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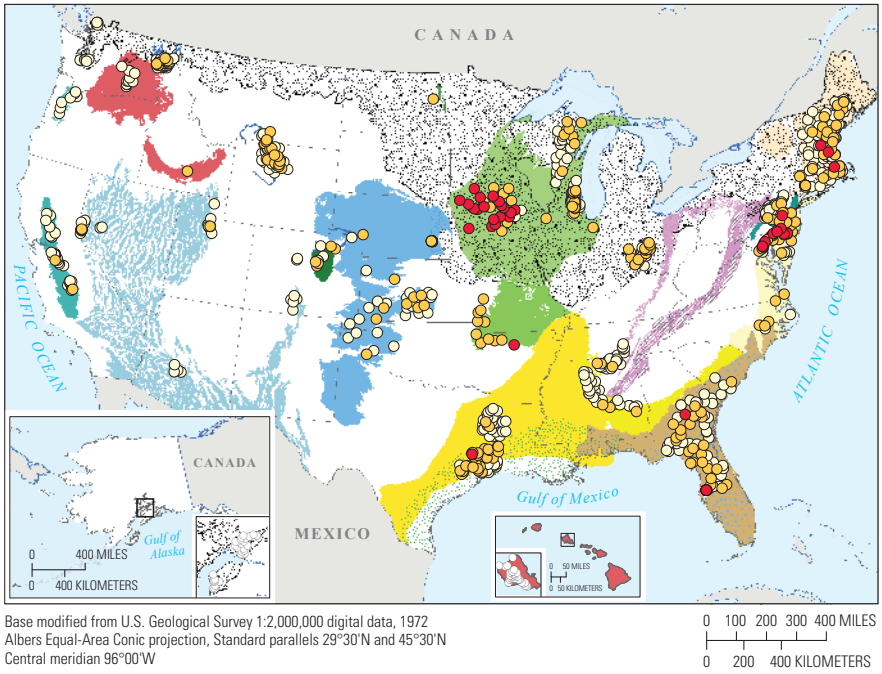
Principal Aquifers Can Contribute Radium to Sources of Drinking Water Under Certain Geochemical Conditions

What are the most important factors affecting dissolved radium concentrations in principal aquifers used for drinking water in the United States? Study results reveal where radium was detected and how rock type and chemical processes control radium occurrence. Knowledge of the geochemical conditions may help water-resource managers anticipate where radium may be elevated in groundwater and minimize exposure to radium, which contributes to cancer risk.

Summary of Major Findings

- Concentrations of radium in principal aquifers used for drinking water throughout the United States generally were below 5 picocuries per liter (pCi/L), the U.S. Environmental Protection Agency (USEPA) maximum contaminant level (MCL) for combined radium—radium-226 (Ra-226) plus radium-228 (Ra-228)—in public water supplies. About 3 percent of sampled wells had combined radium concentrations greater than the MCL.
- Elevated concentrations of combined radium were more common in groundwater in the eastern and central United States than in other regions of the Nation. About 98 percent of the wells that contained combined radium at concentrations greater than the MCL were east of the High Plains.
- The highest concentrations of combined radium were in the Mid-Continent and Ozark Plateau Cambro-Ordovician aquifer system and the Northern Atlantic Coastal Plain aquifer system. More than 20 percent of sampled wells in these aquifers had combined radium concentrations that were greater than or equal to the MCL.
- Concentrations of Ra-226 correlated with those of Ra-228. Radium-226 and Ra-228 occur most frequently together in unconsolidated sand aquifers, and their presence is strongly linked to groundwater chemistry.
- Three common geochemical factors are associated with the highest radium concentrations in groundwater: (1) oxygen-poor water, (2) acidic conditions (low pH), and (3) high concentrations of dissolved solids.



