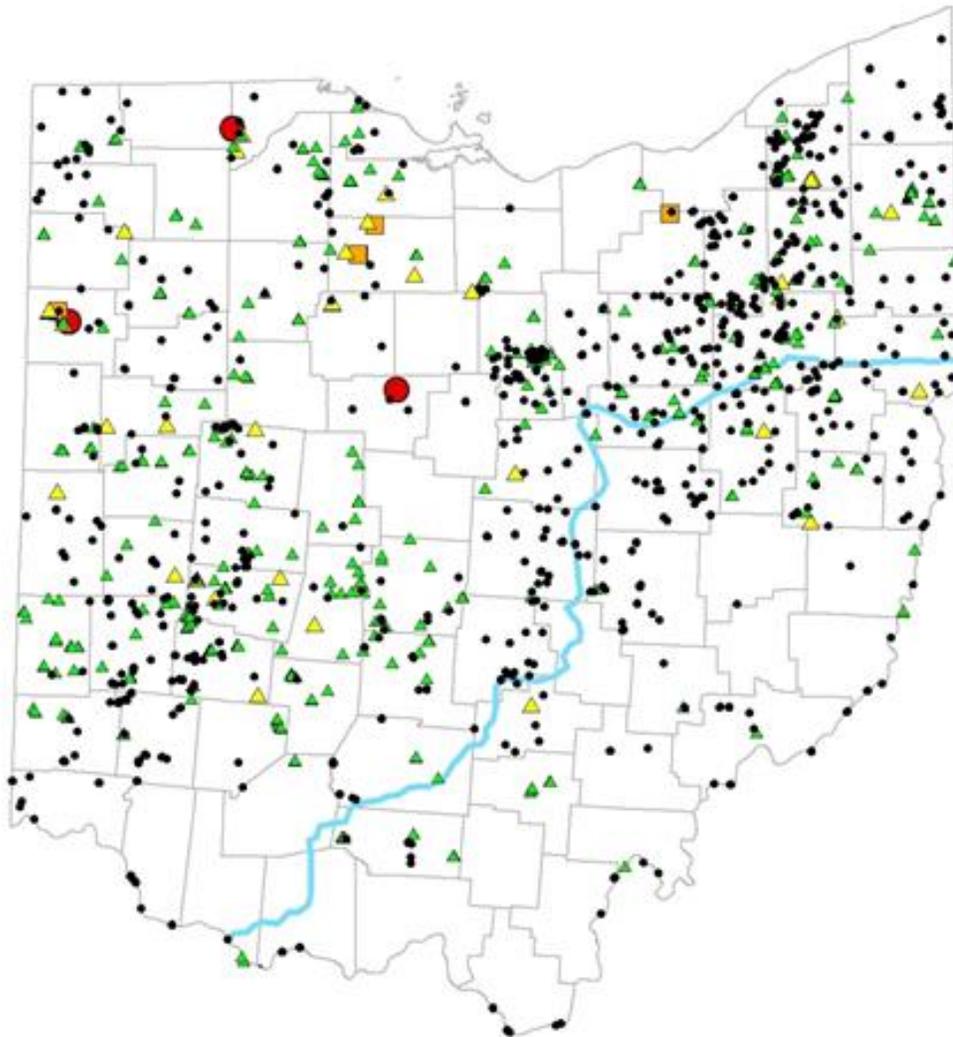


# Radionuclides in Ohio's Ground Water



Division of Drinking and Ground Waters  
Technical Series on Ground Water Quality  
July 2015

## **The Technical Series on Ground Water Quality**

This series of reports provides information to the professional/technical community about ground water quality in Ohio's aquifers. These reports use data from:

- the ambient ground water quality monitoring program; and
- the public water system compliance programs.

These data, representative of raw water, are used to characterize the distribution of selected parameters in ground water across Ohio. The goal is to provide water quality information from the major aquifers, exhibit areas with elevated concentrations, and identify geologic and geochemical controls. This information is useful for assessing local ground water quality, water resource planning, and evaluating areas where specific water treatment may be necessary.

A series of parallel fact sheets, targeted for the general public, provide basic information on the distribution of the selected parameters in ground water. The information in the fact sheets is presented in a less technical format, addresses health effects, outlines treatment options and provides links to additional information.

