

# Polonium-210 Occurrence in Minnesota's Aquifers: A Pilot Study

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# Summary

Ionizing radiation is part of our natural environment, and radioactive elements (radionuclides) from rock and soil are commonly found in Minnesota's groundwater and drinking water. Polonium-210 (Po-210) is a naturally occurring radionuclide that has been found in drinking water aquifers in other states, but very little is known about its occurrence in Minnesota.

Human exposure to Po-210 from drinking water could have a large impact on human health. Po-210 is easily absorbed by the body and is distributed to all tissues, where its radioactive decay results in ionizing radiation exposure. Po-210 produces an especially damaging type of radiation called an alpha particle. Radiation is a known human carcinogen and exposure increases the lifetime risk of developing cancer.

The goal of this pilot study was to determine if Po-210 occurs at measureable levels in community water supply wells that were known to have elevated levels of alpha activity. Thirty-six samples (including three samples of drinking water treated to remove radionuclides) across eleven major aquifers were tested for Po-210 activity. Lead-210 (a related radionuclide) was analyzed in ten samples to better understand the Po-210 activity results. Total, or gross alpha, activity was measured to evaluate the relationship between Po-210 and total alpha activity.

Po-210 was found above 0.1 picocuries per liter (pCi/L) in 67 percent of groundwater samples. A threshold of 0.1 pCi/L was chosen as this level represents an approximate 1:100,000 lifetime cancer risk level and was also near the limit of detection by the laboratory. Po-210 was found at variable levels both within and across different aquifers. The highest activity found was 4.99 pCi/L in a Mt. Simon aquifer well located in south-central Minnesota. Po-210 activity varied within the Mt. Simon aquifer from below detectable limits in one sample from southeast Minnesota to three samples above 1 pCi/L in south-central Minnesota. Lead-210 was also detected, with highest activities found in treated water samples in eastern Minnesota. A comparison of Po-210 and lead-210 results suggests that significant amounts of Po-210 comes directly from sediment around the well, rather than from the decay of existing soluble lead-210 already in the water into Po-210.

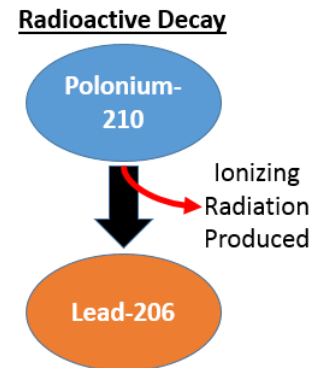
Results from this pilot study show that Po-210 and lead-210 occur in Minnesota aquifers. In some areas, Po-210 and lead-210 could make up a significant portion of the possible radiation dose in drinking water. Three of the 36 wells tested found Po-210 in excess of a 1:10,000 lifetime cancer risk level in untreated water. MDH found that naturally occurring Po-210 and lead-210 are a potential health concern depending upon the effectiveness of water treatment, especially in the Mt. Simon sandstone aquifer. In the wells tested, existing gross alpha screening is adequate to trigger appropriate action for reducing alpha radiation in drinking water. However, this pilot study was only able to address a very limited number of source water wells, prior to treatment in most cases, in a constrained geographical region of Minnesota. Occurrence of Po-210 at high levels in other aquifers in Minnesota is not anticipated but remains an area for possible future investigation. MDH will continue to follow national Po-210 occurrence studies for possible settings where Po-210 may be present at elevated levels.

# Introduction

Many Minnesotans rely on groundwater as a high quality source of drinking water. In some areas, this groundwater resides in soil and rock that naturally contains some uranium and thorium, both radioactive elements. These radioactive elements are unstable and decay naturally into other radioactive elements (such as polonium and radon). While the total amount of radioactive elements in soil is small, it is significant enough to raise concerns about health risks in some situations. For two decades, public health agencies have been communicating the health risk from exposure to one decay product of uranium— radon gas. Other radioactive elements can be present in groundwater and lead to radiation exposure if that water is used for drinking.