Base modified from U.S. Geological Survey 1:2,000,000 digital data, 1972
Albers Equal-Area Conic projection, Standard parallels 29°30'N and 45°30'N
Central meridian 96°00'W

0 100 200 300 400 MILES
0 200 400 KILOMETERS

EXPLANATION

Principal aquifer system

- Glacial sand and gravel
- Crystalline rock New England/New York
- Northern Atlantic Coastal Plain
- Appalachian Piedmont Mesozoic Basins
- Valley and Ridge and Mississippian limestone
- Coastal Plain—Texas Upland, Gulf lowland, Mississippi Embayment
- Floridan and surficial
- Mid-Continent and Ozark Plateau Cambro-Ordovician
- High Plains Tertiary alluvium and fill
- Coastal Plain, Southeastern
- Western Foreland Tertiary/ Cretaceous sandstone
- Rio Grande and Basin and Range basin fill
- Pacific Coastal (Central Valley and Willamette Basins) alluvium and fill
- Western alluvium and fill
- Basalt

Well and radium concentration—In picocuries per liter

- Less than 1
- 1 to 4.99
- Greater than or equal to 5

Figure 1. Elevated radium concentrations occur most commonly in aquifers in the eastern and central United States for the wells sampled in 15 principal aquifer systems across the United States. Ninety-eight percent of the wells that exceeded the combined radium (radium-226 plus radium-228) drinking-water standard of 5 picocuries per liter established by the U.S. Environmental Protection Agency were in aquifers east of the High Plains (modified from U.S. Geological Survey, 2003).

What is an MCL?

A maximum contaminant level (MCL) is a legally enforceable USEPA drinking-water standard. An MCL sets the maximum permissible level of a contaminant in water that is delivered to users of a public water system that provides water for human consumption through at least 15 service connections or regularly serves at least 25 individuals. The USEPA sets MCLs as close as feasible to the maximum contaminant level goal (MCLG), which is the maximum level of a contaminant in drinking water that has an adequate margin of safety and has not been shown or proved to cause adverse health effects. When establishing MCLs, the USEPA takes into account the best available analytical and treatment technologies and cost considerations. The MCL for combined radium applies to the finished drinking water provided by public water systems (after treatment and before distribution). In this study, radium concentrations in untreated source water for public-supply wells and private (domestic) wells were compared to the MCLs to provide an initial perspective on the potential significance of detected contaminants to human health. Concentrations measured in source water do not necessarily reflect the quality of finished water from public water systems.

Study Design

The U.S. Geological Survey (USGS), with participation from the USEPA, conducted a study characterizing the occurrence of radium in groundwater in major aquifers of the United States as part of the National Water-Quality Assessment (NAWQA) Program (Szabo and others, 2012). As part of this study, 1,270 wells were sampled over 15 years (during 1990–2005) and analyzed for Ra-226, Ra-228 and other water-quality parameters. A subset of 645 water samples were analyzed for the short-lived radium radionuclide, radium-224 (Ra-224). All samples were analyzed for major ions and trace elements, and field parameters, such as dissolved oxygen and pH were measured. Wells generally were sampled once, and raw-water samples were collected and filtered (0.45 micron) as close to the wellhead as possible and before any treatment. Of the sampled wells, 48 percent were used for domestic supply, 14 percent were used for public drinking-water supply, and 34 percent were observation wells for sampling recently recharged water in drinking-water-supply aquifers. Type or use was unknown for 4 percent of the sampled wells. All of the water samples were analyzed for at least one or the other longer-lived radium radionuclides, Ra-226 or Ra-228. A minimum of 30 samples were collected and analyzed for at least Ra-226 or Ra-228 from each principal aquifer. More details on the sampling and analyses can be obtained in Szabo and others (2012).

This assessment is the first to characterize occurrence of the three major radionuclides of radium throughout much of the United States, specifically in 15 principal aquifer systems in 45 States (fig. 1). The principal aquifer systems used in the study by Szabo and others (2012) are grouped slightly different from the classification of Lapham and others (2005) and the classification of DeSimone and others (2009). This study also relates radium occurrence with extensive geologic, geochemical, and hydrologic information and data previously lacking on this scale, particularly in the west-central and western United States. In addition, the study extends work from previous national surveys by Longtin (1988) and Focazio and others (2001).

What is Radium?