### **Disclaimer**

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### Abstract

In Ohio, gross alpha and radium radiation are generally low, with very few public water systems exceeding the gross alpha Maximum Contamination Contaminant Level (MCL) value of 15 pCi/L or the combined radium-226/radium-228 MCL value of 5 pCi/L. The widespread low levels in Ohio's ground water suggest the predominant sources of gross alpha and radium are from low concentrations of naturally occurring uranium and thorium and their daughter products (including radium and radon) within Ohio's geologic strata. The distribution of radionuclide detections is consistent with the occurrence of uranium and thorium, based on their geochemistry and geologic processes, but the amount and limitations of the data do not allow complete documentation. These low levels of radionuclide radiation are consistent with Ohio's surficial and shallow bedrock geology and the geologic environments of Ohio's major aquifers. The processes that concentrate uranium and thorium in Ohio geologic settings are discussed to support the conclusion that the concentrations of radionuclides are low in Ohio's aquifers.

### Introduction

#### Properties of Radionuclides

Radionuclides are atoms with unstable nuclei. The decay of unstable parent atoms produces daughter atoms with the release of energy or radiation. The types of radiation released during radioactive decay include alpha particles, beta particles and gamma rays (type of photon). In a decay series, the decay of radioactive parent and daughter atoms produces a series of intermediate radionuclides, until a stable atom is produced. The rate of decay is described by the half-life of an isotope (same element with different number of neutrons). In one half-life, 50 percent of the atoms of a radioactive isotope break down to a daughter product with release of radiation. Isotopes with short half-lives produce more decays per unit time, referred to as higher activity, than isotopes with longer half-lives.

#### Sources of Radionuclides in Ohio's Ground Water

The most common radionuclides found in ground water are decay products of naturally occurring uranium and thorium. Radionuclides usually enter ground water through natural processes of chemical weathering and dissolution of minerals containing uranium and thorium or their daughter products. Uranium and thorium have crustal abundances of 2-3 and 12 parts per million (ppm), respectively.

The solubility of the radionuclides and the compounds they form control their concentrations in ground water and surface water. Because the various decay products have different solubilities and mobilization characteristics from their parent atoms, decay products can migrate away from their parent radionuclides. The intensity of radioactivity in water (gross alpha or gross beta) depends on the presence, concentration and activity of parent and daughter radionuclides. Because the parent and daughter atoms exhibit different geochemical solubilities and half-lives, a high concentration of the parent does not assure a high concentration of a daughter product or a high level of radiation in water. The concentration and activity of dissolved isotopes are the critical factors in determining the radioactivity of a water sample. The complexities of radioactive decay, the decay products present, the range of isotope half-lives and the health concerns of radioactivity make evaluating radionuclides more complicated than measuring the concentration of a single element or compound.

Radioactive materials are used in medical diagnostics and treatments, electricity production, commercial products, research and nuclear weapons. Human activities such as disposal of radioactive wastes may increase the levels of radioactive materials found locally. Contamination of ground water can

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occur from natural sources or from release of anthropogenic radioactive materials due to improper waste storage, leaks or transportation accidents. The widespread occurrence of gross alpha radiation and radium and the lack of local areas with significantly elevated radioactivity in ground water used by Ohio's public water systems (PWSs), suggest that naturally occurring uranium and thorium are the primary source of these contaminants in Ohio.

### Safe Levels of Radionuclides in Drinking Water

The MCL is the maximum allowable level of a contaminant that may be present in drinking water distributed by a public drinking water system without a significant risk of causing health problems. For radionuclides, MCLs are established for radiation levels and concentrations, and consequently, multiple units of measure are used depending on the characteristic to be measured.

- The amount of radiation emitted per minute in a liter of water is measured as picocuries per liter (pCi/L), 1 pCi/L = 2.2 disintegrations per minute per liter.
- A radiation dose, for example, the amount of ionizing radiation delivered to the body in a year, is measured as millirems per year (mrem/yr).
- A concentration, the amount of a dissolved contaminant in a liter of water, is measured as micrograms per liter ( $\mu\text{g/L}$ ); at low concentrations, 1  $\mu\text{g/L}$  is equivalent to one part per billion (ppb).

The MCLs for radionuclides are: 15 picocuries per liter for alpha particle emissions, excluding radon and uranium; 5 pCi/L for combined radium-226/radium-228 isotopes; 4 mrem/yr for beta particle and photon emitters; and 30  $\mu\text{g/L}$  for uranium.