## Background

Radionuclides are unstable radioactive elements that change into a new element over time (called radioactive decay). When this decay occurs, a form of energy known as ionizing radiation is produced (see Figure 1). Ionizing radiation, or 'radioactivity', is surprisingly common in the environment. The earth naturally produces a low level of background radiation from radionuclides, which results in a constant human exposure to ionizing radiation. Exposure to background radiation is unavoidable, but it is important to limit sources of radiation exposure as much as possible. Po-210 is one of many naturally-occurring radionuclides in the earth's crust and can be present in water aquifers. For some Minnesotans, drinking water containing radionuclides can be a significant cancer risk even though it is a small exposure to ionizing radiation. These small daily exposures, if continued for many decades, can add up to an elevated cancer risk (1:5,000, for instance). This increased risk from radionuclides is very small when considering the proportion of people that get cancer within their lifetime (nearly 1:2). Nevertheless, environmental exposures to ionizing radiation increase a person's overall lifetime cancer risk and a major public health goal is to minimize the exposure of communities to radiation.



*Figure 1. The decay of a radionuclide produces a new element and releases energy in the form of ionizing radiation.*

## Health Risks from Polonium-210

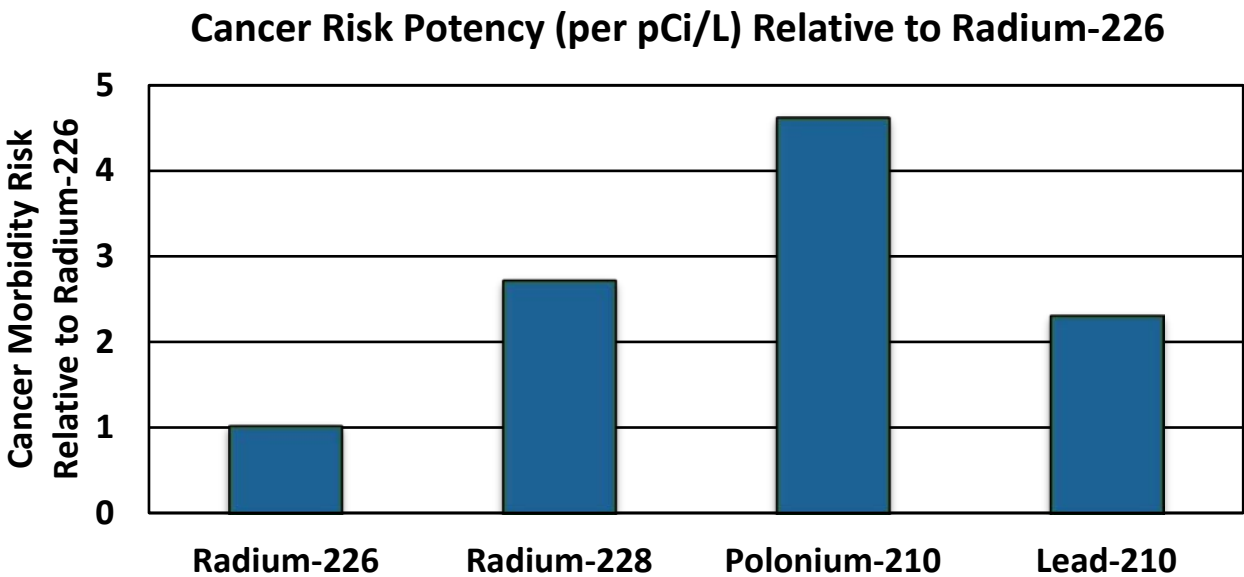
Ionizing radiation is a well-known human carcinogen. Exposure to small amounts over a lifetime results in a gradually building, but slight, increase in lifetime cancer risk (NAS/National Research Council, 2006). However, not all exposures to ionizing radiation cause equal damage. This is because of the properties of the different forms of radiation. To help understand this difference, imagine a tennis ball and a baseball; they are both balls, but a baseball does more damage than a tennis ball when it hits someone. Likewise, different types of radiation can cause different amounts of damage to people. The three forms ionizing radiation can take are:

photons (gamma, x-rays, and some ultraviolet), beta particles, and alpha particles. Alpha particles are particularly harmful to our bodies because the damage they create is not easily repaired. Alpha radiation (the baseball) is generally considered to be twenty times more potent than x-rays and gamma rays (the tennis balls).

Even though alpha particles are very damaging, they cannot pass through the skin. The greatest health risk from alpha exposure comes from eating, drinking, or breathing alpha-containing substances, such as breathing radon gas or drinking water with radionuclides.

