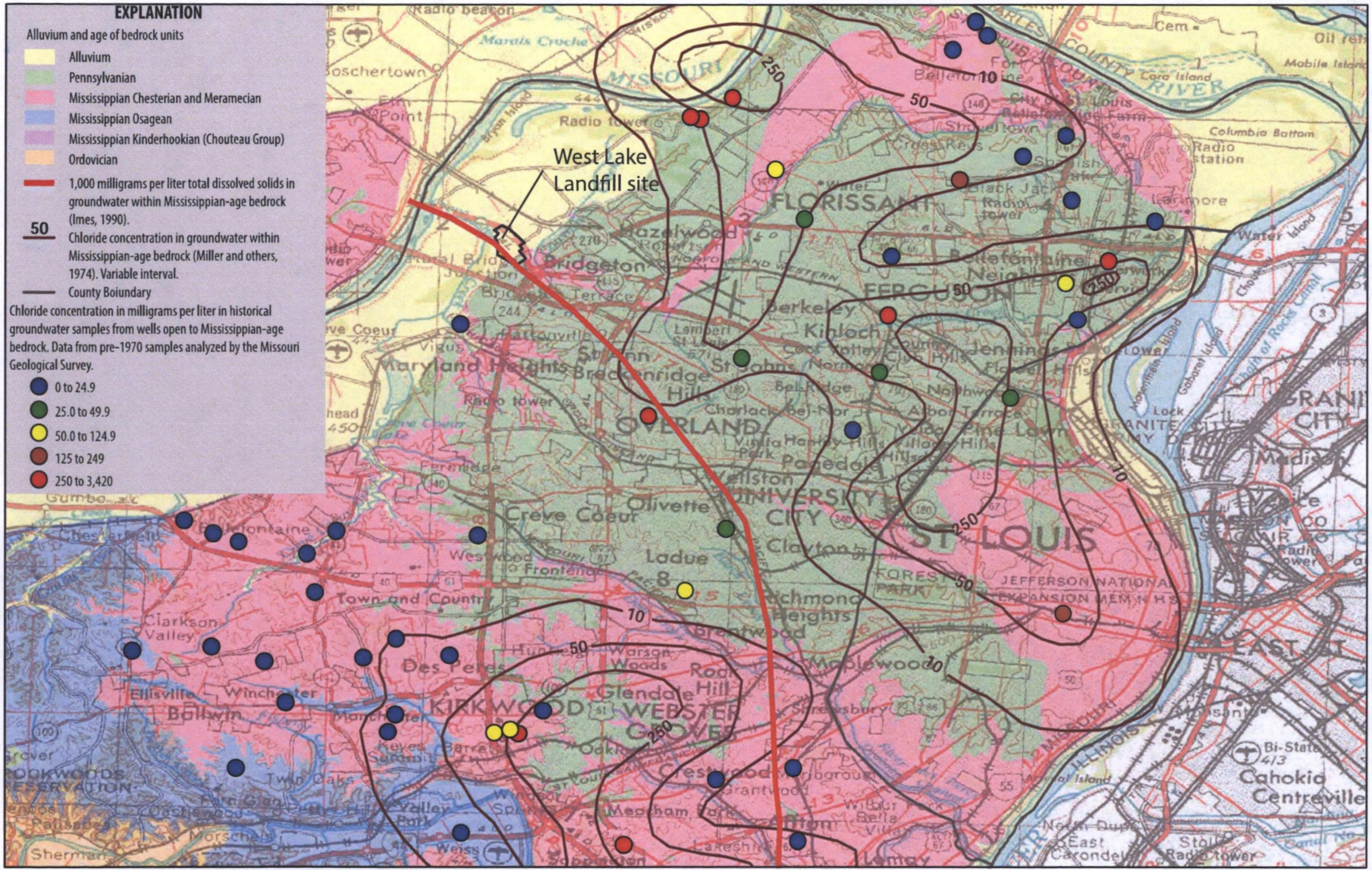
Base from U.S. Geological Survey 1:250,000 digital raster graphic,1969, GCS North American Projection, NAD 83
Geologic structure, bedrock, and alluvium from Missouri Department of Natural Resources, Missouri Environmental Geology Atlas 2007
Insert map modified from Higley and others (2003)

Figure 1. Geologic structure, age of bedrock units, and generalized potentiometric surface within the Mississippian-age bedrock in St. Louis County and St. Louis City.



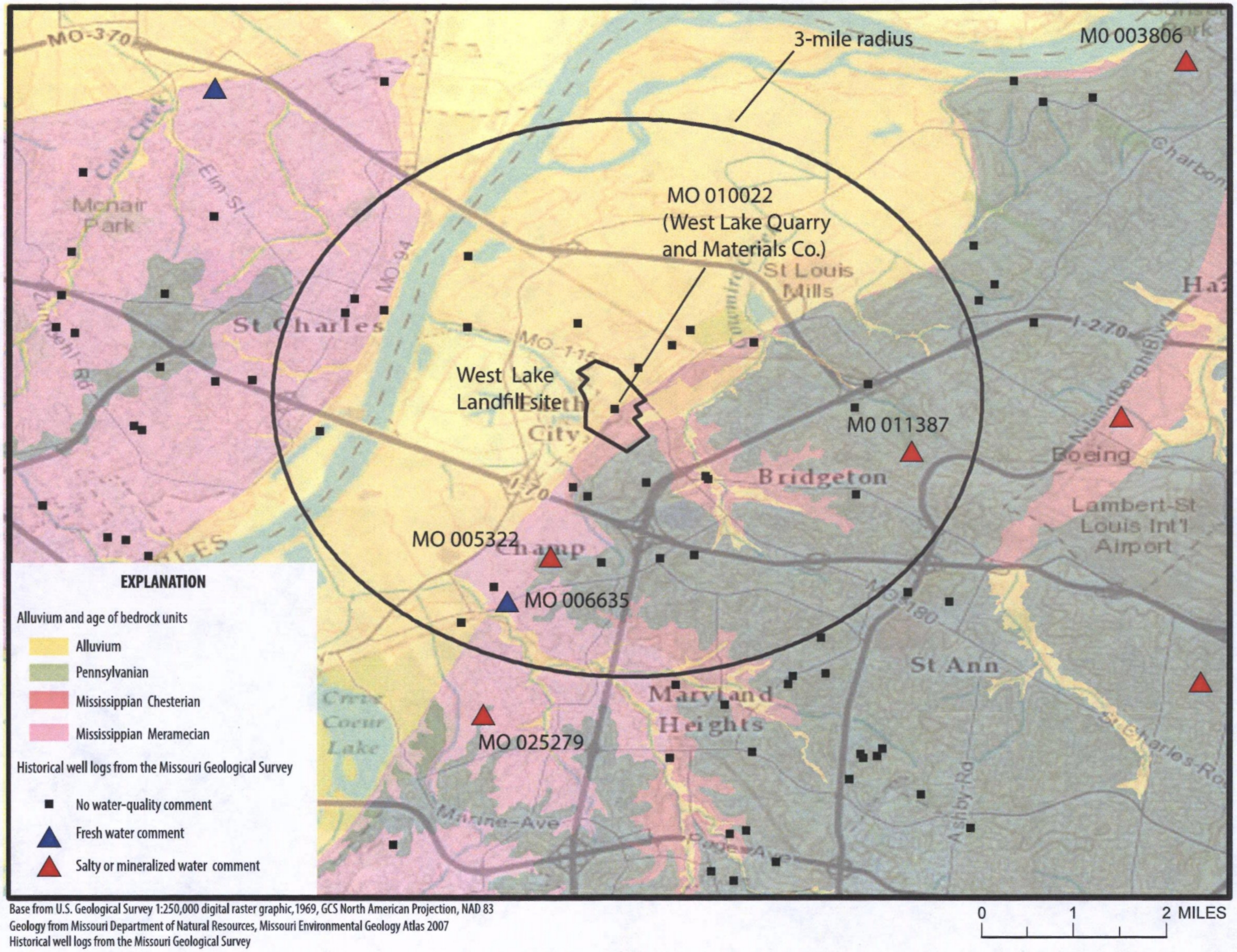
Base from U.S. Geological Survey aerial imagery, 1974
Operable Unit (OU) Areas, site boundary, and Radiologically Impacted Areas (RIM) from U.S. Environmental Protection Agency (EPA) digital data

Figure 2. 1974 aerial photograph of the West Lake Landfill site, former landfill areas, areas containing radiologically impacted material (RIM), and monitoring wells at the West Lake Landfill site.



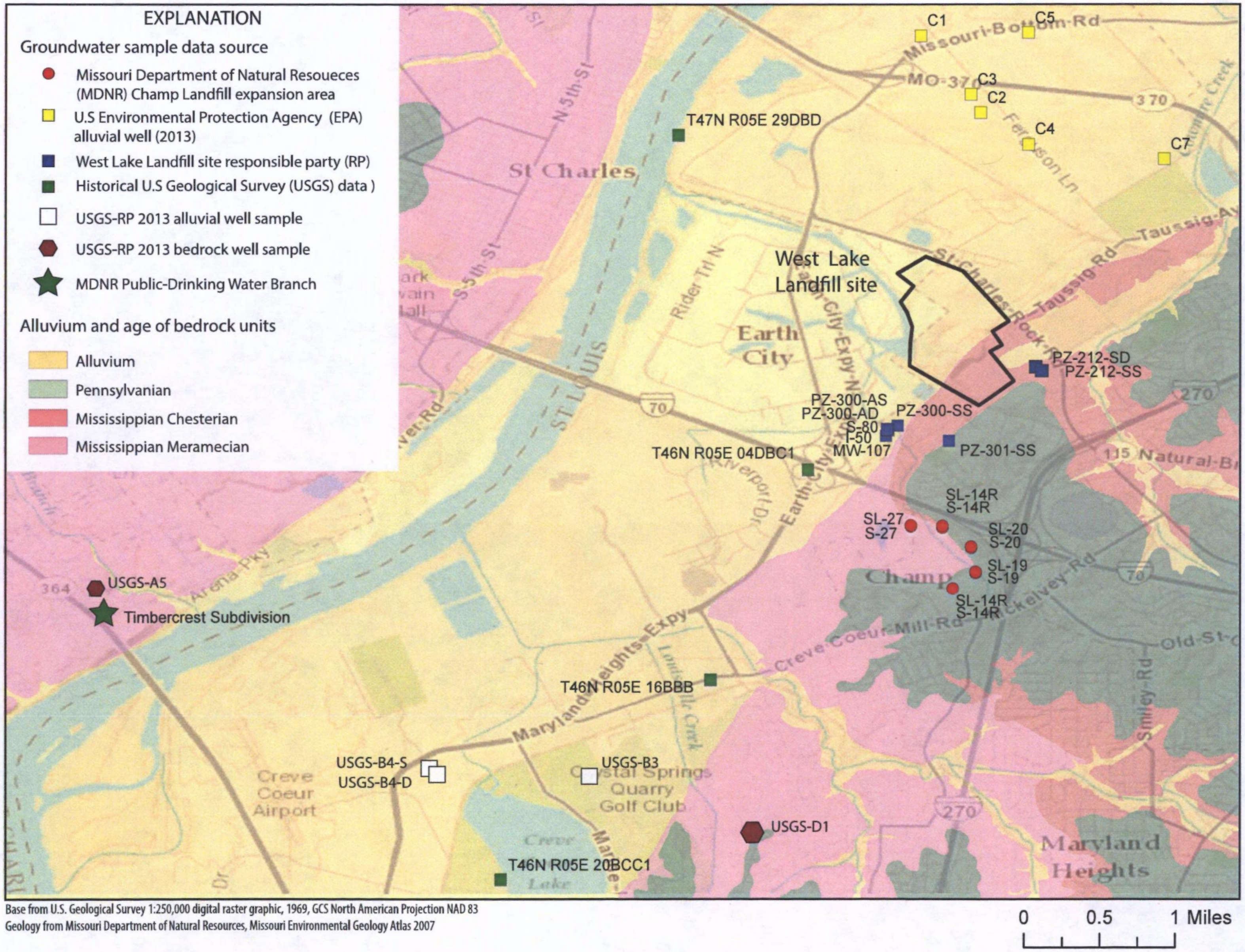
Base from U.S. Geological Survey 1:250,000 digital raster graphic, 1969, GCS North American Projection, NAD 83
Geologic structure, bedrock, and alluvium from Missouri Department of Natural Resources, Missouri Environmental Geology Atlas, 2007

Figure 3. Contours of chloride concentration in the Mississippian-age bedrock from Miller and others (1974) and chloride concentration in historical (1934–70) water samples from water wells analyzed by the Missouri Geological Survey (MGS). Historical MGS sample data pulled from the USGS National Water Information System (http://waterdata.usgs.gov/nwis). Note chloride contours and data values are from different sources and may not directly correspond to each other.



Base from U.S. Geological Survey 1:250,000 digital raster graphic, 1969, GCS North American Projection, NAD 83
Geology from Missouri Department of Natural Resources, Missouri Environmental Geology Atlas 2007
Historical well logs from the Missouri Geological Survey

Figure 4. Water wells with historical geologic logs drilled within a 3-mile radius of the West Lake Landfill site and geologic logs with water-quality comments.



Base from U.S. Geological Survey 1:250,000 digital raster graphic, 1969, GCS North American Projection NAD 83
Geology from Missouri Department of Natural Resources, Missouri Environmental Geology Atlas 2007

Figure 5. Location of background groundwater samples within 5 miles of the West Lake Landfill site.

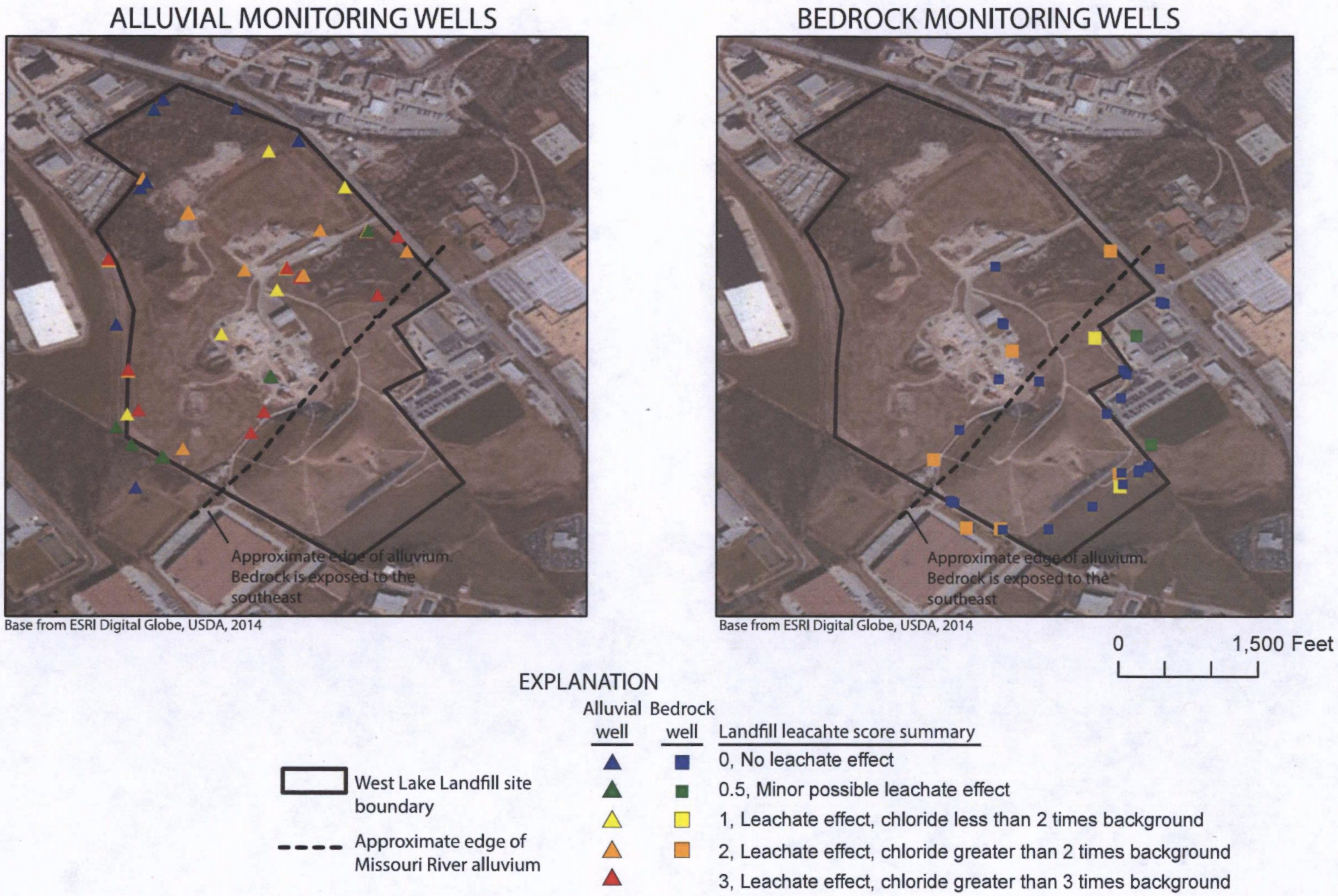


Figure 6. Monitoring wells at the West Lake Landfill site with landfill leachate effects. The presence of landfill leachate identified using concentrations of bromide (Br), chloride (Cl) and iodide (I).