### Radionuclide Data Evaluated

The data used in these technical reports is generally raw water from the Ambient Ground Water Quality Monitoring Program (AGWQMP). The AGWQMP, however, does not include data on radionuclide parameters, so data collected for compliance programs was used to describe the distribution of radionuclides. PWS compliance sampling data is stored in the Safe Drinking Water Information System (SDWIS) and data collected between 1999 and April 2013 was used in this analysis. Only community public water systems are required to collect radionuclide parameter data, and these samples are generally collected at entry points to the distribution system. Thus, most of the sample results are treated samples, although some compliance data, including new well samples, are raw water samples and their results include radionuclide parameters. Treatment varies from system to system, but many community systems soften their water. Softening removes calcium and radium, one of the radionuclides of concern. Other treatment processes, such as rad removal and reverse osmosis, can also reduce concentrations of radionuclides. Thus, the treated water at many public water systems exhibits radiation levels that are likely to be less than the raw, untreated water.

The use of predominantly treated water compliance data is not ideal. However, this report is based only on data for PWSs that do not implement softening, rad removal or reverse osmosis (RO). While this makes the data more representative of raw water, it still includes data for systems that are implementing other types of treatment (for example, iron removal) that could remove radium.

**Gross Alpha** - Gross alpha particle activity is a measure of the total radioactivity due to alpha particle emission produced by radioactive decay. As such, gross alpha is an integrated measure of alpha-emitting radionuclides dissolved in water and depends on the concentration and activity of parent and daughter radionuclides that release alpha radiation. The main alpha emitters in ground water are associated with the radioactive decay series of uranium and thorium. Drinking water samples that exceed the MCL of 15

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pCi/L have additional analysis completed to determine if the presence of radon or uranium have caused the exceedence of the MCL. If radon or uranium are not significant contributors to gross alpha, the system will go to quarterly sampling to determine if an MCL is exceeded.

**Gross Beta** - Gross beta activity is an integrated measure of total beta radiation regardless of the source of the radioactive decay. The level of gross beta in water depends on the concentration and activity of radionuclides that release beta radiation. The low levels of gross beta in Ohio result in limited gross beta data. For compliance sampling, if a drinking water gross beta sample exceeds a trigger level of 50 pCi/L, additional analysis is completed to determine the source of the beta radiation and if an MCL has been exceeded. From 1999 to 2013, there were a total of 512 gross beta sample results from 276 PWSs including 104 raw water samples, for ground water across Ohio. None of these samples are above 20 pCi/L, 40 percent of the MCL trigger, where additional sampling and analysis would be required. Analysis of these results document that gross beta levels are low throughout Ohio. Statewide spatial analysis of elevated gross beta concentrations exhibits no relationship of gross beta activity with the geology of local aquifers. Consequently, gross beta will not be discussed further.

**Radium** - Radium was discovered in 1898 by Madame and Pierre Curie, who isolated it from uranium ore. Radium is unstable and was named radium because it emits rays, the energy or radiation that results from the radioactive decay of radium atoms. There are four natural isotopes of radium. Radium-226 (half-life of 1,600 years) is the most common and is generated in the decay series of uranium-238. Radium-226 decays to radon-222. Radium-228 (half-life of 6.7 years) is the next most common and is generated in the decay series of thorium-232. The decay of radium-226 emits alpha particles, and the decay of radium-228 emits beta particles. Radium is generally immobile in oxidized environments (in contact with free oxygen from the atmosphere) and mobile in reduced environments where the oxygen has been consumed by organisms or chemical processes. The distribution of radium-228 and radium-226 are discussed below.

### Distribution of Radionuclides in Ohio

#### Distribution of Gross Alpha

The distribution of alpha radiation in Ohio's ground water-based public water systems is presented in Figure 1 on the following page. These data are compliance data (3,458 samples) collected by 1,210 community and non-community non-transient PWSs to meet sampling requirements. The samples are mostly distribution water but include 493 raw water samples. Some gross alpha results include alpha radiation from uranium and radon decay, which would not be included in the calculation for determining a MCL exceedance for gross alpha. Thus, the data presented in Figure 1 are averaged results of all data at a water system and do not represent an MCL determination. A gross alpha MCL determination is an annual average and excludes uranium and radon radiation. Only a few average results exceeded the gross alpha MCL concentration of 15 pCi/L (3 of 1210 PWSs exceeded the gross alpha MCL value; 71 percent of systems have only non-detect results).

Figure 1 confirms that average gross alpha values are generally low across Ohio. This is expected because uranium and thorium are not concentrated within Ohio's major aquifers. Natural decay of uranium or thorium generates alpha radiation and radium and it is expected that lower levels of alpha radiation and radium will be found in water associated with lower concentrations of these radioactive, parent elements. The Appalachian Basin siliciclastic and platform carbonate sediments that constitute Ohio's major bedrock aquifers are generally not enriched in uranium and thorium due to the lack of igneous rocks in the stratigraphic section.