Radium is a naturally occurring radioactive element (or radionuclide) and known carcinogen that generally is present at low levels in all soil, water, and rocks. It is derived from the common long-lived radioactive elements, uranium and thorium, which decay slowly to produce other radioactive elements, such as radium. Three commonly occurring radionuclides of radium are Ra-224, Ra-226, and Ra-228, each of which varies in abundance because each decays to a daughter product at a different rate (known as “half-life”). Radium-226 is the most abundant radium isotope in the environment in terms of its mass, primarily because of its long half-life. Ra-226 and Ra-228 have half-lives measurable in terms of years and, therefore, once dissolved can be mobile for considerable periods of time in groundwater. Radium-224 has a shorter half-life, measured in days; however, all radium radionuclides can contribute to the total amount of radioactivity in a water sample. This total level of radioactivity, measured as gross alpha-particle or beta-particle activity, often is used as a cost-effective screening tool to determine the general presence of radionuclides in water samples.

How Does Radium Get From Rocks Into Groundwater?

Radium is derived from the radioactive decay of naturally occurring uranium and thorium in rocks and sediment. Most rocks and sediment contain some uranium and thorium and, thereby, contain radium as well, but usually in small quantities. Uranium and thorium are most common in granitic and metamorphic crystalline rocks and in associated weathered sedimentary deposits in the central United States and mountainous regions of the East and West (Grauch, 1976; Duval and Riggle, 1999; Ayotte and others, 2007). Groundwater flows slowly through pores or cracks in underground rocks and aquifer sediments and dissolves radium-bearing minerals as it moves. Chemical processes in groundwater and on rock surfaces, such as mineral dissolution, desorption, and ion-exchange reactions, control the release of radium from aquifer solids. Radium can also be physically pushed into pore water during isotope decay.

What Human Health Concerns Are Related to Radium?

Exposure to radium over long periods of time can increase the risk of cancer. Radium can enter the body in drinking water, food, or inhaled dust particles that contain radium. Radium in the body behaves similarly to calcium and can replace calcium in tissues, particularly bone. Long-term exposure to radium increases the risk of developing diseases such as bone and sinus cancer, lymphoma, and leukemia (Mays and others, 1985; U.S. Environmental Protection Agency, 1999). Radium in drinking water is a serious concern because the uptake

by bone and other tissues is especially efficient for radium already dissolved in water. Because radium readily accumulates in the body, it is considered to pose a greater cancer risk than most other radioactive elements. Radiation exposure from radium received externally through washing, showering, or other uses of water is less of a concern since human skin tends to block exposure to alpha radiation and minimize penetration of beta radiation.

What Level of Radium in Drinking Water Is a Significant Health Risk?

The MCL of 5 pCi/L, established by the USEPA, is for combined radium, defined as the sum of Ra-226 and Ra-228 (U.S. Environmental Protection Agency, 2000). Although not a requirement, the USEPA encourages public water-supply systems to perform sampling and analysis for Ra-224, because this radium radionuclide and its decay products can contribute substantially to gross alpha-particle activity (Focazio and others, 2001).

How Do the Study Results Compare to the MCL?

Although radium radionuclides were present in raw water from sampled wells tapping every principal aquifer, the combined radium concentration generally was below the established MCL. Overall, few sampled wells (about 3 percent, or 40 of 1,266 wells) contained individual concentrations or combined concentrations of Ra-226 and Ra-228 greater than the MCL. No single isotope of radium was consistently responsible for the MCL being exceeded. Of the 15 principal aquifers studied, 7 had combined radium concentrations that were greater than the MCL in at least one well (fig. 2). Two principal aquifers had detections of combined radium at levels above the MCL much more frequently than the other aquifers. These included the Mid-Continent and Ozark Plateau Cambro-Ordovician aquifer system and the Northern Atlantic Coastal Plain aquifer system. Both these aquifer are composed of materials low in the parent compounds uranium and thorium, which indicates that other factors, such as geochemistry, are important in determining concentrations of radium in the water.