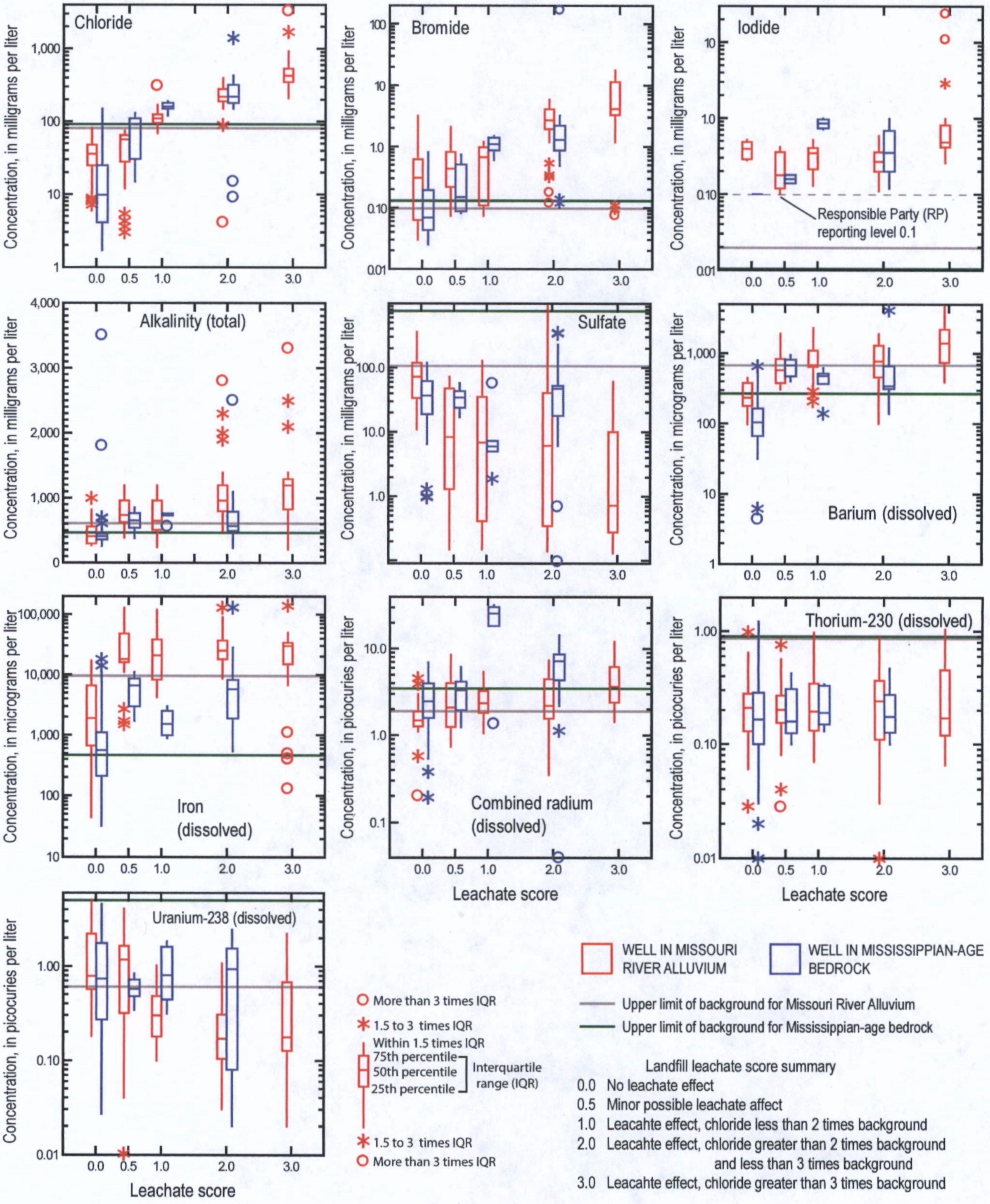


Figure 7. Boxplots of constituents in samples from monitoring wells at the West Lake Landfill site in relation to landfill leachate effects.

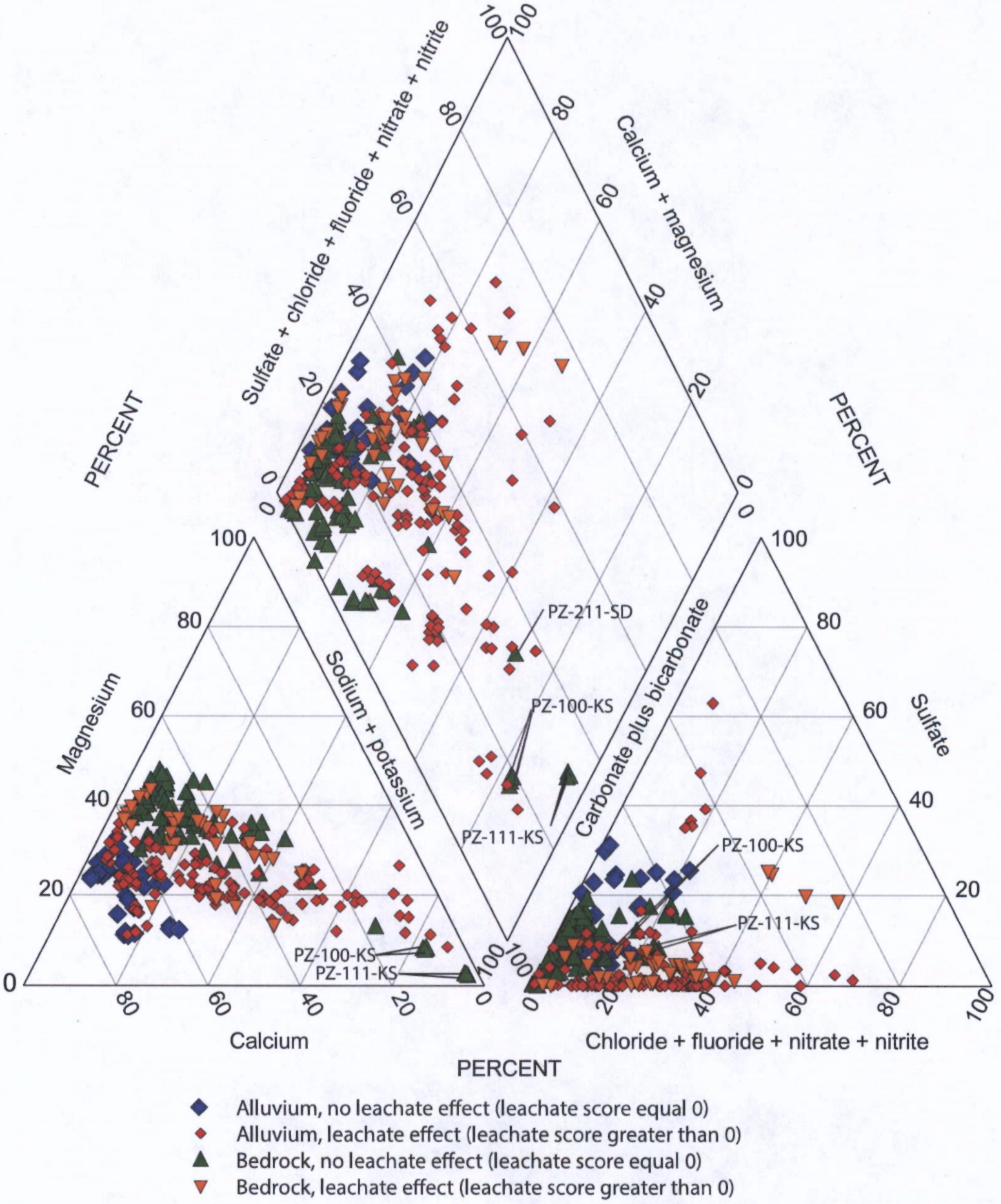


Figure 8. Trilinear diagram of major ions in groundwater samples from the West Lake Landfill site.

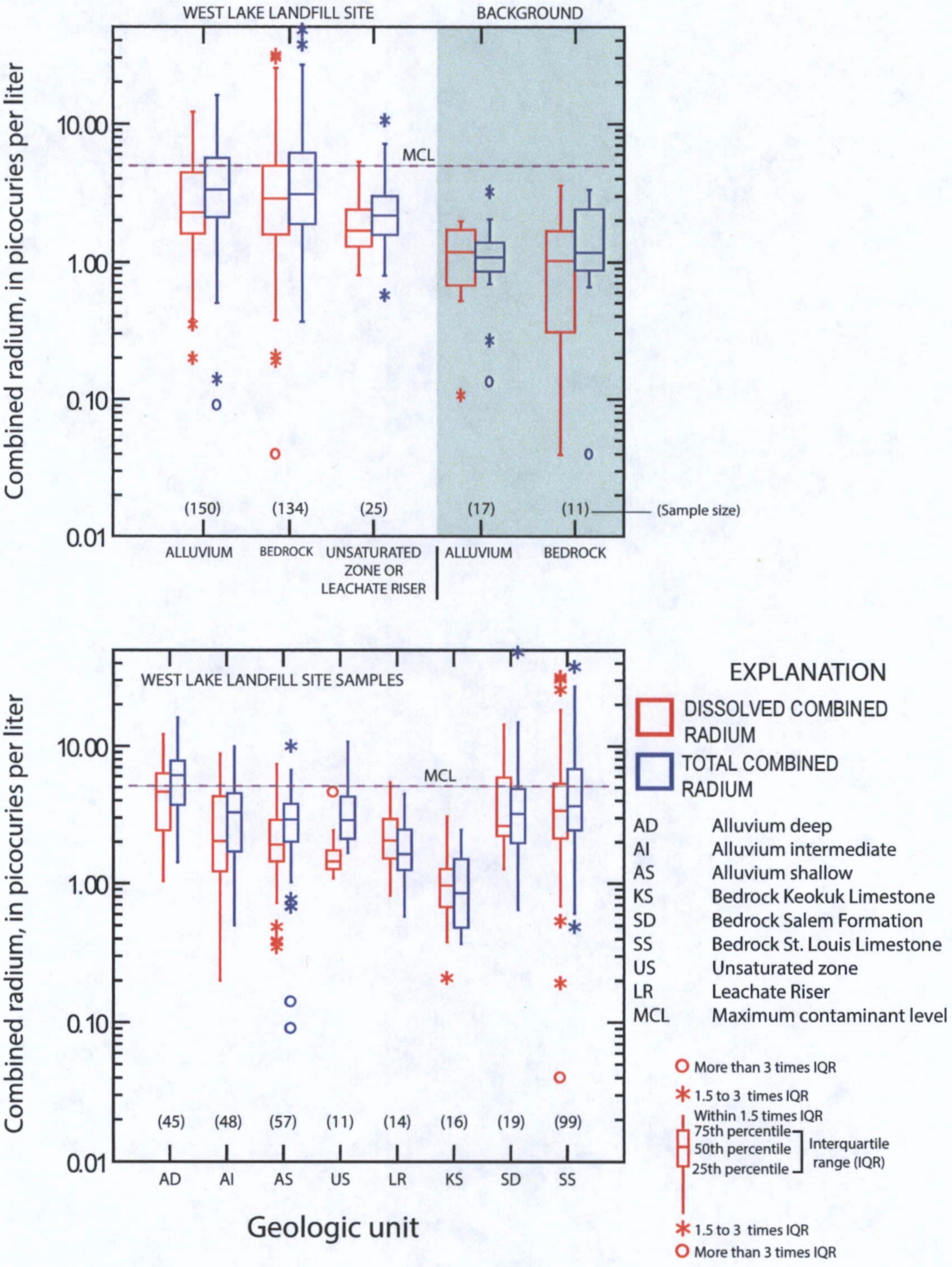


Figure 9. Box plots showing concentrations of dissolved and total combined radium in groundwater samples from monitoring wells in various geologic units at the West Lake Landfill site. Data from 2012-14.

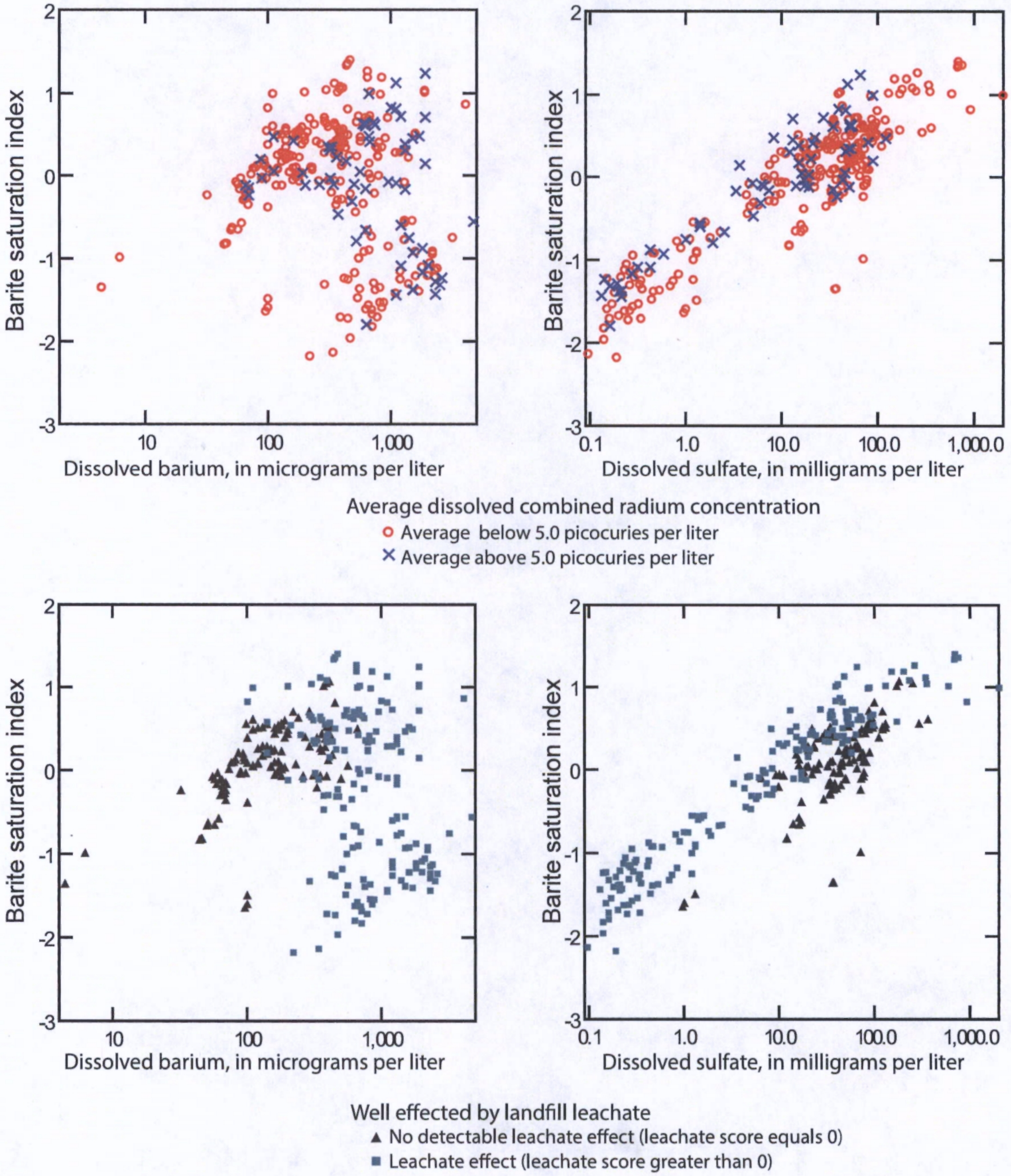


Figure 10. Barite saturation index in relation to dissolved barium (Ba) or sulfate (SO4) concentrations, the presence of dissolved combined radium above the maximum contaminant level (MCL), and the effects from landfill leachate in groundwater samples from the West Lake Landfill site (2012–14).