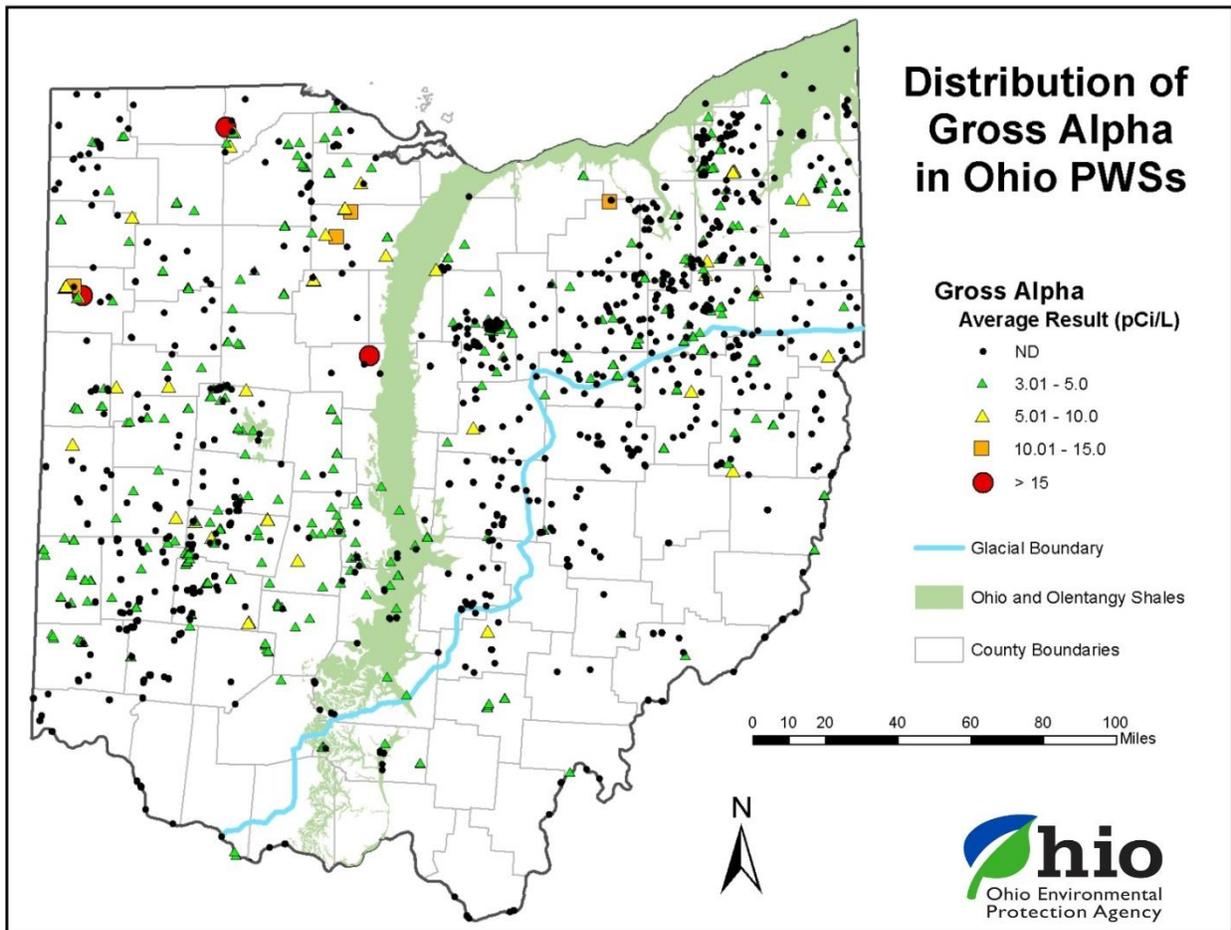


Figure 1. Average gross alpha in distribution water in PWSs without softening and RO.

The western half of Ohio where carbonate aquifers predominate exhibits more sites with average gross alpha above 15 pCi/L than eastern Ohio. Carbonate depositional environments can concentrate uranium in areas where organic matter is abundant. Subsequent weathering and dissolution of limestones and dolomites, concentrates uranium in the insoluble residue associated with calcareous or terra rosa soils. Calcareous soils developed on weathered till in western Ohio have been eroded and incorporated into younger tills deposited by more recent advances of glaciers. Harrell et al. (1993) identified these calcareous soils as a source of radon in Ohio. Radon-222 is generated in the radioactive decay series of uranium-238. The radioactive decay series releases alpha and beta radiation. Thus, calcareous soils developed on exposed tills or incorporated within Wisconsin tills provide another low level radioactive source that may contribute to alpha radiation in western Ohio.