The United States Environmental Protection Agency (USEPA) has established four Maximum Contaminant Level (MCL) standards for radionuclides, along with an MCL goal (MCLG) of zero, used for all known human carcinogens (USEPA, 2000). The gross (total) alpha MCL is set at 15 pCi/L and includes alpha particle production in water by the mixture of radionuclides present, including Po-210. The radium MCL is set at 5 pCi/L for the sum total activity of radium-226 and radium-228. These levels are intended as conservative values to limit radiation exposure in water to very low levels due to the many other sources of radiation exposure in our daily lives (such as natural background, air travel, and medical procedures).

While there is no MCL for Po-210 or lead-210, MDH used published USEPA cancer risk coefficients to estimate a 1 in 10,000 lifetime cancer risk level at 1.1 pCi/L for Po-210 and 2.2 pCi/L for lead-210. These 1 in 10,000 lifetime cancer risk levels are within the risk range (1 in 1,000,000 to 1 in 10,000 cancer risk) used by USEPA for protection of public health. Po-210 is more potent and potentially more harmful than other radionuclides found in Minnesota's groundwater. Lead-210 is about as potent as radium, which is commonly measured (see Fig. 2).



**Figure 2.** Groundwater radionuclide relative potency. At equal pCi/L activity in groundwater, radionuclides present large differences in lifetime cancer risk, spanning nearly five-fold for these four radionuclides known to occur in Minnesota.

## Study Aims

Prior to this pilot study, little was known about the occurrence of Po-210 in Minnesota's groundwater resources. Po-210 had been detected in other states at levels of concern (Ruberu, Liu, & Perera, 2007; Seiler & Wiemels, 2012). Radium-226, a radionuclide that eventually produces Po-210 (through lead-210 and other radionuclides), has been found at elevated levels in Minnesota's aquifers (Lively et al., 1992; Lundy, 2010), leading to concern that Po-210 may be present as well.

The USEPA had previously identified Po-210 and Pb-210 as contaminants of interest in the Unregulated Contaminant Monitoring Rule, but no monitoring under this program was completed due to a lack of analytical methods (USEPA, 2000). More sensitive instrumentation and better processing techniques have been developed over the past decade, which now allows measurement of Po-210 at low levels for a reasonable cost.

This pilot study was designed to assess the presence or absence of Po-210 in many of Minnesota's drinking water aquifers currently impacted by elevated levels of alpha-emitting radionuclides. Because gross alpha measurements do not include a consideration of the cancer potency of individual radionuclides, the presence of Po-210 in drinking water supplies deserves careful consideration. This pilot study focused on measuring Po-210 because it is the most potent of the alpha emitters and is known to naturally occur in groundwater. In addition, this study evaluated relationships between measurements of gross alpha, Po-210, and other radionuclides, in order to test possible ways to estimate Po-210 levels from other, more easily and routinely conducted analyses.

## Methods

The feasibility of quantifying Po-210 at very low activity levels in drinking water and selecting wells for sampling that were likely to contain Po-210 were the subject of method considerations. These two major methods areas are discussed below.

### Feasibility of Analysis

MDH selected a contract laboratory that could measure Po-210 in drinking water at low, environmentally relevant, levels. Samples containing greater than 1 pCi/L of Po-210 were also analyzed for lead-210 activity, another radionuclide of concern in drinking water. The MDH Public Health Laboratory (PHL) measured gross alpha levels in most samples.

In this study's early planning stages it was discovered that the United States Geologic Survey (USGS) planned to include Po-210 and lead-210 in the National Ambient Water Quality Assessment (NAWQA) sampling, which began in the summer of 2014. MDH collaborated with NAWQA staff with the immediate goal of taking split, or identical, water samples and sending them to different contract laboratories to compare analytical results. The USGS data were not



available at the time of this report, but personal communication between MDH staff and USGS staff noted fairly good agreement between laboratories.

## Sampling Site Selection

Selecting groundwater wells to sample for this study was challenging because Po-210 has not been measured in enough places for us to understand what conditions cause elevation of Po-210 in water. Groundwater with Po-210 varies considerably across the country, based on data from wells in Maryland, Florida, and Nevada (Seiler & Wiemels, 2012). However, in Minnesota, data on gross alpha were available and might indicate sites where Po-210 could be in water based on the presence of alpha particles in the water. Since the finalization of USEPA's Radionuclide Rule in 2000, water systems have monitored for gross alpha particle activity in groundwater for compliance with the MCL of 15 pCi/L. MDH used these data to select sample sites for the pilot based on gross alpha levels, while also spreading sampling out among a variety of aquifers and conditions.