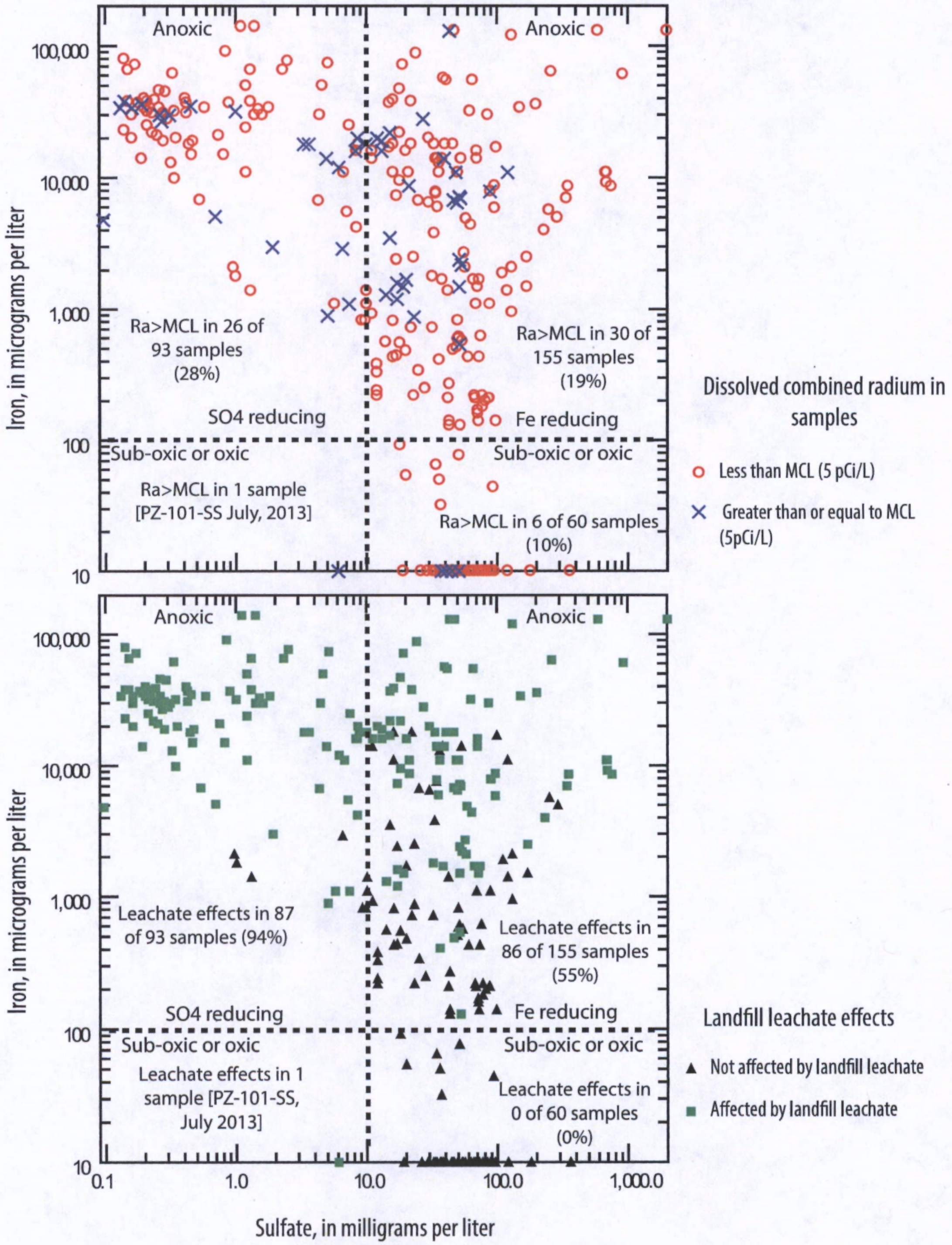
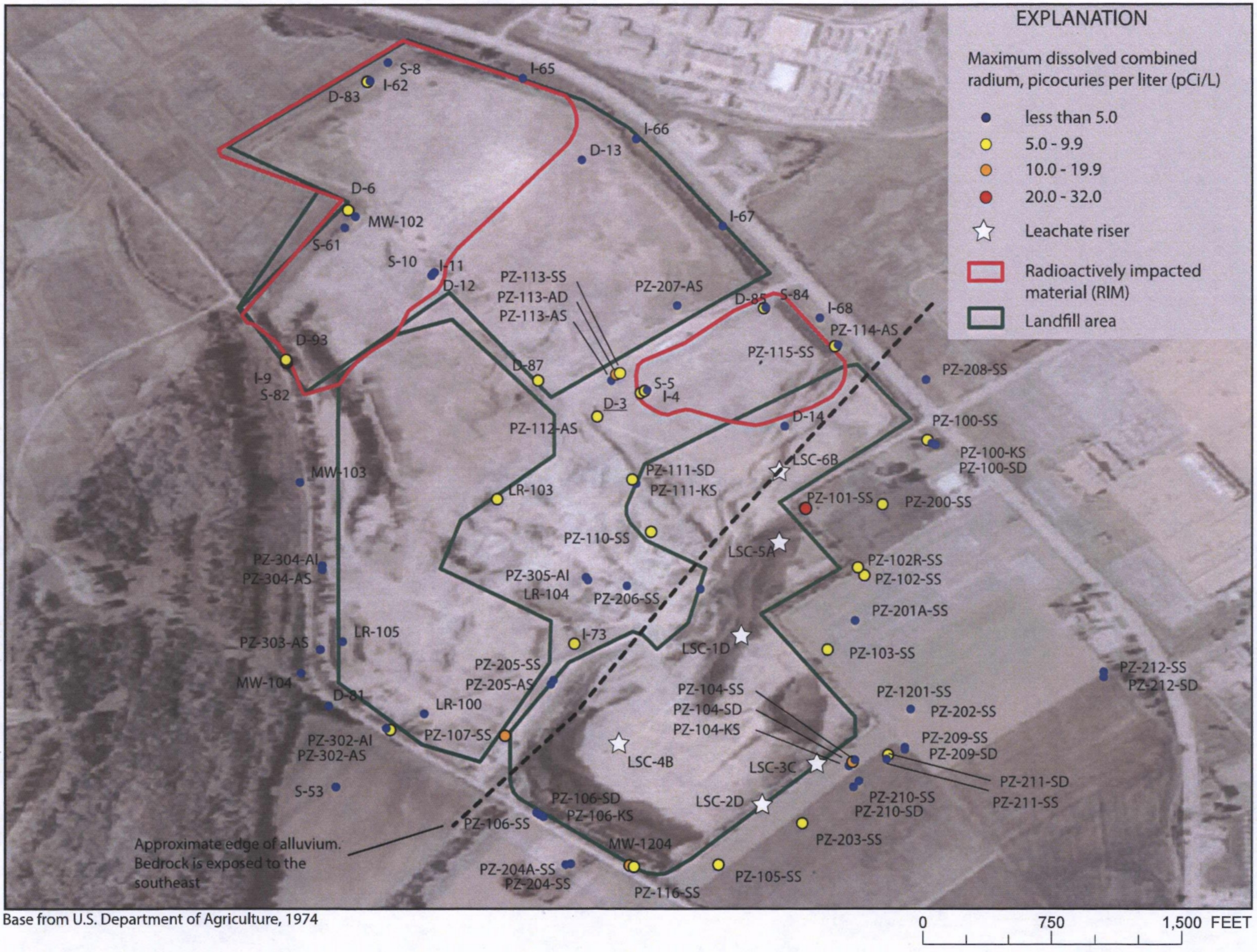
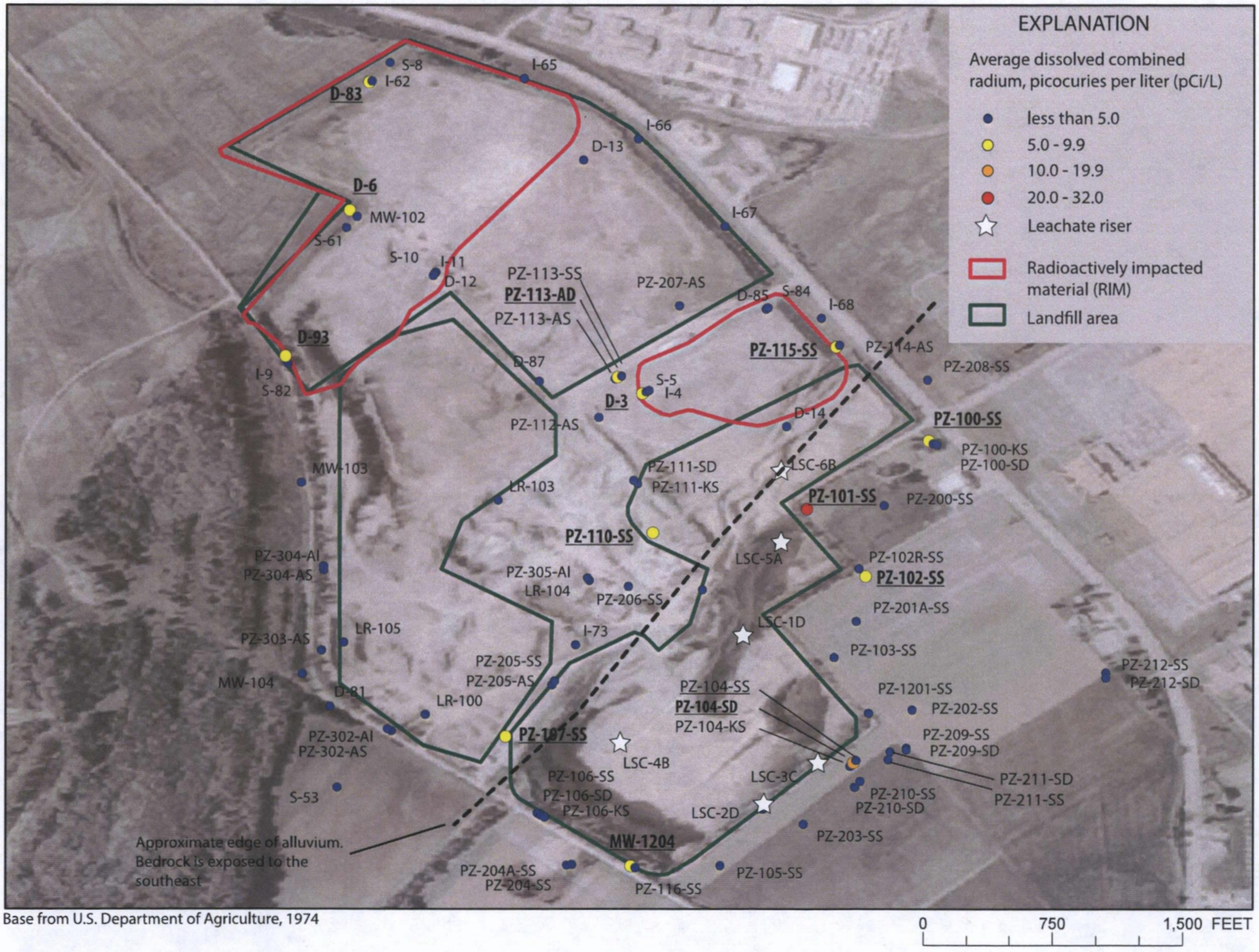


Figure 11. Relation between dissolved concentrations of sulfate (SO_4), iron (Fe), and dissolved combined radium above the maximum contaminant level (MCL) in groundwater samples from the West Lake Landfill site.



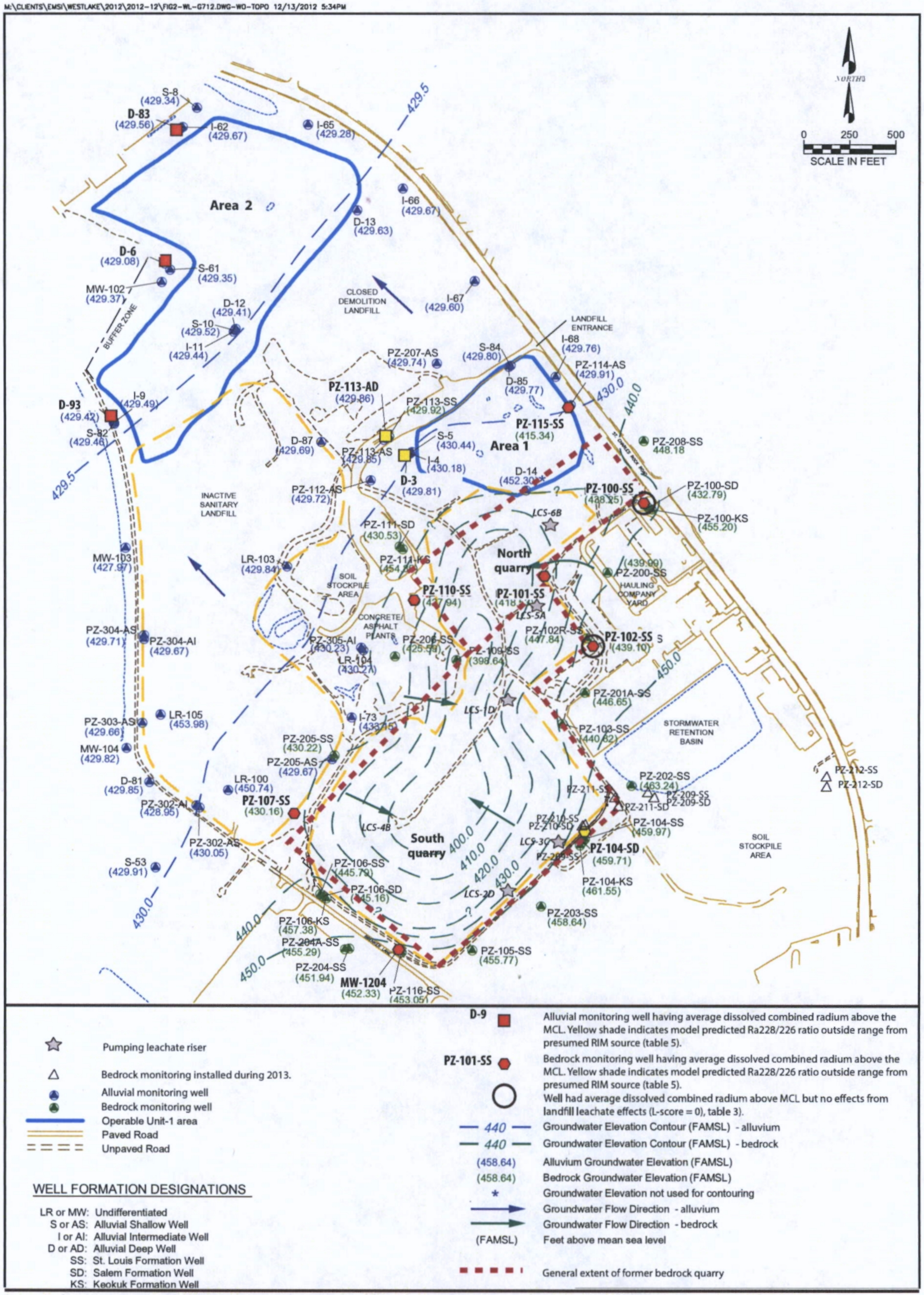
Base from U.S. Department of Agriculture, 1974

Figure 12. Maximum concentration of dissolved combined radium detected in groundwater samples from the West Lake Landfill site (2012–14).



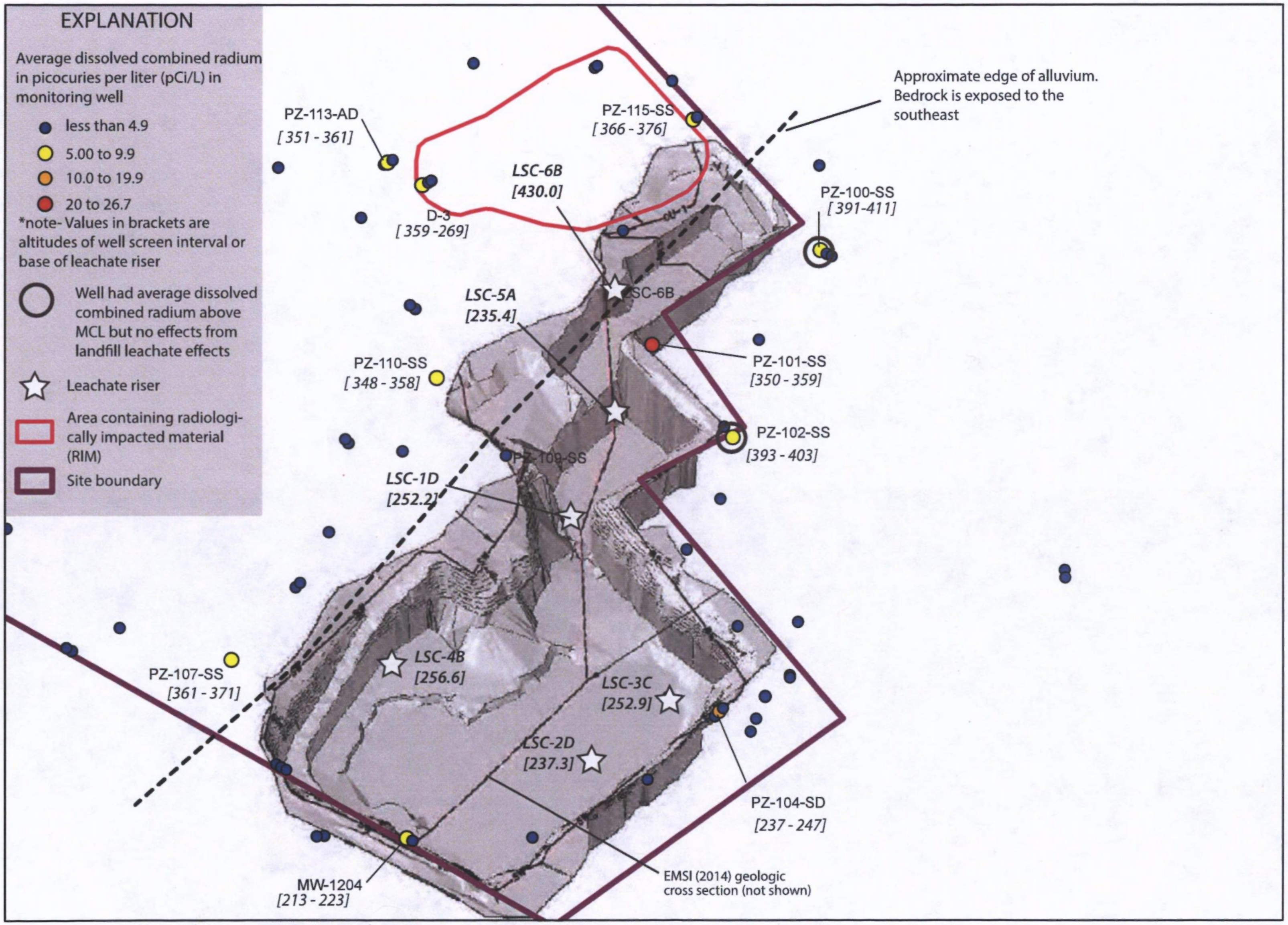
Base from U.S. Department of Agriculture, 1974

Figure 13. Average concentration of dissolved combined radium in groundwater samples from the West Lake Landfill site (2012–14). Wells with average dissolved combined radium values above the MCL underlined and bold-face.