The green overlay in Figure 1 outlines the surface exposure or subcrop below the glacial drift of black shales in central and northeastern Ohio. As explained below, black shales exhibit elevated uranium; however, no strong correlation between gross alpha in drinking water and the distribution of black shale is apparent. Public water systems do not use the black shales as aquifers due to their low permeability and poor water quality. Consequently, the Devonian shales, in spite of their elevated uranium, do not have a significant impact on gross alpha concentrations in drinking water distributed by public water systems. This statement may not be true for private wells with smaller yield requirements that utilize black shales as the local aquifer.

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Although igneous rocks are not present in Ohio aquifers, except as glacial erratics, uranium is mobile in the surface environment and can be concentrated in sedimentary environments. The Devonian black shales provide an example of uranium enrichment (10-40 ppm), which can produce areas of local elevated gross alpha radiation. The Ohio Department of Natural Resources (ODNR) reports that the elevated radon (radon-222 is a daughter product of uranium-238) associated with glacial deposits in Ohio is the result of incorporation of fragments of black shale in tills (Harrell, McKenna and Kumar, 1993). This interpretation is supported by the correlation of the location of Devonian Shale deposits, the direction of Wisconsin ice flow and areas of elevated radon.

Black shales exhibit elevated uranium because their organic material creates reducing conditions, which cause precipitation of dissolved uranium during shale deposition. Uranium is soluble in oxidizing environments (close association with the atmosphere) and precipitates rapidly when uranium-rich oxidized water encounters reducing environments (low to no oxygen) or interacts with organic material.

### Distribution of Radium-228

Figure 2 illustrates the distribution of average results for radium-228 and documents the presence of low levels across Ohio. There are a total of 2668 radium-228 sample results associated with 990 PWSs. Of these samples, 296 are raw water samples. Only one system has a radium-228 sample average that exceeds the radium MCL value (1 out 990 sites; 85 percent of sites have only non-detect results). The radium MCL is used here as a benchmark, but the data plotted are not calculated as compliance data. For compliance purposes, the MCL is an annual average for radium-226 and -228. The data plotted in Figure 2 is an average of all the data from a system, and only includes radium-228 results.