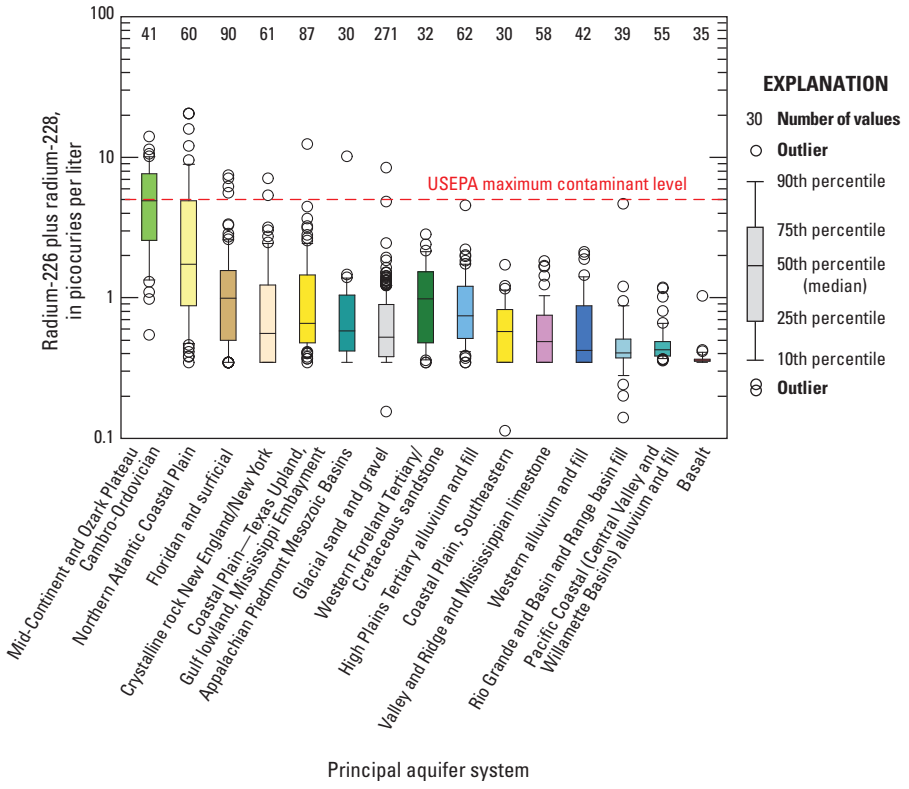


Figure 2. More than 20 percent of groundwater samples from the Mid-Continent and Ozark Plateau Cambro-Ordovician aquifer system and the Northern Atlantic Coastal Plain aquifer system had combined radium (radium-226 plus radium-228) concentrations greater than or equal to the drinking-water maximum contaminant level of 5 picocuries per liter. The next five listed principal aquifers have at least one sample that exceeded the drinking-water standard for combined radium concentrations. Aquifers that exceeded the drinking-water standard usually had geochemical conditions favorable for radium release from aquifer solids, including oxygen-poor water or acidic (low pH) conditions, usually with high concentrations of dissolved solids.

Aquifers of the Northern Atlantic Coastal Plain, primarily composed of quartz-rich sand (above), had some of the highest concentrations of radium primarily because of low pH conditions that release radium from the sediments.

What Three Water Types Are Likely to Contain Radium?

Three water types were identified by Szabo and others (2012) as likely to have elevated concentrations of radium in groundwater, including (1) low-oxygen (dissolved oxygen less than [<] 1 milligram per liter [mg/L]) water with iron or manganese; (2) acidic (pH < 6) water with high concentrations of nitrate or other acidic anions (sulfate, chloride) and calcium, strontium, or barium; and (3) water with high concentrations of dissolved solids containing one or more of the following in abundance: calcium, barium, magnesium, strontium, potassium, sulfate, or bicarbonate. These water types are indicative of conditions that may prevent radium from adsorbing onto aquifer sediments and increase the mobility of radium into surrounding water. High concentrations of combined radium and individual radionuclides Ra-226 and Ra-228 were found in water containing at least one of these chemical characteristics from the principal aquifers studied, because radium is most soluble and mobile in these geochemical conditions. Of these water types, low-oxygen conditions were the most abundant and explained the occurrence of the highest concentrations of radium in more principal aquifers than the other two water types (fig. 3).

Which Principal Aquifers Are Most Vulnerable to Radium Contamination?