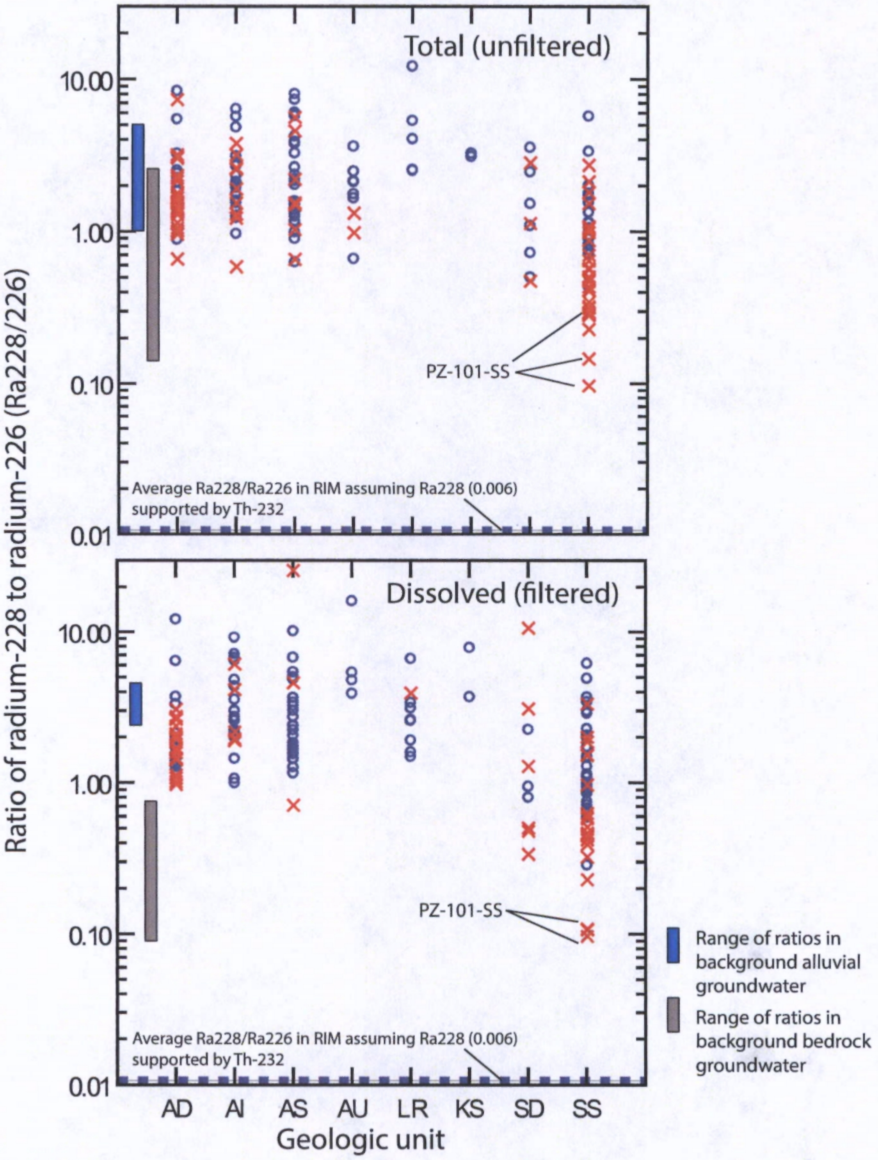
Base map and potentiometric contours from Engineering Management Solutions Inc., 2012

Figure 14. Monitoring wells with average dissolved combined radium above the maximum contaminant level (MCL) and degree of landfill leachate effects in those wells, and results of radium isotope mass balance in relation to water levels measured during July 2012.



Base map provided by Paul Rosasco, Environmental Management Support, Inc., written commun., 2014.

Figure 15. Approximate location of monitoring wells and leachate risers in the vicinity of the South and North quarry that have dissolved combined radium above the maximum contaminant level, and elevation of quarry floor and well screen intervals.

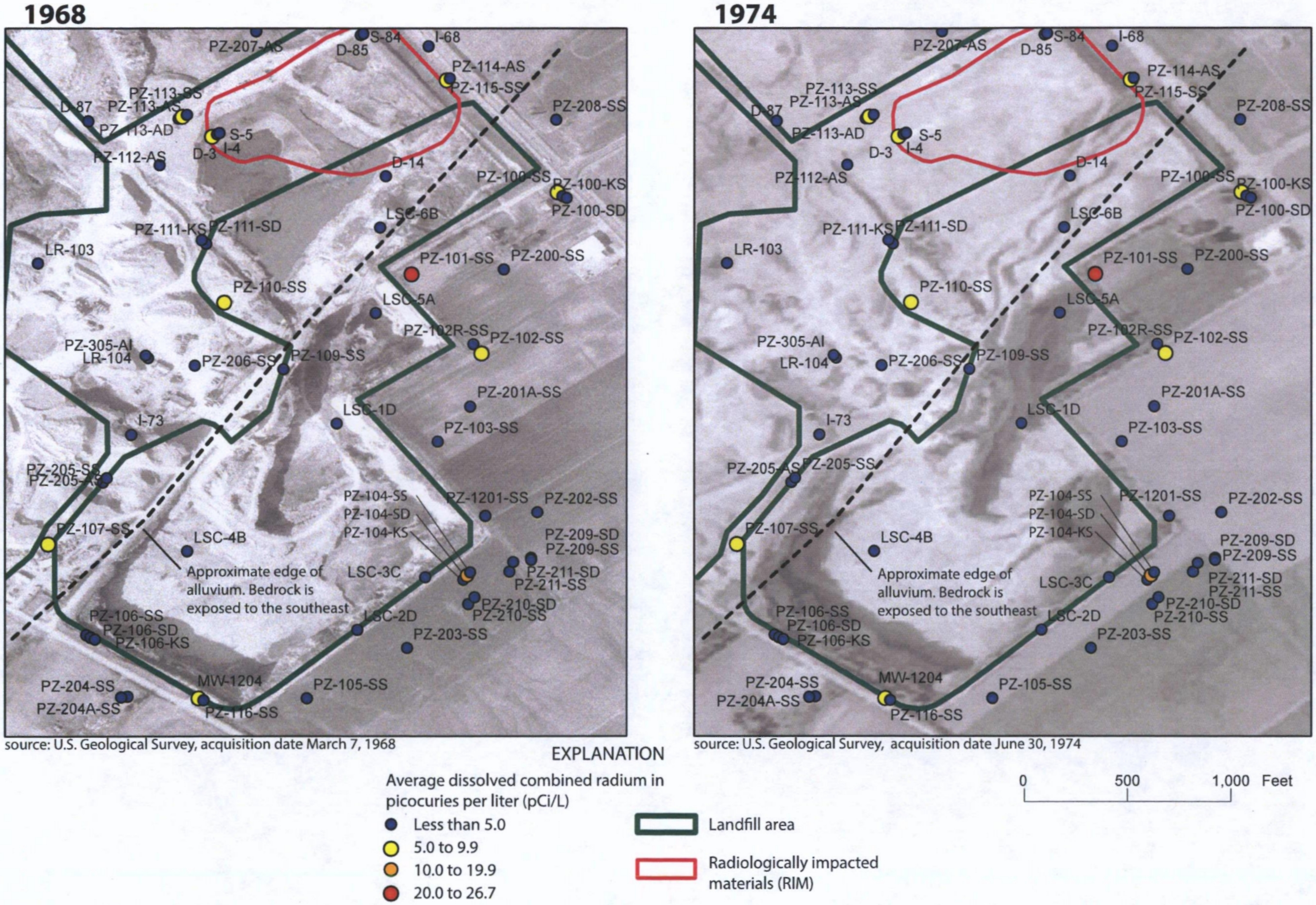


AD	Alluvium, deep	LR	Leachate Riser
AI	Alluvium, intermediate	KS	Bedrock, Keokuk Limestone
AS	Alluvium, shallow	SD	Bedrock, Salem Formation
AU	Unsaturated zone	SS	Bedrock, St. Louis Limestone

○ Combined radium below 5 pCi/L (picocuries per liter) maximum contaminant level (MCL)

× Combined radium above 5 pCi/L (picocuries per liter) maximum contaminant level (MCL)

Figure 16. Plots showing ratio of radium-228 to radium-226 (Ra228/226) in groundwater samples from monitoring wells completed in various geologic units at the West Lake Landfill site in relation to ratios in background groundwater samples and radiologically impacted material (RIM). Ratio of Ra228/226 in radiologically impacted material (RIM) based on sample data contained in McLaren and Hart Inc. (1996).



source: U.S. Geological Survey, acquisition date March 7, 1968

source: U.S. Geological Survey, acquisition date June 30, 1974

Figure 17. Average dissolved combined radium in monitoring wells near the North and South quarry areas at the West Lake Landfill site in relation to site features shown on 1968 and 1974 aerial photographs. Imagery obtained from U.S. Geological Survey (2014).

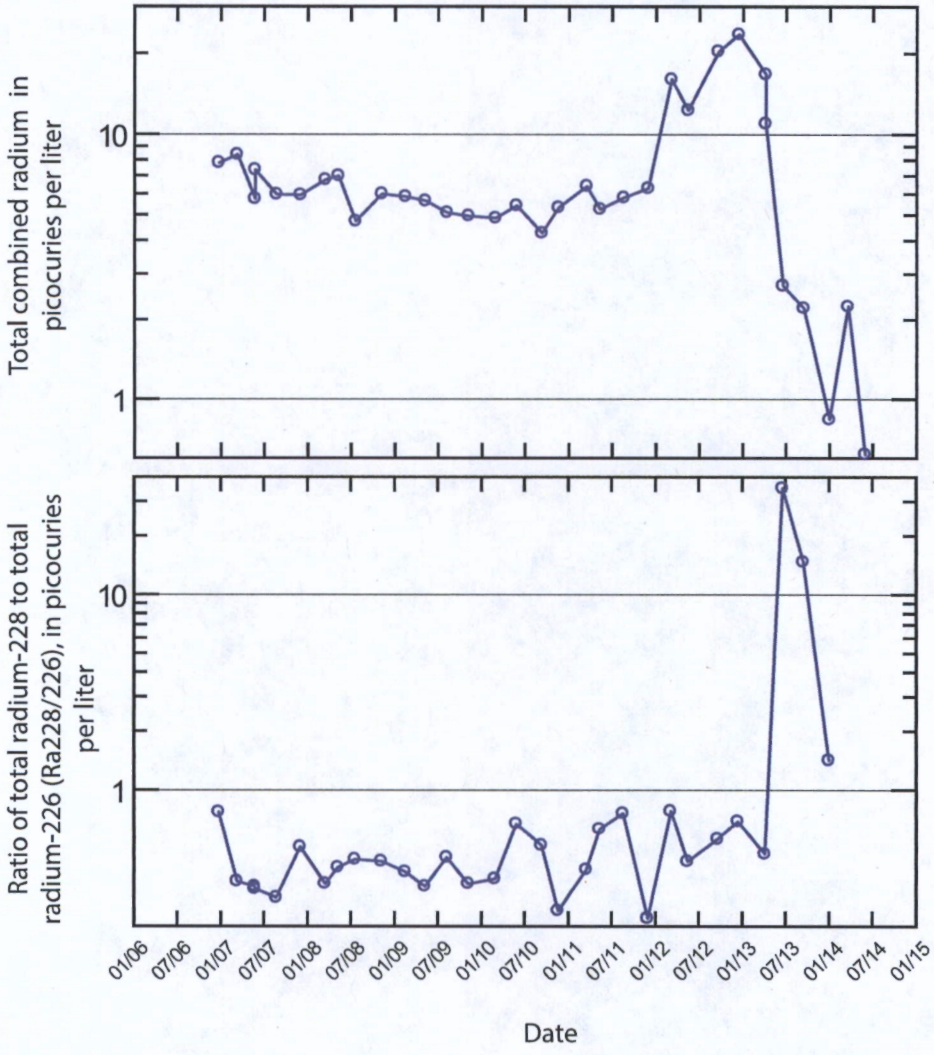


Figure 18. Concentrations of total combined radium and ratio of total radium-228 to radium-226 (Ra228/226) in samples from leachate discharged from the Bridgeton Landfill (2006–14).

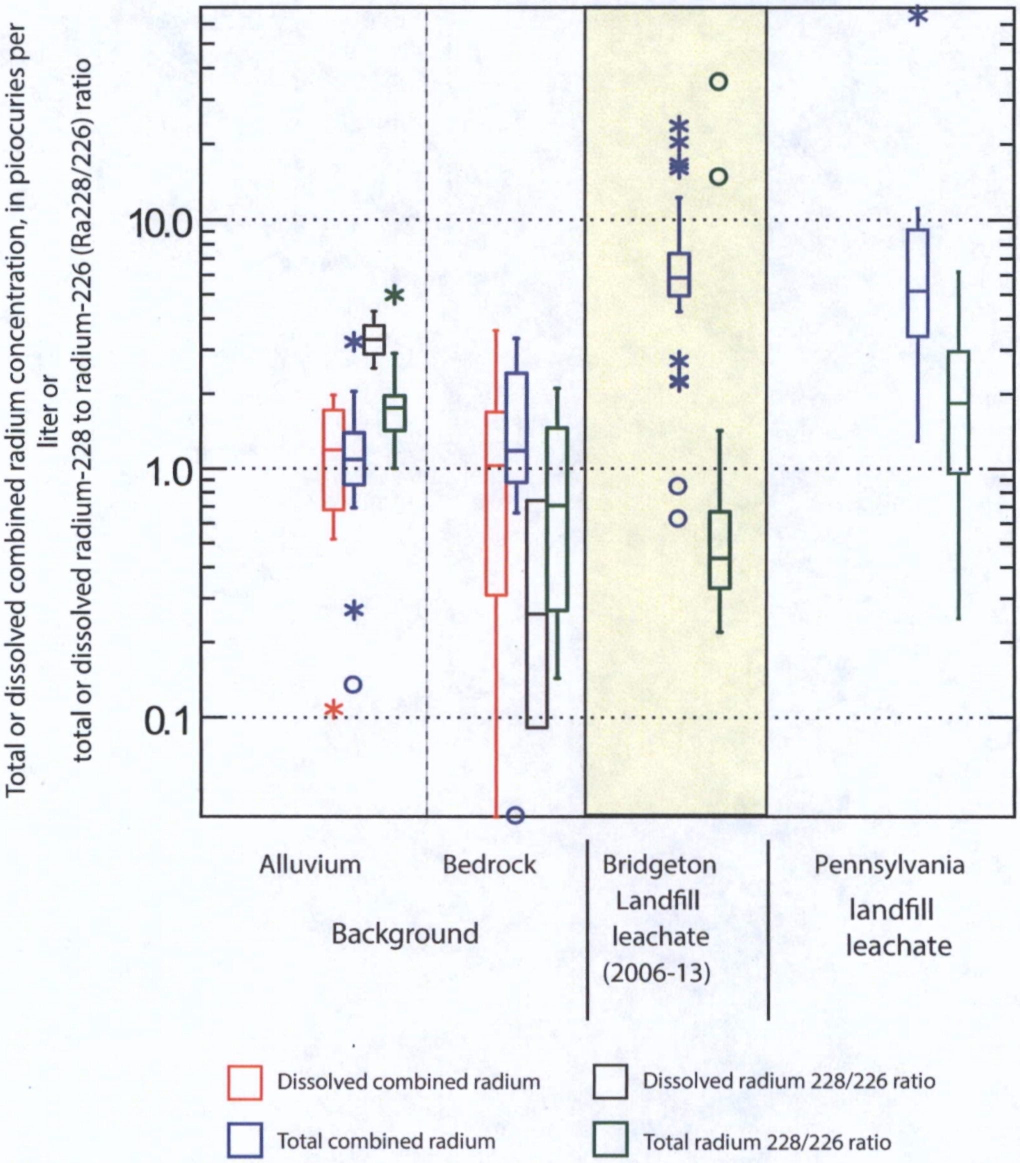


Figure 19. Comparison of dissolved and total combined radium and ratio of total and dissolved radium-228 to radium-226 (Ra228/226) in background groundwater samples to leachate pumped from the Bridgeton Landfill (2006–13) and selected Pennsylvania landfills. Pennsylvania leachate data from Civil and Environmental Consultants, Inc. (2005).

Table 1. Bedrock units, regional aquifer designations, and monitoring well designations at the West Lake Landfill site and Champ Landfill.

System	Series	Missouri Department of Natural Resources (DNR) Name	West lake Landfill designation	Champ Landfill designation	Geohydrologic unit or equivalent
Pennsylvanian	Missourian	Undifferentiated			Stratigraphic equivalent to Western Interior Plains confining system
	Desmoinesian	Undifferentiated			
	Atokan	Undifferentiated			
Mississippian	Chesterian	Ste. Genevieve Limestone			Stratigraphic equivalent to Springfield Plateau aquifer system
	Meramecian	St. Louis Limestone	SS	SL	
		Salem Limestone	SD	S	
		Warsaw Formation			
	Osagean	Keokuk Limestone[1]	KS		
		Burlington Limestone[1]			
		Fern Glen Formation			
	Kinderhookian	Chouteau Group			Ozark confining unit
Devonian	Upper	Bushberg Sandstone			
		Grassy Creek Shale			
Silurian	Unknown	Bainbridge Limestone			
Ordovician	Cincinnatian	Maquoketa Shale			
	Mohawkian	Kimmswick Limestone			Ozark aquifer
		Decorah Formation			
		Plattin Formation			
		Joachim Dolomite			
		St. Peter Sandstone			
	Ibexian	Powell Dolomite			
		Roubidoux Formation			
		Gasconade Dolomite			
Cambrian	Croixian	Potosi Dolomite			

[1] The Keokuk Limestone and Burlington Limestone often are undifferentiated in the St. Louis region.