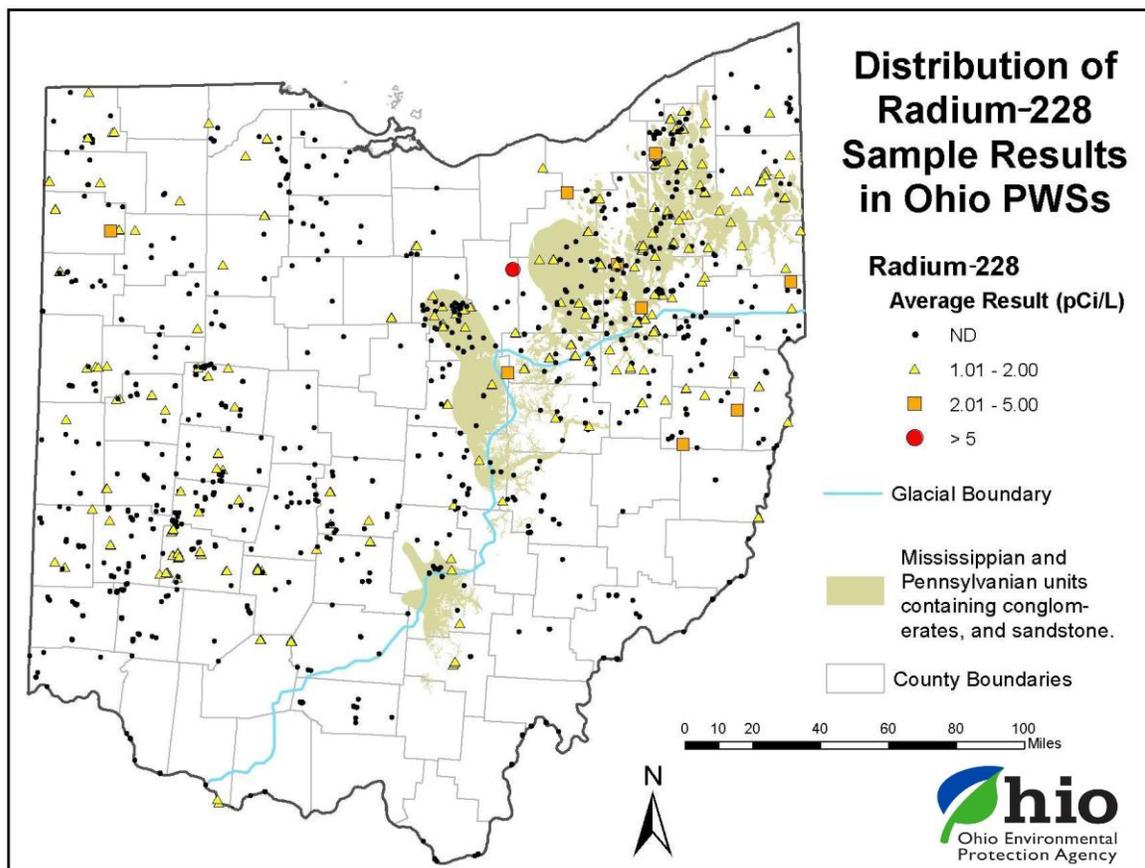


Figure 2. Average radium-228 in distribution systems of PWSs without softening and RO.

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The largest concentration of radium-228 detections occurs in northeast Ohio as illustrated in Figure 2. This area correlates roughly with the subcrop of Mississippian and Pennsylvanian units containing conglomeratic units, like the Mississippian Black Hand Sandstone and Pennsylvania Lower and Middle Pottsville including the Sharron Conglomerate/Sandstone and Massillon Sandstone indicated with the grey-green overlay. This correlation is crude and partly reflects the fact that these sandstone and conglomerate units are targeted by local well drillers as productive aquifers.

Radium-228 is derived from the radioactive decay of thorium 232. Thorium generally occurs in accessory minerals (thorite,  $\text{ThSiO}_4$  and monazite,  $(\text{Ce, La, Y, Th}) \text{PO}_4$ ), which are resistant to near surface weathering and dissolution. These thorium-rich, heavy minerals are concentrated by stream processes as detrital grains in stream or beach deposits. Consequently, radium-228 is associated with detrital concentrations of thorium-rich accessory minerals in sandstones or conglomerates. The Mississippian Black Hand Sandstone, Pennsylvanian Massillon Sandstone and Pennsylvanian Sharon Sandstone/Conglomerate illustrated in Figure 2 are good candidates for concentrations of thorium-rich detrital minerals. Another possibility for thorium accessory mineral concentration is within glacial sand and gravel deposits in Ohio's buried valley aquifers. Many of the radium-228 detections exhibited in Figure 2 are associated with buried valley or alluvial aquifers. With radium mobile under reducing conditions it is possible that the radium-228 has migrated some distance from its thorium source in reduced aquifers.

To evaluate this detrital concentration hypothesis for thorium concentration and distribution, and subsequent release of radium-228, the sites were associated with the geologic setting of the primary aquifer and evaluated. Figure 3 plots the maximum radium-228 results against groups of aquifer hydrogeologic settings. The sandstone/shale setting exhibits the highest maximum values of radium-228 and the carbonate setting exhibits the lowest, with considerable overlap at low concentrations. This is what the hypothesis predicts.