The greatest frequency of elevated radium in groundwater occurs primarily in the eastern and central United States (fig. 1). About 98 percent of the sampled wells that had combined radium concentrations greater than the MCL were located east of the High Plains. More than 20 percent of wells sampled in the Northern Atlantic Coastal Plain aquifer system and Mid-Continent and Ozark Plateau Cambro-Ordovician aquifer system had combined radium concentrations greater than or equal to the MCL. The single highest concentration of combined radium in this study (20.4 pCi/L) was detected in water from the Northern Atlantic Coastal Plain aquifer system (fig. 2). Among the remaining five principal aquifers with elevated radium concentrations, the Floridan aquifer system had the greatest frequency of sampled wells with combined radium concentrations greater than the MCL, followed by the granitic and metamorphic crystalline rocks of New England/New York, the Mesozoic sedimentary basins in or adjoining the Appalachian Piedmont, the Gulf Coast Coastal Plain aquifer systems, and the glacial deposits.

Results of a previous study have shown that some parts of the Midwest have significantly higher than average combined radium concentrations in groundwater (U.S. Environmental Protection Agency, 2010); however, results of this current study indicate that radium occurrence is not only related to source rock type but also to aquifer geochemistry. Many aquifers containing the highest concentrations of combined radium in the eastern and central United States had oxygen-poor water containing particularly high concentrations of iron and manganese and, on occasion, high dissolved solids concentrations. These factors were found mostly in the Mid-Continent and Ozark Plateau Cambro-Ordovician and Floridan aquifer systems (figs. 2, 3), but also in the Appalachian Piedmont Mesozoic Basins, Crystalline Rocks of New England/New York, and the Glacial sand and gravel aquifers.

In the East, the shallow parts of the Northern Atlantic Coastal Plain aquifer system are unconfined and composed of unconsolidated quartz-rich sand with little acid-buffering capacity, which typically results in acidic (low pH) conditions that are optimal for dissolving or desorbing radium. The shallow part of this aquifer system is susceptible to the effects of urban and agricultural activities on the land surface, including septic-effluent disposal and fertilizer and lime applications, all of which can leach nitrate, phosphate, calcium, magnesium, and other dissolved solids into groundwater.

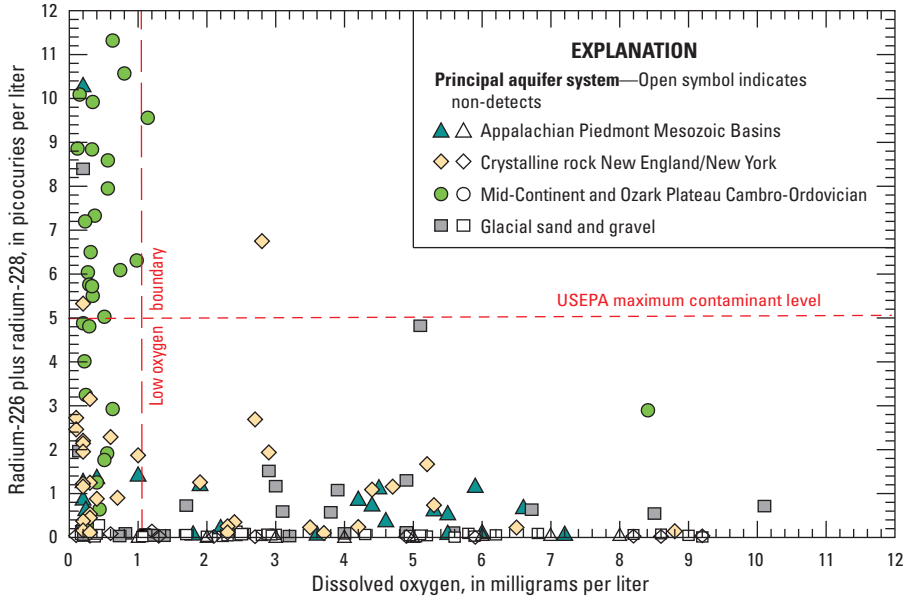


Figure 3. Oxygen-poor groundwater contains elevated concentrations of combined radium (radium-226 plus radium-228) compared to groundwater with higher dissolved oxygen concentrations. Low-oxygen conditions decrease the likelihood that radium will adsorb to aquifer materials and increases the mobility of radium into groundwater. In this study, low-oxygen conditions were present in many aquifers, but were generally found more frequenty in the Appalachian Piedmont Mesozoic Basins, crystalline rock New England/New York, Mid-Continent and Ozark Plateau Cambro-Ordovician, and parts of the glacial aquifer systems in the eastern United States.