Table 2. Estimates of upper limit of background concentrations for selected constituents in groundwater in the Missouri River alluvium and Mississippian-age bedrock in the vicinity of the West Lake Landfill site.

[RP, Responsible party; mg/L, milligram per liter; CaCO3, calcium carbonate; --, no data; ug/L, microgram per liter; pCi/L, picocuries per liter; Ra228/226, ratio of radium-228 to radium-226]

		Mississippian-age bedrock		Missouri River alluvium		Missouri River alluvium (no offsite RP wells)	
Constituent[1]	Unit	Number of samples	Upper limit of background	Number of samples	Upper limit of background	Number of samples	Upper limit of background
Calcium, dissolved	mg/L	148 (18)	250 (137.8)	12	174	7	160
Magnesium, dissolved	mg/L	153 (22)	94.7 (57.8)	12	60.8	7	31
Sodium, dissolved	mg/L	154	310	14	71.6	9	27.38
Potassium, dissolved	mg/L	13	8.5	10	6.1	5	6.1
Alkalinity as $CaCO_3$, total	mg/L	18	478	9	625	9	625
Chloride, dissolved	mg/L	158	51.4 (91 RP)	17	236	11	79.5
Sulfate, dissolved	mg/L	146 (15)	878 (525)	14	109	9	103
Fluoride, dissolved	mg/L	23	4.4	3	0.3	3	0.314
Iodide, dissolved	mg/L	2	0.0074	2	0.0218	2	0.0218
Bromide, dissolved	mg/L	6	0.12	4	0.09	4	0.091
Nitrate plus nitrate (NO_2+NO_3), dissolved	mg/L	147	0.3	8	0.1	3	0.000
Ammonia (NH_3), dissolved	mg/L	131	0.69	0	--	0	--
Arsenic, dissolved	ug/L	5	2.0	3	4.1	3	4.1
Barium, dissolved	ug/L	149 (14)	271 (229)	6	670	6	670
Boron, dissolved	ug/L	141 (8)	2,200 (636)	4	93	4	93
Iron, dissolved	ug/L	144 (13)	8,500 (581)	6	9,600	6	9,600.00
Lead, dissolved	ug/L	141	8.1	4	1.9	4	1.9
Lithium, dissolved	ug/L	2	35	2	39.7	2	39.7
Manganese, dissolved	ug/L	142 (10)	744 (280)	6	740	6	740
Nickel, dissolved	ug/L	97	9.6	2	4.4	2	4.4
Strontium, dissolved	ug/L	6	4,200	4	803	4	803
Zinc, dissolved	ug/L	141	319	5	110	5	110
Chloride/Bromide ratio (range)	mass ratio	6	464-1,100	4	442 - 1,917	--	--
Iodide/sodium ratio (range)	mass ratio	2	0.00018-0.00024	2	0.00041-0.0008	--	--
Radium-226, dissolved	pCi/L	11	3.24	11	0.64	--	--
Radium-228, dissolved	pCi/L	9	0.65	11	1.45	--	--
Thorium-228, dissolved	pCi/L	11	0.15	11	0.20	--	--
Thorium-230, dissolved	pCi/L	9	0.893	11	0.909	--	--
Thorium-232, dissolved	pCi/L	11	0.11	11	0.21	--	--

Table 2. Estimates of upper limit of background concentrations for selected constituents in groundwater in the Missouri River alluvium and Mississippian-age bedrock in the vicinity of the West Lake Landfill site.

[RP, Responsible party; mg/L, milligram per liter; CaCO3, calcium carbonate; --, no data; ug/L, microgram per liter; pCi/L, picocuries per liter; Ra228/226, ratio of radium-228 to radium-226]

		Mississippian-age bedrock		Missouri River alluvium		Missouri River alluvium (no offsite RP wells)	
Constituent[1]	Unit	Number of samples	Upper limit of background	Number of samples	Upper limit of background	Number of samples	Upper limit of background
Uranium-234, dissolved	pCi/L	10	13.48	11	0.87	--	--
Uranium-235, dissolved	pCi/L	11	0.44	11	0.26	--	--
Uranium-238, dissolved	pCi/L	11	5.064	7	0.63	--	--
Radium-226, total	pCi/L	9	3.324	17	0.93	--	--
Radium-228, total	pCi/L	9	1.04	17	2.20	--	--
Thorium-228, total	pCi/L	11	0.36	17	0.65	--	--
Thorium-230, total	pCi/L	9	1.366	17	1.31	--	--
Thorium-232, total	pCi/L	11	0.16	17	0.675	--	--
Uranium-234, total	pCi/L	10	16.4655	17	2.86	--	--
Uranium-235, total	pCi/L	11	0.56	12	0.28	--	--
Uranium-238, total	pCi/L	11	4.642	17	1.78	--	--
Combined radium, dissolved	pCi/L	11	3.56	11	1.98	--	--
Combined radium, total	pCi/L	7	3.34	17	2.81	--	--
Ra228/226 ratio, dissolved	ratio	2	0.74	3	4.28	--	--
Ra228/226 ratio, total	ratio	4	2.11	9	4.98	--	--

[1] Background concentrations for bedrock include results from unfiltered samples from Champ landfill, and values may be influenced by aquifer particulates. Values in parentheses exclude data from Champ landfill.

Table 3. Summary of average concentrations of selected constituents in monitoring wells at the West Lake Landfill site (2012–14); frequency that dissolved chloride, bromide, iodide, combined radium, or total combined radium exceeded background; and assigned leachate score.

[Leachate score of 1 or greater indicates that well is affected by landfill leachate with scores of 3 indicate average chloride more than 3 times background, leachate scores of 2 indicate average chloride more than twice background. Leachate score of 0.5 indicate possible effects from landfill leachate. Concentrations of combined dissolved or total radium or Ra228/226 ratio not used in determining effects of landfill leachate. Wells PZ-212-SD and PZ-212-SS are considered background and not included. Selected columns shadded for clarity.

MCL, maximum contaminant level; Ra, radium; pCi/L, picocuries per liter; Cl, chloride; mg/L, milligrams per liter; Ra228/226, ratio of radium-228 to radium-226; ug/L, micrograms per liter; Br, bromide; I, iodide; Na, sodium; ALL, alluvium; BED, bedrock; SS, St. Louis Limestone; SD, Salem Formation; KS, Keokuk Limestone; AS, shallow alluvium; AI, intermediate alluvium; AD, deep alluvium; LR, leachate riser; US, unsaturated zone; --, no data]

Well Number and geologic unit			Average of all samples from monitoring well						Number of samples that exceeded background (shaded column indicates constituent used to determine leachate effects)						Number of samples combined Ra exceeded MCL (5 pCi/L)		
Well Number	Media	Unit	Cl (mg/L)	Total combined Ra (pCi/L)	Dissolved combined Ra (pCi/L)	Dissolved Ra228/226	Fuel-VOC (sum in ug/L)	Cl-VOC (sum in ug/L)	Cl	Br	I	Na	Combined Ra	Total combined Ra	Dissolved	Total	Leachate score (L_score)
Wells with average dissolved combined radium (Ra) above 5.0 pCi/L																	
D-3	ALL	AD	420	8.22	7.40	1.63	11.4	0.0	4	4	4	4	4	4	3	4	3
D-6	ALL	AD	183	7.15	5.98	1.39	23.5	1.3	4	4	4	4	4	4	3	4	2
D-83	ALL	AD	62.0	7.11	6.62	1.40	8.4	0.0	0	3	0	4	4	4	4	4	0.5
D-93	ALL	AD	343	5.53	5.63	1.62	6.2	188.5	4	4	4	4	4	4	3	2	3
MW-1204	BED	SS	426	14.8	5.1	0.60	4,158	0.0	2	2	2	2	3	4	1	4	2
PZ-100-SS	BED	SS	4.0	5.69	5.1	0.91	2.9	0.0	0	0	0	0	4	4	2	3	0
PZ-101-SS	BED	SS	170	20.8	26.7	0.10	23.7	0.0	4	4	4	3	4	4	4	4	1
PZ-102-SS	BED	SS	5.1	13.0	5.4	0.57	0.0	0.0	0	0	0	0	4	4	3	4	0
PZ-104-SD	BED	SD	190	7.11	11.1	0.65	5,196	0.0	4	4	3	4	4	4	4	3	2
PZ-107-SS	BED	SS	300	12.1	8.7	0.36	6.7	0.2	3	4	4	4	4	4	4	4	2
PZ-110-SS	BED	SS	190	6.25	6.3	0.80	0.0	12.7	4	4	4	4	4	4	3	4	2
PZ-113-AD	ALL	AD	463	9.31	8.44	2.14	4.1	0.0	4	4	4	4	4	4	3	4	3
PZ-115-SS	BED	SS	205	7.98	7.4	0.18	1.7	0.0	4	4	3	0	4	4	4	4	2
Wells with average dissolved combined radium (Ra) below 5.0 pCi/L																	
D-12	ALL	AD	245	1.94	2.11	4.60	9.9	4.4	4	4	4	4	2	1	0	0	2
D-13	ALL	AD	143	4.03	2.88	1.78	41.9	0.0	3	1	0	3	4	3	0	1	1
D-14	ALL	AD	467	4.35	2.84	1.87	286	1.5	3	3	3	3	3	4	0	2	3
D-81	ALL	AD	21.3	3.54	2.27	5.59	0.0	3.4	0	4	0	2	3	2	0	1	0.5
D-85	ALL	AD	241	11.4	3.82	2.05	238	0.2	2	2	3	4	4	4	1	4	2
D-87	ALL	AD	270	5.50	4.75	2.03	0.6	0.7	4	4	4	4	3	4	3	3	2
I-11	ALL	AI	203	3.84	3.68	3.12	22.0	8.1	4	4	4	4	4	4	0	0	2
I-4	ALL	AI	168	5.68	3.52	4.16	229	0.2	4	4	3	4	2	3	2	3	2
I-62	ALL	AI	52.5	1.58	1.61	6.40	2.2	0.0	0	0	0	0	1	0	0	0	0

Table 3. Summary of average concentrations of selected constituents in monitoring wells at the West Lake Landfill site (2012–14); frequency that dissolved chloride, bromide, iodide, combined radium, or total combined radium exceeded background; and assigned leachate score.

[Leachate score of 1 or greater indicates that well is affected by landfill leachate with scores of 3 indicate average chloride more than 3 times background, leachate scores of 2 indicate average chloride more than twice background. Leachate score of 0.5 indicate possible effects from landfill leachate. Concentrations of combined dissolved or total radium or Ra228/226 ratio not used in determining effects of landfill leachate. Wells PZ-212-SD and PZ-212-SS are considered background and not included. Selected columns shadded for clarity.

MCL, maximum contaminant level; Ra, radium; pCi/L, picocuries per liter; Cl, chloride; mg/L, milligrams per liter; Ra228/226, ratio of radium-228 to radium-226; ug/L, micrograms per liter; Br, bromide; I, iodide; Na, sodium; ALL, alluvium; BED, bedrock; SS, St. Louis Limestone; SD, Salem Formation; KS, Keokuk Limestone; AS, shallow alluvium; AI, intermediate alluvium; AD, deep alluvium; LR, leachate riser; US, unsaturated zone; --, no data]

Well Number and geologic unit			Average of all samples from monitoring well						Number of samples that exceeded background (shaded column indicates constituent used to determine leachate effects)						Number of samples combined Ra exceeded MCL (5 pCi/L)		
Well Number	Media	Unit	Cl (mg/L)	Total combined Ra (pCi/L)	Dissolved combined Ra (pCi/L)	Dissolved Ra228/226	Fuel-VOC (sum in ug/L)	Cl-VOC (sum in ug/L)	Cl	Br	I	Na	Combined Ra	Total combined Ra	Dissolved	Total	Leachate score (L_score)
I-65	ALL	AI	57.3	2.21	1.00	--	0.0	0.0	0	0	0	4	0	1	0	0	0
I-66	ALL	AI	64	1.29	1.18	7.10	0.3	0.0	1	0	0	4	0	0	0	0	0
I-67	ALL	AI	130	1.62	2.14	5.92	0.6	0.0	4	2	0	4	1	0	0	0	1
I-68	ALL	AI	523	4.86	3.03	3.62	0.0	0.0	3	2	0	4	4	4	0	2	3
I-73	ALL	AI	1,463	4.85	4.35	1.45	467	33.1	4	4	4	4	3	3	1	1	3
I-9	ALL	AI	315	5.26	4.55	2.73	0.7	15.1	4	4	4	4	4	4	2	3	2
LR-100	ALL	LR	178	1.06	1.36	2.61	389.7	0.0	4	4	4	4	1	0	0	0	2
LR-103	ALL	LR	135	2.49	3.33	2.60	1.1	0.9	4	4	4	4	4	1	1	0	1
LR-104	ALL	LR	38.0	2.75	2.24	4.18	0.4	1.7	0	4	4	0	2	1	0	0	0.5
LR-105	ALL	LR	795	1.45	2.47	1.58	610	0.0	2	2	2	2	2	0	0	0	3
MW-102	ALL	US	10.2	2.81	1.57	4.76	0.0	0.0	0	0	0	0	0	2	0	0	0
MW-103	ALL	US	26.3	4.48	2.16	16.04	0.3	0.0	0	3	0	0	1	2	0	1	0
MW-104	ALL	US	4.1	4.15	1.57	4.62	1.0	0.3	0	4	0	0	1	2	0	1	0.5
PZ-100-KS	BED	KS	54.3	0.76	1.0	3.69	0.0	0.0	0	4	0	4	0	0	0	0	0
PZ-100-SD	BED	SD	1.7	3.14	2.7	--	1.0	0.0	0	0	0	0	0	2	0	0	0
PZ-102R-SS	BED	SS	8.5	4.25	3.8	0.80	0.4	0.0	0	0	0	0	2	4	1	0	0
PZ-103-SS	BED	SS	6.8	10.7	4.9	0.66	378	0.0	0	0	0	2	4	4	2	3	0
PZ-104-KS	BED	KS	24.0	0.95	0.7	--	0.0	0.0	0	1	0	0	0	0	0	0	0
PZ-104-SS	BED	SS	4.2	2.97	2.8	1.44	7,240	0.0	0	0	0	0	0	1	0	0	0
PZ-105-SS	BED	SS	86.8	2.97	3.5	1.52	0.0	0.0	1	0	0	0	1	1	1	0	0
PZ-106-KS	BED	KS	13.5	1.18	1.5	7.80	0.0	0.0	0	0	0	0	0	0	0	0	0
PZ-106-SD	BED	SD	11.8	1.94	2.0	2.24	0.0	0.0	0	1	0	0	0	0	0	0	0
PZ-106-SS	BED	SS	21.8	4.96	3.4	3.42	0.0	0.0	0	0	0	0	3	4	0	2	0
PZ-109-SS	BED	SS	3.8	3.49	3.6	0.88	0.0	0.0	0	0	0	0	2	1	0	1	0

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[Leachate score of 1 or greater indicates that well is affected by landfill leachate with scores of 3 indicate average chloride more than 3 times background, leachate scores of 2 indicate average chloride more than twice background. Leachate score of 0.5 indicate possible effects from landfill leachate. Concentrations of combined dissolved or total radium or Ra228/226 ratio not used in determining effects of landfill leachate. Wells PZ-212-SD and PZ-212-SS are considered background and not included. Selected columns shadded for clarity.