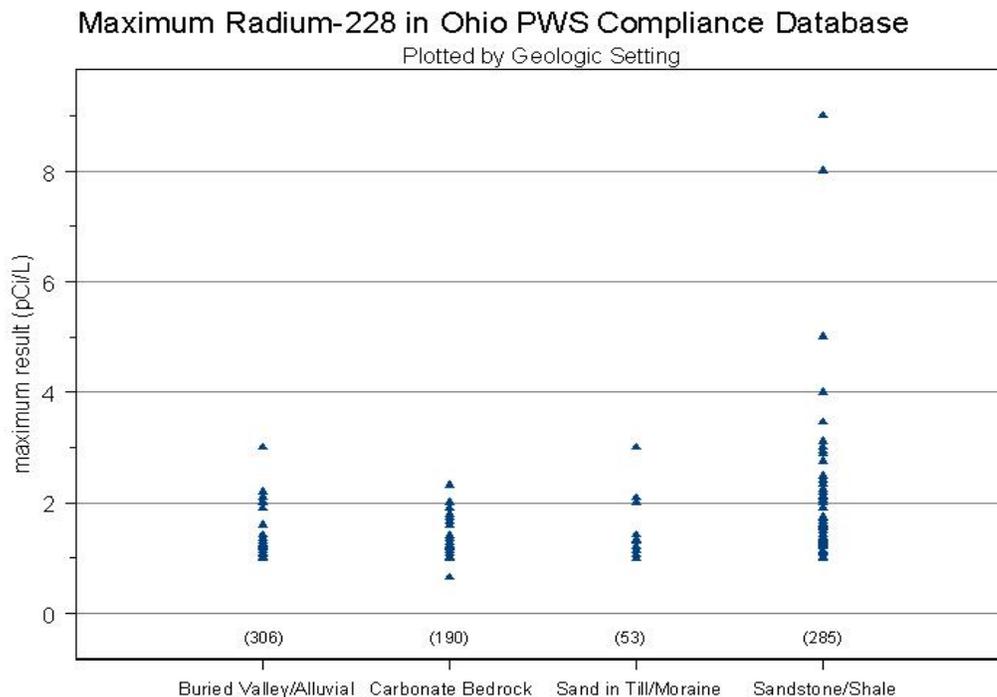


Figure 3. Maximum radium-228 plotted against aquifer setting.

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The core of the Appalachian Mountains, including igneous rocks with thorium accessory minerals, was a source area for sandstones deposited in eastern Ohio. Fluvial processes transported sediments and concentrated heavy, accessory minerals. Subsequent decay of thorium has resulted in higher concentrations of radium-228 in these rocks. The marine carbonates deposited in western Ohio had little to no continental detritus resulting in low thorium concentrations and little production of radium-228. The sand and gravel deposits within the much younger buried valleys are dominated by reworked glacial material, which includes some Canadian Shield debris, but it is diluted by locally eroded sedimentary bedrock with low concentrations of thorium-rich accessory minerals. This analysis supports the hypothesis that thorium accessory minerals in sandstone units can control the distribution of radium-228 exhibited in Figure 2. Although the detrital concentration of thorium provides an explanation for the distribution of radium-228, it is important to point out that even the sites with elevated radium-228 exhibit relatively low levels of radium-228.

### Distribution of Radium-226

Figure 4 illustrates the distribution of average results for radium-226. The data is limited because public water system regulations require radium-226 analysis only if the gross alpha is greater than 5 pCi/L or the sum of gross alpha and radium-228 exceed 5 pCi/L. The radium-226 data plotted include 310 samples distributed among 177 PWSs. Only two sites had sample average exceeding the radium MCL value (2 out of 177 sites). The limited data presented in Figure 4 does not identify significant concentrations of radium-226. There is an area of detections within the Pennsylvanian Middle and Lower Pottsville Formation in northeast Ohio, and some detections are scattered across northwest Ohio over the carbonate aquifers.