Treatment Options for Radium in Drinking Water

A number of treatment options are available to public water suppliers and homeowners for removing radium from drinking water. Ion-exchange methods, also known as water softeners, can remove radium from water, in addition to calcium and magnesium, which cause hardness (Lucas, 1987; Szabo and others, 2008). Sodium is used to replace (exchange) these constituents in water. For some people, such as for those on a low-salt diet, sodium can cause other health concerns. A major benefit, however, is that this type of treatment system can readily be used to treat water for the whole house. Reverse osmosis systems may provide an alternative means of treatment; however, this type of treatment system can produce only small volumes of treated water and cannot be used to treat water for the whole house. Also, green sands containing the mineral glauconite provide a natural treatment option for home owners and treatment facilities. Public water supplies have many other choices for treating the water to remove radium, including ion exchange, iron removal, and lime softening among others (Zapecza and Szabo, 1988). For more information on compliance options for the USEPA Radionuclides Rule, see *http://water.epa.gov/lawsregs/rulesregs/sdwa/radionuclides/compliancehelp.cfm*. Public water suppliers and private homeowners can contact the National Sanitation Foundation (*www.nsf.org*) and the Water Quality Association (*www.wqa.org*) for assistance in determining the best treatment options. In addition, the National Ground Water Association (*www.ngwa.org*) is dedicated to education efforts aimed at reducing contaminant risks to water supplies and provides a summary of treatment considerations and an overview of strategies for well owners (Treyens, 2009).

These chemical constituents can be involved in ion exchange and acidification reactions that are not readily buffered by quartz sand and thereby stimulate the release of radium from aquifer materials into the groundwater.

East of the High Plains, waters with high dissolved solids typically had low dissolved oxygen concentrations (< 1 mg/L) and generally did not contain enough sulfates to form insoluble barium-radium sulfate mineral. As a result, radium was mobile and detectable. Groundwater in the High Plains and to the west also contained high dissolved solids but was usually rich in oxygen and contained concentrations of sulfate high enough to form insoluble barium-radium sulfate mineral. High concentrations of dissolved oxygen and sulfate can limit radium concentrations in groundwater by increasing the likelihood that radium is adsorbed or precipitated.

Drinking-water sources in groundwater aquifers may be vulnerable to contamination from radium, a known carcinogen. Knowledge of the geochemical conditions in the Nation's aquifers helps water resource managers understand where high concentrations of radium may occur in groundwater.

Possible Implications and Use of These Findings

Understanding the factors controlling radium occurrence in groundwater can help managers anticipate conditions in which radium may be elevated, prioritize monitoring studies, improve decisions related to land-use planning, and assist well owners in determining suitable locations for new drinking-water supply wells. The findings help to identify how and why radium occurrence can vary in wells from different geographic regions, principal aquifers, and geochemical environments and may help target the appropriate level of assessment, testing, and protection to aquifers and geographic areas of most concern for elevated radium concentrations and, thus, human health concerns. Principal aquifers with relatively high drinking-water ingestion risks for radium identified in this study (fig. 2) were the same aquifers identified by Focazio and others (2001), despite different sampling approaches used (random and targeted, respectively).

An example of the usefulness of the application of these findings is New Jersey's Private Well Testing Act (New Jersey Register, 2002). Its regulations became effective in 2002, requiring testing for gross alpha-particle activity in domestic wells of the Northern Atlantic Coastal Plain aquifer system in New Jersey, which is one of the most vulnerable regions in the United States to radium occurrence (Szabo and others, 2005). Also, the information can be particularly useful to private homeowners with domestic wells, because these water systems generally are not monitored regularly for radium, unlike public-supply wells that are monitored under the Safe Drinking Water Act (U.S. Environmental Protection Agency, 2000; Focazio and others, 2001). Improved information is needed on the number of people who rely on drinking water from domestic wells in specific regions that tap specific aquifers, and continued public education and testing of domestic wells is advised, particularly for naturally occurring inorganic chemicals (DeSimone and others, 2009).

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This fact sheet highlights the major findings and implications of the study and serves as a companion product to a journal article by Szabo and others (2012) that presents more detailed and technical information The URL to access the report online is *http://water.usgs.gov/nawqa/trace/radium/*.

This publication is accessible online at *http://pubs.usgs.gov/fs/2010/3113/*.