MCL, maximum contaminant level; Ra, radium; pCi/L, picocuries per liter; Cl, chloride; mg/L, milligrams per liter; Ra228/226, ratio of radium-228 to radium-226; ug/L, micrograms per liter; Br, bromide; I, iodide; Na, sodium; ALL, alluvium; BED, bedrock; SS, St. Louis Limestone; SD, Salem Formation; KS, Keokuk Limestone; AS, shallow alluvium; AI, intermediate alluvium; AD, deep alluvium; LR, leachate riser; US, unsaturated zone; --, no data]

Well Number and geologic unit			Average of all samples from monitoring well						Number of samples that exceeded background (shaded column indicates constituent used to determine leachate effects)						Number of samples combined Ra exceeded MCL (5 pCi/L)		
Well Number	Media	Unit	Cl (mg/L)	Total combined Ra (pCi/L)	Dissolved combined Ra (pCi/L)	Dissolved Ra228/226	Fuel-VOC (sum in ug/L)	Cl-VOC (sum in ug/L)	Cl	Br	I	Na	Combined Ra	Total combined Ra	Dissolved	Total	Leachate score (L_score)
PZ-111-KS	BED	KS	145	1.23	0.9	--	0.0	0.0	4	4	0	4	0	0	0	0	0[a]
PZ-111-SD	BED	SD	9.6	2.70	3.5	1.62	0.0	0.0	0	0	0	0	1	1	1	0	0
PZ-112-AS	ALL	AS	118	4.73	3.54	1.69	13,081	0.6	4	4	3	4	3	4	1	2	1
PZ-113-AS	ALL	AS	99	2.17	1.88	2.51	8.5	0.0	4	4	4	4	0	1	0	0	1
PZ-113-SS	BED	SS	10.7	4.80	4.6	1.14	0.0	0.0	0	0	0	0	4	3	1	2	0
PZ-114-AS	ALL	AS	245	1.38	1.28	2.22	239.9	0.5	4	4	2	4	1	0	0	0	2
PZ-116-SS	BED	SS	3.8	1.06	2.3	5.52	0.0	0.0	0	0	0	0	1	0	1	0	0
PZ-200-SS	BED	SS	100	5.38	4.5	0.85	1.2	0.0	3	2	0	0	2	3	1	2	0.5
PZ-201A-SS	BED	SS	3.9	1.57	1.3	--	4.6	0.0	0	0	0	0	0	0	0	0	0
PZ-202-SS	BED	SS	56.3	2.81	2.2	2.44	69.0	0.0	1	2	2	0	0	1	0	0	0.5
PZ-203-SS	BED	SS	4.7	3.08	2.9	1.87	0.3	0.0	0	0	0	0	1	2	1	0	0
PZ-204A-SS	BED	SS	408	3.21	2.2	1.55	30.6	0.0	4	3	2	4	0	1	0	1	2
PZ-204-SS	BED	SS	2.9	1.76	1.6	--	0.0	0.0	0	0	0	0	0	0	0	0	0
PZ-205-AS	ALL	AS	340	3.00	2.23	1.82	5,775	0.2	4	4	4	4	3	2	0	0	3
PZ-205-SS	BED	SS	32.0	2.94	2.4	1.08	0.0	0.0	0	3	0	0	0	1	0	0	0
PZ-206-SS	BED	SS	25.0	2.91	2.3	1.40	0.0	0.2	0	4	0	0	0	1	0	0	0
PZ-207-AS	ALL	AS	220	2.60	1.91	2.38	105	0.0	4	4	4	4	2	1	0	0	2
PZ-208-SS	BED	SS	84.0	2.21	1.9	2.56	0.0	0.7	1	0	0	0	0	0	0	0	0
PZ-209-SD[b]	BED	SD	7.2	7.78	1.1	--	0.8	0.0	0	0	0	1	0	1	0	0	0
PZ-209-SS[b]	BED	SS	7.0	1.64	1.5	--	0.0	0.0	0	0	0	0	0	0	0	0	0
PZ-210-SD[b]	BED	SD	140	0.72	1.1	--	45.3	0.0	2	2	2	2	0	0	0	0	1
PZ-210-SS[b]	BED	SS	14.5	0.47	1.0	--	1.0	0.0	0	2	0	0	0	0	0	0	0
PZ-211-SD[b]	BED	SD	32.5	24.6	3.4	10.66	0.0	0.0	0	0	0	1	1	1	0	0	0
PZ-211-SS[b]	BED	SS	5.5	1.18	0.7	--	2.0	0.0	0	0	0	0	0	0	0	0	0

Table 3. Summary of average concentrations of selected constituents in monitoring wells at the West Lake Landfill site (2012–14); frequency that dissolved chloride, bromide, iodide, combined radium, or total combined radium exceeded background; and assigned leachate score.

[Leachate score of 1 or greater indicates that well is affected by landfill leachate with scores of 3 indicate average chloride more than 3 times background, leachate scores of 2 indicate average chloride more than twice background. Leachate score of 0.5 indicate possible effects from landfill leachate. Concentrations of combined dissolved or total radium or Ra228/226 ratio not used in determining effects of landfill leachate. Wells PZ-212-SD and PZ-212-SS are considered background and not included. Selected columns shadded for clarity.

MCL, maximum contaminant level; Ra, radium; pCi/L, picocuries per liter; Cl, chloride; mg/L, milligrams per liter; Ra228/226, ratio of radium-228 to radium-226; ug/L, micrograms per liter; Br, bromide; I, iodide; Na, sodium; ALL, alluvium; BED, bedrock; SS, St. Louis Limestone; SD, Salem Formation; KS, Keokuk Limestone; AS, shallow alluvium; AI, intermediate alluvium; AD, deep alluvium; LR, leachate riser; US, unsaturated zone; --, no data]

Well Number and geologic unit			Average of all samples from monitoring well						Number of samples that exceeded background (shaded column indicates constituent used to determine leachate effects)						Number of samples combined Ra exceeded MCL (5 pCi/L)		
Well Number	Media	Unit	Cl (mg/L)	Total combined Ra (pCi/L)	Dissolved combined Ra (pCi/L)	Dissolved Ra228/226	Fuel-VOC (sum in ug/L)	Cl-VOC (sum in ug/L)	Cl	Br	I	Na	Combined Ra	Total combined Ra	Dissolved	Total	Leachate score (L_score)
PZ-302-AI	ALL	AI	66.5	2.11	1.71	3.00	0.0	0.4	0	4	3	4	1	1	0	0	0.5
PZ-302-AS	ALL	AS	51.0	3.23	4.37	25.81	141.9	0.0	0	2	2	2	1	1	1	0	0.5
PZ-303-AS	ALL	AS	89	3.29	2.30	5.06	3,417	16.2	3	4	4	4	3	3	0	0	1
PZ-304-AI	ALL	AI	290	4.27	3.64	2.04	78.0	8.4	4	4	4	4	3	4	0	1	2
PZ-304-AS	ALL	AS	380	4.05	3.13	1.50	302	1.0	4	4	4	4	3	4	0	1	3
PZ-305-AI	ALL	AI	59.3	3.53	2.44	2.92	27.0	0.0	0	4	4	0	2	3	0	0	0.5
S-10	ALL	AS	190	1.71	0.96	--	43.6	14.0	4	4	4	4	0	2	0	0	2
S-5	ALL	AS	310	4.06	2.26	4.83	219	0.0	4	4	3	4	2	3	0	2	2
S-53	ALL	AS	31.0	2.54	2.15	5.91	0.7	0.0	0	3	2	3	2	1	0	1	0
S-61	ALL	AS	8.6	2.35	1.65	4.28	0.4	6.8	0	0	0	0	0	1	0	0	0
S-8	ALL	AS	37.0	2.23	1.77	3.60	0.0	0.0	0	0	0	0	1	1	0	0	0
S-82	ALL	AS	270	5.28	3.84	2.40	3.4	5.5	3	4	4	4	3	3	1	1	2
S-84	ALL	AS	61.3	3.09	2.29	5.19	55.4	2.2	0	4	4	4	3	2	0	0	0.5

[a] Although average Cl and Br are above background, well is one of four wells installed into the Keokuk Limestone beneath the Warsaw Formation. The increased Cl and Br but low I concentrations and upward hydraulic head between the KS series wells and shallower SD and SS series wells indicates that the increased Cl and Br probably are reflective of natural background salinity that can exist within the Mississippian bedrock in the region.

[b] Well installed summer 2013 and only two samples available.

Table 4. Ranked correlation coefficients between degree of landfill leachate effect (L_score), and selected constituents in all 2012–14 groundwater samples from the West Lake Landfill site.

[all constituents in dissolved phase unless noted otherwise; U-234, uranium-234; U-238, uranium-238; VOCs, volatile organic compounds; gray shade indicates values greater than 0.40; red shade indicates values less than -0.40]

Constituent	L_score	Calcium	Magnesium	Sodium	Potassium	Barium	Iron	Manganese	Total alkalinity	Sulfate	U-234	U-238
L_score	1.00	--	--	--	--	--	--	--	--	--	--	--
Calcium	0.60	1.00	--	--	--	--	--	--	--	--	--	--
Magnesium	0.67	0.64	1.00	--	--	--	--	--	--	--	--	--
Sodium	0.73	0.28	0.37	1.00	--	--	--	--	--	--	--	--
Potassium	0.65	0.25	0.41	0.61	1.00	--	--	--	--	--	--	--
Barium	0.76	0.63	0.59	0.50	0.53	1.00	--	--	--	--	--	--
Iron	0.72	0.63	0.59	0.49	0.50	0.80	1.00	--	--	--	--	--
Manganese	0.48	0.76	0.38	0.22	0.22	0.59	0.66	1.00	--	--	--	--
Total alkalinity	0.72	0.53	0.73	0.58	0.59	0.69	0.70	0.44	1.00	--	--	--
Sulfate	-0.41	-0.03	-0.27	-0.27	-0.29	-0.55	-0.49	-0.09	-0.45	1.00	--	--
U-234	-0.44	-0.21	-0.29	-0.27	-0.35	-0.45	-0.49	-0.21	-0.40	0.43	1.00	--
U-238	-0.42	-0.12	-0.26	-0.27	-0.36	-0.42	-0.47	-0.11	-0.37	0.47	0.89	1.00
Combined radium	0.30	0.17	0.34	0.13	0.21	0.40	0.17	-0.04	0.21	-0.28	-0.10	-0.13
Fuel VOCs	0.58	0.28	0.48	0.41	0.47	0.56	0.61	0.31	0.55	-0.43	-0.39	-0.42

Table 5. Results of radium isotope mass balance simulations for monitoring well having average dissolved combined radium above 5.0 picocuries per liter.

[pCi/L, picocuries per liter; Ra228/226; ratio of radium-228 to radium-226; RIM, radiologically impacted material; ALL, alluvium; BED, bedrock; AD, deep alluvial well; SS, St. Louis Limestone; SD, Salem Limestone]

Monitoring well	Media	Geologic unit	Average dissolved combined radium in samples from well (pCi/L)	Average dissolved Ra228/226 ratio in samples from well (pCi/L)	Excess combined radium above background (average in well minus background concentration) (pCi/L)	Fraction of average combined radium in well above background and assumed to be from RIM source	Model predicted average Ra228/226 ratio in samples from well assuming lowest measured background Ra228/226 ratio	Model predicted average Ra228/226 ratio in samples from well assuming highest measured background Ra228/226 ratio	Does measured average Ra228/226 ratio lie within predicted range indicating leaching from RIM is a possible origin of combined radium above background in well?	Predicted lower Cl (mg/L)	Predicted upper Cl (mg/L)	Average measured Cl (mg/L)	Well possibly down-gradient from RIM area?	Ra228/226 ratios consistent with possible RIM origin?	Measured Cl consistent with RIM mixing model?	Possible runoff from Area 1?	Model group	Measured Cl (low)	Measured Cl (high)
D-93	ALL	AD	5.63	1.62	3.65	0.65	0.36	1.76	Yes	93	1,480	343	A2	Y	Y	N	A	330	350
D-6	ALL	AD	5.98	1.39	4.00	0.67	0.34	1.65	Yes	93	1,525	183	A2	Y	Y	N	A	150	200
PZ-110-SS	BED	SS	6.33	0.80	2.77	0.44	0.05	1.19	Yes	95	1,031	190	A1	Y	Y	N	A	180	200
PZ-115-SS	BED	SS	7.42	0.18	3.86	0.52	0.05	1.02	Yes	96	1,209	250	A1	Y	Y	N	A	150	250
PZ-101-SS	BED	SS	26.70	0.10	23.14	0.87	0.02	0.29	Yes	99	1,954	170	A1	Y	Y	Y	A	150	190
MW-1204	BED	SS	5.10	0.60	1.54	0.30	0.06	1.47	Yes	94	740	426	N	Y	Y	Y	B	9.1	1,400
PZ-104-SD	BED	SD	11.09	0.65	7.53	0.68	0.03	0.68	Yes	97	1,550	190	N	Y	Y	Y	B	160	250
D-3	ALL	AD	7.40	1.63	5.42	0.73	0.27	1.34	No	95	1,662	420	A1	N	Y	N	C	240	490
D-83	ALL	AD	6.62	1.40	4.64	0.70	0.30	1.49	Yes	94	1,594	62	A2	Y	N	N	C	54	75
PZ-113-AD	ALL	AD	8.44	2.14	6.46	0.77	0.24	1.17	No	95	1,733	482	A1	N	Y	N	C	450	480
PZ-100-SS	BED	SS	5.12	0.91	1.56	0.30	0.06	1.47	Yes	94	745	4	N	Y	N	N	D	3.4	4.9
PZ-102-SS	BED	SS	5.41	0.57	1.85	0.34	0.06	1.39	Yes	94	825	5	N	Y	N	N	D	4.3	6.5
PZ-107-SS	BED	SS	8.75	0.36	5.19	0.59	0.04	0.86	Yes	96	1,366	300	N	Y	Y	N	D	260	340

1- Upper limit of dissolved combined radium in background groundwater samples from the Missouri River alluvium is 1.98 pCi/L and 3.56 pCi/L for the Mississippian-age bedrock.

2- Range of Ra228/226 (dissolved or total) in background groundwater samples from the Missouri River alluvium is 1.0 to 4.98.

2- Range of Ra228/226 (dissolved or total) in background groundwater samples from the Mississippian-age bedrock is 0.09 to 2.11.

APPENDIX A

TABLE A-1. List of fuel-related volatile organic compounds (VOCs) and chlorinated VOCs.

[Fuel-VOCs, fuel related volatile organic compound; VOCs, volatile organic compounds]

Fuel-VOCs	**Chlorinated VOCs**
Benzene	Tetrachloroethene (PCE).
Chlorobenzene	Trichloroethene (TCE).
Methyl-*tert*-butyl ether (MTBE)	*cis*-1,2-Dichlorobenzene.
Toluene	Vinyl chloride.
1,4-Dichlorobenzene	--
Ethylbenzene	--
Isopropyl benzene	--
1,2-dimethyl benzene	--
Xylene	--
1,2-Dichlorobenzene	--
1,2,4-Trichlorobenzene	--
1,2,4-Trimethylbenzene	--
1,3,5-Trimethylbenzene	--
1,3-Dichlorobenzene	--

Table A2. Pairwise ranked correlation coefficients for selected constituents in alluvial monitoring well samples from the West Lake Landfill site

[values for dissolved phase unless noted otherwise; gray shade indicates values between 0.40 and 1.0; red shade indicates values between -0.40 and -1.0; Ba; barium; Ca, calcium; Fe, iron; Mn, manganese; Na, sodium; _t, indicated total or unfiltered sample; Br, bromide; Cl, chloride; I, iodide; SO_4, sulfate; Ra-226; radium-226; Ra228, radium-228; Ra228/226, ratio of radium-228 to radium-226; VOCs, volatile organic compounds]

Parameter or constituent	Ba	Ca	Fe	Mn	Na	Fe_t	Mn_t	Total alkalinity	Br	Cl	I	SO_4	Ra-226	Ra-228	Ra combined	Ra226_t	Ra228_t	Ra_t combined	Ra228/226	Ra228/226_t	Fuel-VOCs
Ba	1.00																				
Ca	0.22	1.00																			
Fe	0.61	0.41	1.00																		
Mn	-0.16	0.49	0.25	1.00																	
Na	0.53	0.01	0.32	-0.26	1.00																
Fe_t	0.48	0.42	0.83	0.36	0.24	1.00															
Mn_t	-0.18	0.47	0.22	0.96	-0.29	0.39	1.00														
Total alkalinity	0.49	0.12	0.50	-0.18	0.66	0.41	-0.24	1.00													
Br	0.61	0.25	0.44	-0.17	0.76	0.35	-0.25	0.74	1.00												
Cl	0.61	0.18	0.37	-0.19	0.89	0.26	-0.22	0.56	0.75	1.00											
I	0.59	0.39	0.50	0.11	0.51	0.39	0.05	0.55	0.73	0.59	1.00										
SO_4	-0.65	0.15	-0.48	0.29	-0.42	-0.35	0.33	-0.55	-0.51	-0.43	-0.44	1.00									
Ra-226	0.79	0.16	0.29	-0.25	0.46	0.18	-0.27	0.36	0.50	0.54	0.41	-0.45	1.00								
Ra-228	0.52	0.15	0.19	-0.08	0.27	0.18	-0.09	0.17	0.31	0.23	0.19	-0.31	0.63	1.00							
Ra combined	0.67	0.18	0.26	-0.13	0.35	0.22	-0.14	0.25	0.40	0.36	0.28	-0.38	0.81	0.95	1.00						
Ra226_t	0.66	0.16	0.23	-0.17	0.32	0.34	-0.11	0.28	0.32	0.33	0.22	-0.30	0.65	0.52	0.62	1.00					
Ra228_t	0.50	0.11	0.24	-0.10	0.27	0.24	-0.08	0.22	0.28	0.22	0.12	-0.30	0.49	0.60	0.61	0.59	1.00				
Ra_t combined	0.60	0.14	0.26	-0.13	0.32	0.33	-0.09	0.27	0.33	0.29	0.17	-0.32	0.59	0.62	0.67	0.80	0.94	1.00			
Ra228/226	-0.58	0.03	-0.20	0.36	-0.34	-0.11	0.36	-0.27	-0.34	-0.46	-0.28	0.21	-0.70	0.16	-0.14	-0.45	-0.12	-0.26	1.00		
Ra228/226_t	-0.26	-0.04	-0.06	0.07	-0.15	-0.27	-0.01	-0.14	-0.06	-0.11	-0.03	0.00	-0.28	-0.04	-0.14	-0.71	0.13	-0.23	0.43	1.00	
Fuel-VOCs	0.42	-0.11	0.54	-0.13	0.44	0.45	-0.13	0.52	0.40	0.42	0.38	-0.44	0.20	0.00	0.10	0.11	0.05	0.10	-0.29	-0.13	1.00

Table A3. Pairwise ranked correlation coefficients for selected constituents in bedrock monitoring well samples from the West Lake Landfill site.