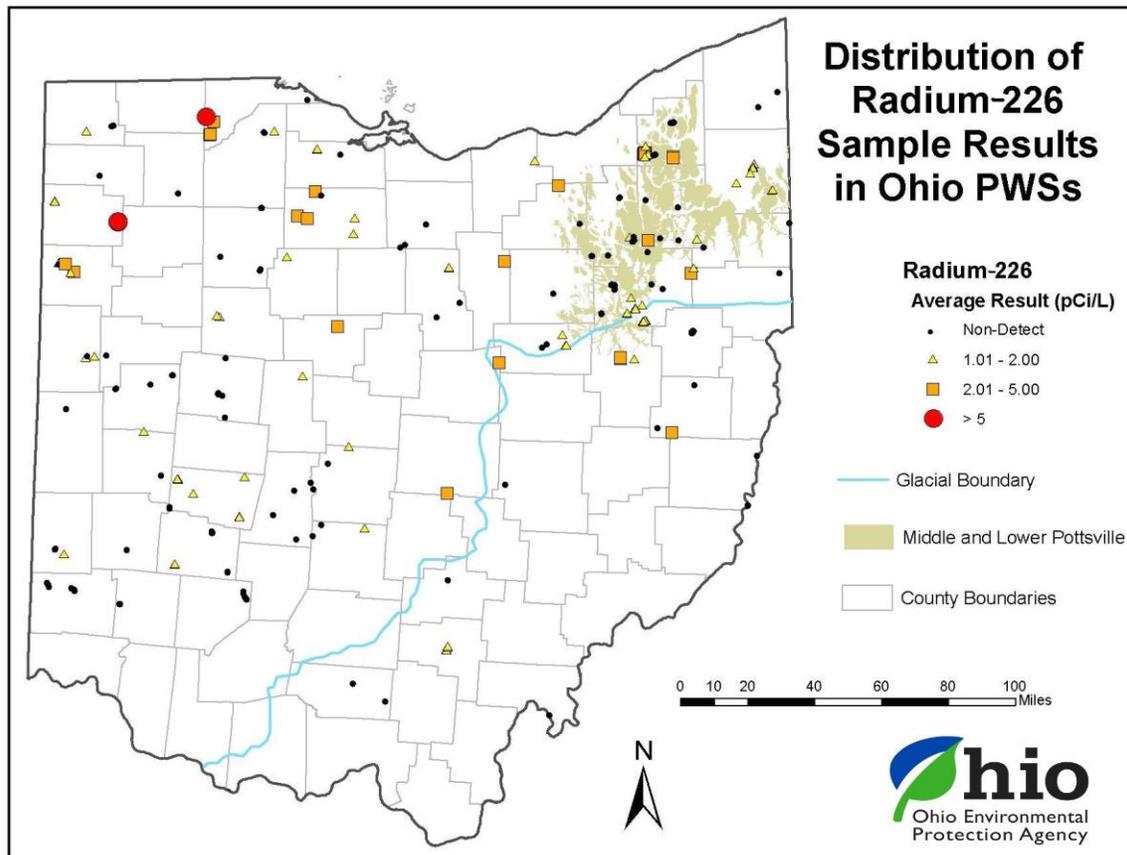


Figure 4. Distribution of average radium-226 in public water with softening or RO.

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Radium-226 is generated by the radiometric decay of uranium-238 and, consequently, areas of elevated radium-226 mirror elevated uranium concentrations. Radium-226 follows the same general pattern as gross alpha because its main source in Ohio is also uranium. The group of detections in northeast Ohio is likely related to uranium associated with Pennsylvania System coals. As discussed previously, reducing depositional environments can concentrate uranium dissolved in oxygenated water. The Pennsylvanian coal swamps were filled with organic material in stagnant, reduced water that provided conditions to concentrate uranium within the swamp deposits that became coal. Radium-226 is soluble under reducing conditions, so wells that penetrate coal seams may produce water with radium-226, if the well is drawing some water from coal seams.

The scattered presence of radium-226 detections in western Ohio is consistent with documentation of uranium concentration in marine carbonate depositional environments. The interaction of surface water with the atmosphere maintains the oxidized condition in most depositional environments. Because uranium is soluble under oxidized conditions, it is generally present in surface water at low concentrations. In carbonate depositional areas with high concentrations of organic material, the environment will become reducing and uranium can be incorporated into the carbonate deposits as complexes with organic matter. In contrast, there is usually little to no contribution of siliclastic material to a marine carbonate (thus limited thorium input), so it is common for uranium to be enriched relative to thorium in marine carbonates (Szabo et.al, 2012; Sturchio, et. al.,2001)

Further enrichment of uranium occurs within soils developed on glacial tills that include carbonate clasts. The dissolution of the carbonate clasts during soil formation concentrates the insoluble debris and forms calcareous soils. Harrell et al. (1993) suggests that these soils are a source of radon in western Ohio due to the presence of enriched uranium. Radon produced from the uranium decay moves through the soil as a gas. The mobility of the radium generated by the uranium decay in these soils is limited by the oxidizing conditions and the absorption of radium onto clays and iron hydroxides within calcareous soils. Thus, although these processes may account for radium- 226 detections in western Ohio, the limited concentration of uranium and the immobility of radium in oxidized soils limit the concentrations of radium-226 in western Ohio ground water.

Although the earlier statements are consistent with the expected distribution and mobility of uranium and radium-226, data constraints (limited number of radium-226 analyses and the fact that these samples are treated water) do not allow confirmation of these processes. The available data could be used to target locations with higher levels of radiation to evaluate the source of site-specific radionuclides. The generally low concentrations of radionuclides in Ohio, however, do not necessitate this effort.

### Conclusion

In Ohio, gross alpha and radium radiation levels are generally low, with few ground water based PWSs exceeding the gross alpha MCL value of 15 pCi/L or the combined radium-226/radium-28 MCL value of 5 pCi/L. The widespread distribution of gross alpha and radium in Ohio's ground water suggests the predominant sources are low concentrations of naturally occurring uranium and thorium and their daughter products within Ohio's geologic strata. The known presence of elevated radium and radon indicates uranium is present in the black shales in sufficient concentrations to produce areas of elevated radon, but these shales are not used as aquifers for PWSs. We have used the limited detection data to illustrate the geologic and geochemical processes that are likely to produce illustrated clusters of detections of gross alpha, radium-228 and radium-226. Although the proposed processes are reasonable and are certainly active, data constraints make it difficult to confirm these processes. These low levels of radiation are consistent with Ohio's surficial and shallow bedrock geology and the active geologic environments of Ohio's major aquifers.

**For more information** - For more information on radionuclides in Ohio's ground water, contact Ohio Environmental Protection Agency's Division of Drinking and Ground Waters at (614) 644-2752.

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