[values for dissolved phase unless noted otherwise; gray shade indicates values between 0.40 and 1.0; red shade indicates values between -0.40 and -1.0; Ba; barium; Ca, calcium; Fe, iron; Mn, manganese; Na, sodium; _t, indicated unfiltered sample; Br, bromide; Cl, chloride; I, iodide; SO_4, sulfate; Ra-226; radium-226; Ra228, radium-228; Ra228/226, ratio of radium-228 to radium-226; VOCs, volatile organic compounds]

Parameter or constituent	Ba	Ca	Fe	Mn	Na	Fe_t	Mn_t	Total alkalinity	Br	Cl	I	SO_4	Ra-226	Ra-228	Ra combined	Ra226_t	Ra228_t	Ra_t combined	Ra228/226	Ra228/226_t
Ba	1.00																			
Ca	0.70	1.00																		
Fe	0.68	0.51	1.00																	
Mn	0.77	0.58	0.80	1.00																
Na	0.09	0.07	0.20	0.13	1.00															
Fe_t	0.69	0.52	0.81	0.81	0.21	1.00														
Mn_t	0.74	0.55	0.75	0.95	0.13	0.88	1.00													
Total alkalinity	0.47	0.56	0.50	0.48	0.32	0.46	0.45	1.00												
Br	0.25	0.45	0.29	0.30	0.55	0.28	0.26	0.55	1.00											
Cl	0.33	0.50	0.35	0.31	0.62	0.39	0.30	0.43	0.81	1.00										
I	0.53	0.59	0.45	0.44	0.46	0.40	0.41	0.57	0.67	0.62	1.00									
SO_4	-0.21	0.14	-0.24	-0.14	0.10	-0.13	-0.09	-0.10	0.21	0.25	-0.02	1.00								
Ra-226	0.60	0.56	0.38	0.38	-0.02	0.38	0.33	0.44	0.21	0.14	0.45	-0.24	1.00							
Ra-228	0.40	0.33	0.24	0.24	0.13	0.35	0.26	0.20	0.12	0.18	0.28	-0.04	0.45	1.00						
Ra combined	0.57	0.51	0.36	0.33	0.06	0.38	0.31	0.38	0.23	0.19	0.45	-0.20	0.89	0.75	1.00					
Ra226_t	0.65	0.59	0.45	0.45	0.07	0.51	0.44	0.44	0.23	0.21	0.45	-0.17	0.84	0.41	0.79	1.00				
Ra228_t	0.35	0.38	0.22	0.26	-0.05	0.29	0.27	0.29	0.05	0.06	0.15	-0.02	0.37	0.46	0.44	0.47	1.00			
Ra_t combined	0.60	0.59	0.42	0.43	0.08	0.46	0.41	0.44	0.23	0.22	0.43	-0.13	0.76	0.48	0.77	0.89	0.74	1.00		
Ra228/226	-0.52	-0.46	-0.29	-0.27	-0.25	-0.31	-0.23	-0.56	-0.31	-0.24	-0.50	0.23	-0.87	0.00	-0.60	-0.71	-0.25	-0.60	1.00	
Ra228/226_t	-0.29	-0.25	-0.16	-0.19	-0.17	-0.24	-0.22	-0.39	-0.13	-0.04	-0.36	0.21	-0.58	0.04	-0.37	-0.78	0.04	-0.53	0.81	1.00
Fuel-VOCs	0.45	0.39	0.46	0.42	0.06	0.35	0.39	0.42	0.23	0.19	0.48	-0.21	0.34	0.16	0.28	0.30	0.18	0.31	-0.41	-0.16

Appendix B. Limitations of data used in this report

Limitations of radionuclide data from the 2012–14 West Lake Landfill site comprehensive groundwater sampling

Data from 32 replicate samples from the 2012–14 comprehensive sampling effort at the West Lake Landfill (WLL) site were examined for comparability by calculating the relative percent difference (RPD) between initial and replicate sample pairs. The RPD was calculated as the absolute value of the difference between the initial and replicate value divided by the average of the two values times 100. The median RPD values of dissolved and total radium-226 (Ra-226) and radium-228 (Ra-228) between the initial and replicate samples (19.2 to 37.9 percent) were smaller than the median values of the analytical combined standard uncertainty (CSU) that ranged from 42 to 47 percent indicating differences in the field replicate samples generally were within the analytical precision. Examination of the 32 replicate sample pairs indicates that at the maximum contaminant level (MCL) of 5.0 pCi/L, the analytical uncertainty expressed as the combined standard CSU, generally represents about 38 percent of the measured value and that the data are adequate for the purposes of comparison to the MCL (fig. B-1); however, as the measured values decrease the CSU becomes increasingly larger in relation to the measured values with the CSU approaching 100 percent of the measured value at combined radium concentrations of about 1.0 picocuries per liter (pCi/L). While useful for comparison to the MCL, the dataset probably is not adequate in discriminating combined radium concentrations at about 1 to 2 pCi/L or smaller.

Precision of the analytical results for dissolved and total Ra-226 and Ra-228 was examined by comparing differences between the reported values (irrespective of "J" or "U" flags) in the regular and replicate samples. Although the narratives in the quarterly monitoring documents (Engineering Management Support, Inc., 2012, 2013a, 2013b; Paul Rosasco, Engineering Management Support, Inc., written commun., 2014) discuss these as duplicate samples, the sample bottles were filled sequentially and are more consistent with a replicate sample. Of the 32 sample pairs, 7 dissolved and 7 total combined radium pairs had differences greater than 50 percent and; with the exception of the August 2012 sample from well PZ-113-AD, differences generally were less than 50 percent when the average measured concentrations were larger than about 6 pCi/L (fig. B-2). Only 6 of the 32 pairs of initial and replicate samples had dissolved or total Ra-226 differences exceeding 50 percent; however, analytical precision for dissolved and total Ra-228 were lower with 10 dissolved and 11 total Ra-228 sample pairs having differences exceeding 50 percent (not shown). The lower precision in the Ra-228 results is apparent using a simple pairwise correlation between initial and replicate values where coefficients for dissolved and total Ra-226 (r of 0.85 and 0.92, respectively) were larger than those for dissolved and total Ra-228 (r of 0.63 and 0.55, respectively). Reporting levels for Ra-228 (median left censored value of 0.85 pCi/L) were nearly four times larger than the median left censored value for Ra-226 of 0.24 pCi/L indicating greater difficulty in determining low concentrations of Ra-228 compared to Ra-226. The smaller precision in the Ra-228 values translates to greater error in Ra228/226 ratios with correlations for dissolved and total Ra228/226 ratios of 0.43 and 0.61 between initial and replicate samples. Of the 33 initial/replicate sample pairs that had detectable Ra-228 and Ra-226, 13 had Ra228/226 differences between the initial and replicate samples exceeding 50 percent (fig. B–3). Unlike precision in concentrations of Ra-228 or Ra-226, which generally increased with increasing concentration, differences in Ra228/226 ratios increased with the value of the ratio. The lower precision in measured Ra-228 concentrations and Ra228/226 ratios is a possible limitation and source of uncertainty in the use of the Ra228/226 ratio in the isotope mass-balance mixing model. The lower precision in measured the Ra-228 concentrations is consistent with that observed from numerous other evaluations of the analytical precision of these radionuclide measurements (Szabo and others, 2008; Szabo and others, 2012).

Perhaps the most important limitation on the usability of the data and findings in this report is the paucity of background radionuclide data in the Mississippian-age bedrock and Missouri River alluvium in the vicinity of the WLL site. While a search for historical radionuclide data was done for the city of St. Louis, and St. Charles and St. Louis Counties, the background dataset of samples within 5 mi of the WLL site is small consisting of only 17 alluvial groundwater and 11 bedrock groundwater samples. Included in this background dataset are samples from nine domestic wells in the Missouri River alluvium and two domestic wells in the Mississippian-age bedrock aquifer. Seven of the alluvial wells were domestic wells that seemed to be "sand point" wells (a 2–5 feet [ft] long metal screen at the bottom of a steel or iron casing driven into the alluvium) that usually are less than about 40 ft deep. Two of the alluvial wells were irrigation wells that were drilled to 80 ft or more and open to the 20 or 30 ft of coarse sands and gravels near the base of the alluvium. These alluvial wells probably are a reasonable comparison to the alluvial monitoring wells at the WLL site that generally have a 10-ft long screen set in a 12–15 ft thick filter sand pack; however, radionuclide data from domestic or public-supply wells could be biased low because domestic wells often have long open-intervals (50 to hundreds of feet), and water samples from these wells is a mixture of water often from many geologic units compared to bedrock monitoring wells at the WLL site that extract water from a limited 10- to 20-ft thick section of the bedrock strata. In domestic or public-supply wells, a small amount of water produced from isolated zones or fractures within the bedrock aquifer that might have larger radionuclide concentrations can be diluted by water produced from other zones. None of the background wells with radionuclide data contain elevated concentrations of major ions such as those seen in background wells at the Champ Landfill or noted on historical well logs from the region, and the absence of radionuclide data from wells with water of higher salinity is a data gap. In addition, isolated radionuclide-enriched minerals have been noted in eastern Missouri. Bell (1963) noted that the Salem Formation locally can contain small grains of fluorite (a mineral often enriched in uranium compared to the carbonate matrix) and also noted that during wet periods the sporadic appearance of a yellow carnotite-type mineral on rock faces of quarries in the Salem Formation near St. Genevieve, Missouri although two samples from the bulk limestone did not have unusually large amounts of uranium (0.0004 and 0.0003 percent). Even including the 2013 supply-well sample data, the small amount of background data available, especially radionuclides in the bedrock units of concern at the WLL site (St. Louis Limestone and Salem Formation), remains an important limitation to understanding the occurrence of radium above the MCL at the WLL site.

Other limitations with the 2012–14 data include monitoring well locations and details on field measurements made during monitoring well sampling. The focus on the existing monitoring well program for the Bridgeton Landfill was compliance monitoring for release of contaminants from that facility into the adjacent alluvium and bedrock. Monitoring wells for the Bridgeton Landfill are clustered near that facility on property owned by the landfill, especially bedrock monitoring wells. It seems that the focus of groundwater monitoring for the West Lake Landfill and radiologically impacted material (RIM) areas was installation of monitoring wells at various depths within the alluvium around landfill materials and again on landfill property. A result of these two different monitoring programs is an absence of bedrock monitoring wells in the middle and western parts of the WLL site, and absence of monitoring wells offsite and generally downgradient of the facility. Results of the 2012–14 comprehensive groundwater monitoring at the site detected landfill leachate effects in a number of monitoring wells, many of which are along the landfill property boundary, indicating that in some areas landfill leachate has possibly moved beyond the extent of the existing monitoring well network. Many monitoring wells at the site, especially bedrock wells, yield small quantities of water, and samples often can be turbid (Ward Herst, Herst and Associates, oral communication, 2013). Because field measurements commonly made during sample collection such as water temperature, pH, dissolved oxygen content, and turbidity were not available in the database provided, the effect of sample turbidity and possible changing redox conditions during sampling on concentrations of radionuclides such as total combined radium cannot be evaluated.

Limitations in the use of the isotope mass-balance mixing model

The isotope and secondary chloride mass-balance mixing model is subject to a variety of uncertainties and limitations, some of which can be, or were, constrained and others that are more open-ended and cannot be constrained with the available data; and the mass-balance model results by themselves are not conclusive. One assumption stated is that the Ra-228 in the RIM is supported (in radioactive equilibrium) by its parent radionuclide thorium-232 (Th-232) such that the ratio of Ra228/226 in the RIM has been stable (0.006 based on 1990s boring data that penetrated and sampled RIM material) since its placement at the WLL site. The Ra228/226 ratio is based on data from 12 samples containing greater than 30 pCi/g combined radium listed in the OU1 Site Characterization Summary Report (Engineering Management Support, Inc., 1997). This value was obtained by substituting the reporting level for left censored Ra-228 values. A ratio of 0.007 was calculated using samples with greater than 5 pCi/L total radium (n=25). Using only samples where both Ra-226 and Ra-228 results were not censored and the total combined radium was greater than 30 pCi/g resulted in an average calculated Ra228/226 ratio for the RIM of 0.005 (n=2). If the Ra-228 in the RIM is not supported by its parent Th-232 (that is, there is substantially less Th-232 than Ra-228 in the RIM, or if there is no Th-232), then the Ra228/226 ratio in the RIM will have changed over time with the decay of one-half the unsupported Ra-228 every 5.8 years (Ra-228 half-life); and the possible range for the RIM may be from about 0.06 in 1973 to as low as about 0.001 by 2014. Increasing the Ra228/226 ratio in the RIM to as large as 0.2 has minimal effect on the mass-balance calculations shown in table 5, except that the average Ra228/226 of 0.10 measured in samples from well PZ-101-SS would fall slightly outside the mass-balance-model predicted average of 0.11 to 0.45 for samples from well PZ-101-SS. Decreasing the assumed Ra228/226 ratio in the RIM to 0.001 had no effect in the mass-balance model outcome as the average measured Ra228/226 ratios in the same 11 wells fell within the model-predicted range and measured Ra228/226 in wells D-3 and PZ-113-AD continued to fall outside the model predicted range.

The isotope mass balance is more sensitive to the range of Ra228/226 ratio in background groundwater than the Ra228/226 ratio in the RIM; for example, doubling the Ra228/226 ratio for background groundwater in the bedrock to 4.22 does not change the outcome of the calculations in table 5; however, reducing the background ratio to 1.11 for the groundwater in the bedrock and 2.50 for the groundwater in the alluvium results in three bedrock wells (PZ-100-SS, PZ-104-SD, and PZ-110-SS) and all alluvial wells having Ra228/226 ratios outside the range for a possible RIM origin. These somewhat lower Ra228/226 ratios are not outside the range of reasonable values when compared to national or global carbonate or clastic sand aquifers. Vinson and others (2012) reported Ra228/226 ratios in the carbonate-controlled geochemistry of the Cambrian-age Jordan aquifer (a sandstone aquifer) generally less than 1 (median of 0.5). This ratio is consistent with a median Ra228/226 ratio of 0.54 (*n*=30) for the MCOO (Mid-continent Cambro-Ordovician dolostone) aquifers in the United States (Szabo and others, 2012). The Ra228/226 isotope activity ratio data for groundwater of the United States show ratios that typically are enriched in Ra-228 relative to Ra-226 in the clastic sand aquifers (Szabo and others, 2012). In their Online Supplemental Table S–4, Szabo and others (2012) report the Ra228/226 isotope activity ratio medians of 3.8 and 4.9 for the High Plains Tertiary aquifer and Mountain-West alluvial fill aquifers, respectively, and 3.0 for Mountain-West Tertiary sand and shale bedrock aquifers; though, it is noted that these median values are not well constrained because of the large number of samples where the quantification of one or both of the isotope concentrations had a high degree of uncertainty. The ratios in the United States sand aquifers were comparable to the activity ratio data for these radium isotopes compiled worldwide from sand and sandstone aquifers by Vengosh and others (2009); they obtained a median Ra228/226 isotope activity ratio of 1.6, with ratios from individual samples exceeding 5, and median ratios from a major sandstone aquifer they studied of about 2.9. The relatively large range of Ra228/226 isotope activity ratios from relatively similar aquifers indicates that constraining this value to a range to which the mass-balance model calculation is sensitive is difficult or unlikely.

In addition to the Ra228/226 ratio used for the background groundwater term, the mass-balance model is sensitive to the upper limit of background dissolved combined radium concentration because this value is used to calculate the fraction of excess (fraction above the MCL) dissolved combined radium in the well; for example, if the maximum background dissolved combined radium for the alluvium and bedrock is set to 12.6 pCi/L (the maximum total combined radium reported in public-supply wells in the region by the MDNR), then only four wells, three of which are alluvial wells (D-93, D-6, PZ-101-SS, and D-83), have average dissolved combined radium above the background and only these could be considered as having a possible RIM source in the model. In that case, chloride (Cl) concentrations in well D-83 would remain outside the predicted range in the secondary Cl mass-balance model. As was discussed in the text, if the upper limit of background dissolved combined radium for the bedrock is more similar to that measured in offsite wells PZ-212-SS and PZ-212-SD and about 1 pCi/L or less, none of the bedrock wells and only four alluvial wells (D-93, D-6, PZ-115-SS, and D-83) would have average dissolved combined radium values within the predicted interval for a possible RIM source. Szabo and others (2012) report combined radium concentrations range between 2 and 5 pCi/L for the High Plains Tertiary aquifer, Mountain-West alluvial-fill aquifers, and the Mountain-West Tertiary sand and shale bedrock aquifers. The uncertainty in the upper limit of background dissolved combined radium concentrations in the bedrock is an important consideration in evaluating the isotope mass-balance model results presented in table 5 because of the sensitivity of the results to the background value used. The model calculation is sensitive to variation within the possible reasonable range for this value.

The secondary Cl mass balance was used to provide additional constraint on the radium isotope mass-balance model; however, the chloride mass-balance model adds little additional constraint to the radium isotope mass balance because of the wide possible range of Cl in landfill leachate. The range of Cl concentrations in landfill leachate of 100 to 2,240 mg/L reported by Tchobanoglous and others (1977) is large but only slightly larger than the upper limit of Cl in background groundwater in the alluvium (79.5 mg/L) or bedrock (91 mg/L). Because of varied age and composition of landfilled materials at the WLL site, it is likely that Cl concentrations vary greatly in leachate generated at the site and probably span the range reported by Tchobanoglous and others (1977). In addition, coupling secondary Cl mass balance to the radium isotope mass balance by using the excess dissolved combined radium to estimate the fraction of RIM and landfill leachate assumes that the proportion of assumed radium from RIM and landfill leachate are constant, which they probably are not. Leachate generated from demolition fill may have smaller chloride concentrations compared to leachate from areas containing sanitary wastes, and RIM spread over both areas could result in different fractions of radium to chloride in leachate assuming the radium is in fact mobilized from these various areas. The different composition of leachate generated from demolition wastes compared to sanitary wastes also may result in different mobilization rates from radium from RIM mixed with the wastes. The range of Cl concentrations reported in the literature was used to account for the inherent uncertainty in chloride concentrations in leachate at the WLL site, but even that large range may underestimate the actual variability. The Cl is also advancing conservatively with leachate plumes, whereas the radium likely is undergoing some degree of attenuation.

Data constraints are broad given the uncertainty in background radium concentrations and Ra228/226 ratios, possible uncertainty in Ra228/226 ratios of the RIM, potential for some non-RIM wastes to contain radium, and wide range of possible Cl concentrations in landfill leachate; and there is potential for a possible bias toward "false positives" (measured Ra228/226 ratio in a well is consistent with a possible RIM origin when there is no RIM origin) in the isotope mass-balance calculations. Because of the large uncertainty, a positive result in the isotope mass-balance calculations does not "prove a RIM origin" and also does not rule out the other hypotheses for the origin of above-MCL combined radium in a well. Negative results (well Ra228/226 ratios are not consistent with a model calculated RIM origin) are probably less affected by the uncertainty of the constraints than are the positive results and, therefore, there is more confidence in them. As an example, the small number of background samples for combined radium in groundwater from the Mississippian-age bedrock (*n*=11, table 2) indicates an upper limit of background dissolved combined radium of 3.56 pCi/L; however, concentrations of dissolved combined radium in offsite monitoring wells near the site (PZ-212-SS and PZ-212-SD) are less than 1.0 pCi/L. Changing the background dissolved combined radium in the isotope mass balance model from 3.56 pCi/L to less than 1.0 pCi/L has a substantial effect on the model output. Using background dissolved combined radium of 3.56 pCi/L as was done in table 5 for all of the eight bedrock wells with average dissolved combined radium being possibly consistent with a RIM source, whereas using less than 1.0 pCi/L for background dissolved combined radium results in all eight of the bedrock wells having Ra228/226 ratios inconsistent with a possible RIM source.

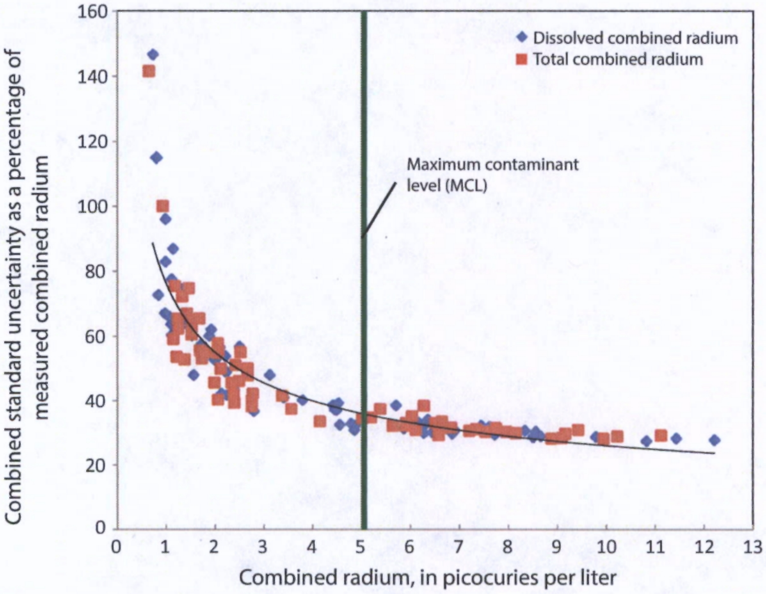

Figure B–1. Plot of combined standard uncertainty (CSU) of combined radium concentration as a percentage of reported combined radium concentration in initial and replicate sample pairs collected during the 2012–14 comprehensive groundwater sampling.

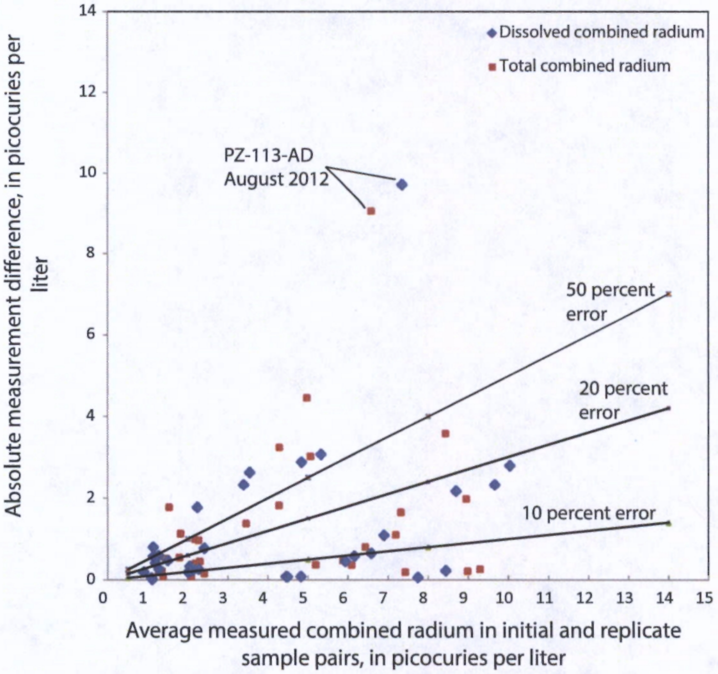

Figure B–2. Differences between measurements of dissolved and total combined radium in initial and replicate groundwater sample pairs collected during the 2012–14 comprehensive groundwater sampling in relation to the mean combined radium activity. -Note, solid lines are the 10, 20, and 50 percent relative percent difference (RPD) between initial and replicate results.

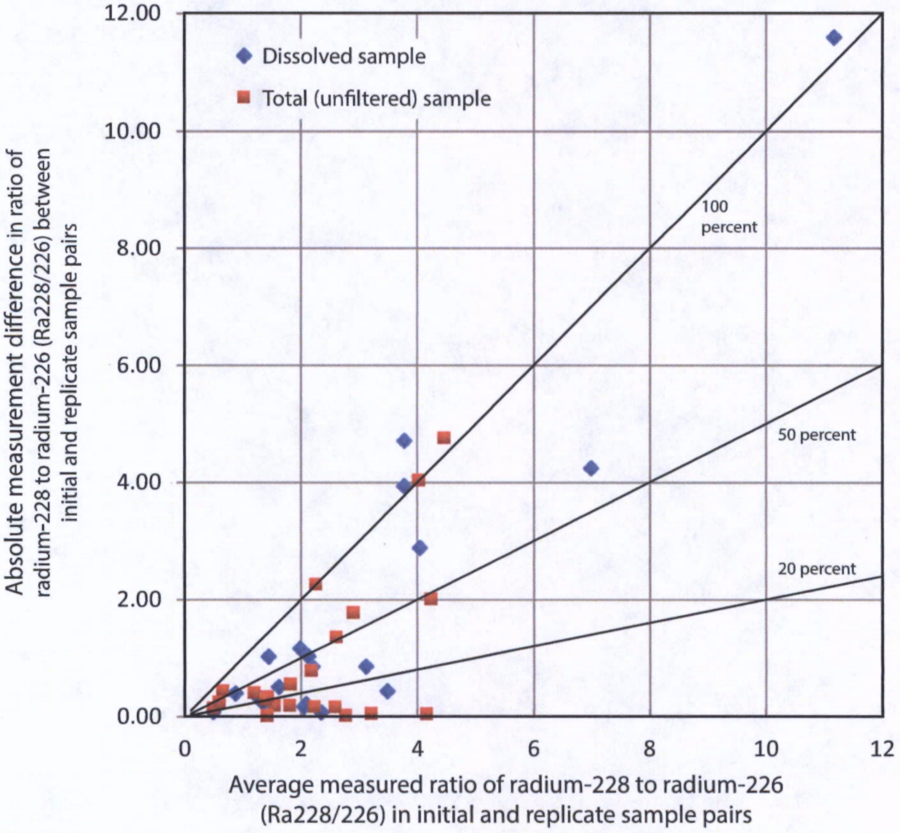

Figure B-3. Differences between ratios of radium-228 to radium-226 (Ra228/226) in initial and replicate groundwater sample pairs collected during the 2012–14 comprehensive groundwater sampling in relation to the average Ra228/226 ratio in the initial and replicate sample. Note-- solid lines are the 20, 50, and 100 percent relative percent difference (RPD) between initial and replicate sample result.

Appendix C. Potentiometric maps prepared by the West Lake Landfill Responsible Parties (RP) during the 2012–13 comprehensive sampling.

Appendix C
Figure C-1
July 30, 2012

LEGEND

●	Alluvium Groundwater Well
●	Bedrock Groundwater Well
━	Operable Unit-1 Area as Defined By ROD
═	Paved Road
╌	Unpaved Road
— 440 —	Groundwater Elevation Contour (FAMSL) - Alluvium
— 440 —	Groundwater Elevation Contour (FAMSL) - St. Louis Fm
(458.64)	Alluvium Groundwater Elevation (FAMSL)
(458.64)	Bedrock Groundwater Elevation (FAMSL)
*	Groundwater Elevation not used for contouring
→	Groundwater Flow Direction - Alluvium
→	Groundwater Flow Direction - St. Louis
(FAMSL)	Feet above mean sea level

WELL FORMATION DESIGNATIONS

LR or MW:	Undifferentiated
S or AS:	Alluvial Shallow Well
I or AI:	Alluvial Intermediate Well
D or AD:	Alluvial Deep Well
SS:	St. Louis Formation Well
SD:	Salem Formation Well
KS:	Keokuk Formation Well

NOTES:

1. Horizontal Coordinates Based on State Plane Missouri East Zone NAD 27
2. Elevations Based on U.S.G.S. Datum.
3. Existing Grade Contours are from the Aerial Survey Completed by the Sanborn Mapping Company on July 20, 2011.
4. Base Map Prepared by Aquaterra Environmental Solutions, Inc.

Figure 2
Alluvial Groundwater Table and
St. Louis FM. Potentiometric Elevation Map
July 30, 2012
West Lake Landfill Operable Unit-1

EMSI Engineering Management Support, Inc.

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Appendix C
Figure C-2
April 2, 2013

LEGEND

- 🔵 Leachate Collection Sump
- 🔵 Alluvium Groundwater Well
- 🟢 Bedrock Groundwater Well
- ━ Operable Unit-1 Area as Defined By ROD
- ━ Estimated Extent of Radiologically Impacted Material
- ═ Paved Road
- ＝ ＝ ＝ Unpaved Road

— 440 —	Groundwater Elevation Contour (FAMSL) - Leachate
— 440 —	Groundwater Elevation Contour (FAMSL) - Alluvium
— 440 —	Groundwater Elevation Contour (FAMSL) - St. Louis Fm
(279.02)	Leachate Groundwater Elevation (FAMSL)
(429.82)	Alluvium Groundwater Elevation (FAMSL)
(458.64)	St. Louis Fm Groundwater Elevation (FAMSL)
*	Groundwater Elevation not used for contouring
→	Groundwater Flow Direction - Leachate
→	Groundwater Flow Direction - Alluvium
→	Groundwater Flow Direction - St. Louis Fm
(FAMSL)	Feet above mean sea level

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Figure 2
Alluvial Groundwater Table and
St. Louis FM. Potentiometric Elevation Map
April 2, 2013
West Lake Landfill Operable Unit-1

EMSI Engineering Management Support, Inc.

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Appendix C
Figure C-3
July 8, 2013

LEGEND

●	Leachate Collection Sump
●	Alluvium Groundwater Well
●	Bedrock Groundwater Well
▬	Operable Unit-1 Area as Defined By ROD
▬	Estimated Extent of Radiologically Impacted Material
═	Paved Road
= = = =	Unpaved Road
— 440 —	Groundwater Elevation Contour (FAMSL) - Alluvium
— 440 —	Groundwater Elevation Contour (FAMSL) - St. Louis Fm
(279.02)	Leachate Groundwater Elevation (FAMSL)
(429.82)	Alluvium Groundwater Elevation (FAMSL)
(458.64)	St. Louis Fm Groundwater Elevation (FAMSL)
*	Groundwater Elevation not used for contouring
	Groundwater Flow Direction - Leachate
	Groundwater Flow Direction - Alluvium
	Groundwater Flow Direction - St. Louis Fm
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Figure 2
Alluvial Groundwater Table and
St. Louis FM. Potentiometric Elevation Map
July 8, 2013
West Lake Landfill Operable Unit-1

EMSI Engineering Management Support, Inc.

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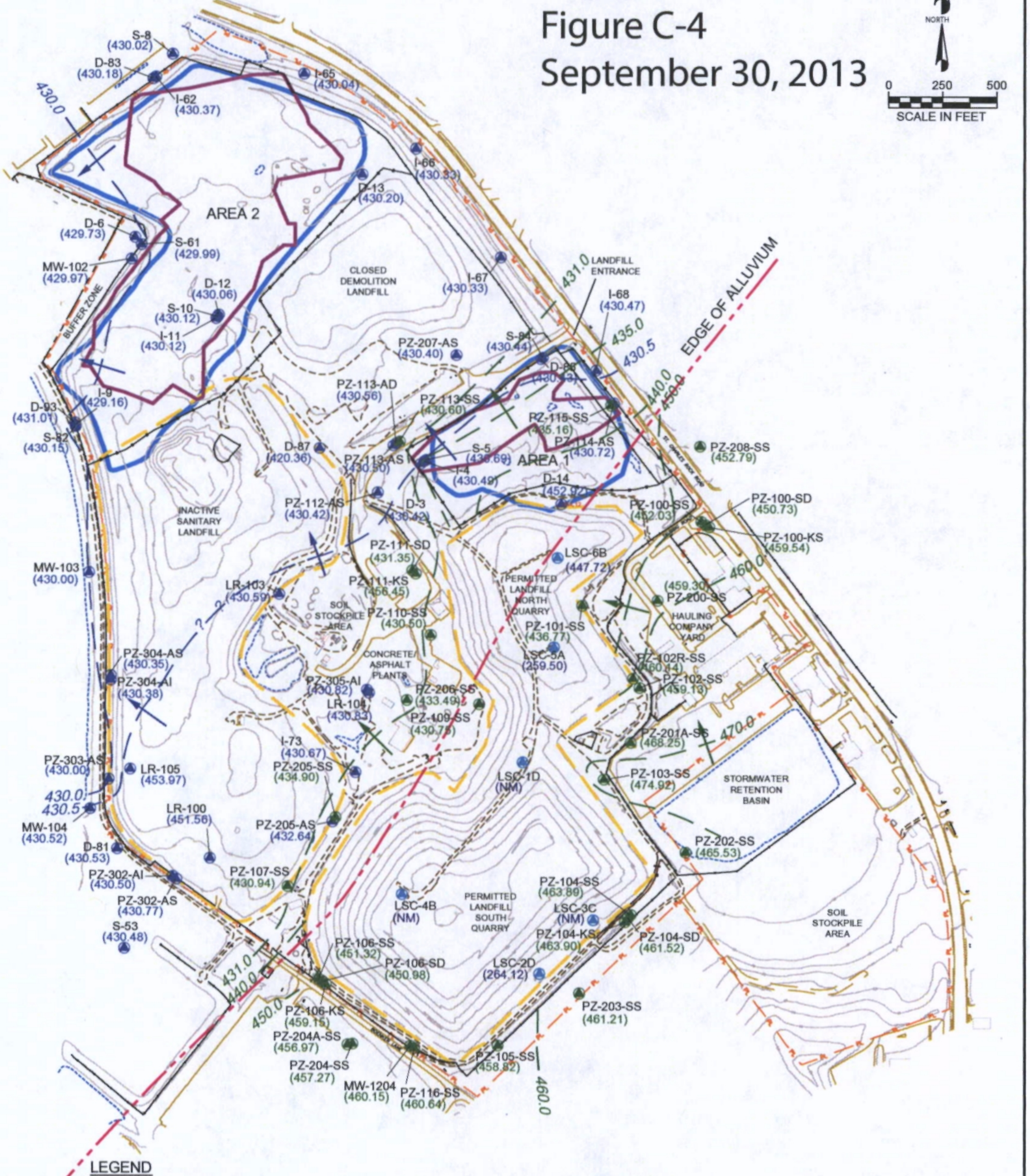
Appendix C
Figure C-4
September 30, 2013



LEGEND

●	Leachate Collection Sump
●	Alluvium Groundwater Well
●	Bedrock Groundwater Well
▬	Operable Unit-1 Area as Defined By ROD
▬	Estimated Extent of Radiologically Impacted Material
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→	Groundwater Flow Direction - Alluvium
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(FAMSL)	Feet above mean sea level

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Figure 2
Alluvial Groundwater Table and
St. Louis FM. Potentiometric Elevation Map
September 30, 2013
West Lake Landfill Operable Unit-1

EMSI Engineering Management Support, Inc.