The sampling sites included 32 wells from 26 drinking water supply systems. All sites were selected based on gross alpha levels in sources of drinking water before the water supplier treated the water to remove radionuclides.

# Results

## Polonium- and Lead-210

A summary of the overall Po-210 data are provided in Table 1. Po-210 was found above MDH's cutoff of meaningful activity, 0.1 pCi/L, in 64 percent of 36 samples. The value listed in Table 1 for average activity of lead-210 is likely an exaggeration of the true lead-210 mean throughout the sampling area because this radionuclide was only measured in samples with elevated Po-210. Table 2 provides Po-210, gross alpha, and lead-210 activity data listed in order of Po-210 activity. The samples with 'entry point' names in Table 2 are not source water well samples, but samples taken as water enters the distribution system following water treatment specific to each system.

Po-210 activity was low in all of the aquifers tested except the Mt. Simon aquifer (Figure 3). Within the Mt. Simon aquifer, there was a high degree of variability, based on geographic location and possible well depth and other unknown factors (Figures 3 and 4).

The highest Po-210 result was 4.99 pCi/L in a Mt. Simon aquifer well in south-central Minnesota. The next highest Po-210 sampling results, activities above 1 pCi/L, were found in nearby Mount Simon aquifer wells. The highest lead-210 activities, 2.9 pCi/L and 1.5 pCi/L, were found in samples taken following water treatment in two east central Minnesota communities. The lead-210 data demonstrate that lead-210 and Po-210 are not in close equilibrium with each

other (their activity in groundwater is not equal). Therefore, Po-210 activity is not coming from lead-210 decay into Po-210 in groundwater (Table 2).

## Gross Alpha

The results of the gross alpha analyses were compared to Po-210 results. Five of the 36 samples did not include gross alpha activity (these samples were collected by USGS for split sampling analysis of Po-210 activity only). In total, there was not a strong correlation between gross alpha activity and Po-210, especially in the 15-30 pCi/L range of gross alpha values.

Changes in gross alpha activity over time were also evaluated, in order to understand the usefulness of a change over time measurement as an indicator of Po-210 activity in future studies. However, the change in gross alpha activity over time was not consistent across samples with elevated Po-210, and meaningful estimations of Po-210 activity could not be made with gross alpha change.

## Discussion

Minnesota's groundwater contains a wide variety of radionuclides from the decay of uranium and thorium. Each distinct radionuclide has its own potency for increasing cancer risk. Many different radionuclides are contributing to the overall alpha particle activity (gross alpha) that is routinely measured in drinking water as part of USEPA Safe Drinking Water Act regulations. MDH was interested in measuring Po-210, as it has the greatest cancer potency of these commonly occurring radionuclides. MDH found that the proportion of Po-210 activity to total gross alpha activity in each water sample to be quite low. This result was reassuring – Po-210 did not make up a large proportion of gross alpha activity. Therefore, compliance with the gross alpha MCL of 15 pCi/L in the aquifers tested is likely to indicate low Po-210, but the exact correlation between gross alpha and Po-210 is problematic, and the pilot study only covered a small number of wells in a constrained geographic region of the state.

Gross alpha activity, either by single measurement or change in activity over time, did not correlate well with Po-210 activity in groundwater. This is in agreement with other studies demonstrating difficulty using gross alpha methods to capture Po-210 activity in groundwater (M F. Arndt et al., 2010; M. F. Arndt & West, 2008). One of the limitations of this study is that wells with low gross alpha activity were not tested, and a well with low gross alpha level could still contain a meaningful level of Po-210, due to the lack of sensitivity for Po-210 in gross alpha testing.

The presence of Po-210 in groundwater was not usually associated with equal activity of lead-210. As lead-210 decay produces Po-210, the presence of Po-210 could be explained by lead-210 decay if these two radionuclides were nearly equal in activity in groundwater. Results from this study suggest otherwise, that most Po-210 comes directly from surrounding rock and sediment, rather than from the decay of lead-210 in groundwater.

Despite the low overall contribution of Po-210 to gross alpha in these samples, the seemingly small amounts present are an important component of possible human exposure to radiation from untreated drinking water sources. The median (0.13 pCi/L), average (0.39 pCi/L), and maximum (4.99 pCi/L) levels of Po-210 correspond to lifetime cancer risks of approximately 1 in 100,000, 1 in 30,000, and 1 in 2,000, respectively. Three of 36 wells in this study contained Po-210 levels greater than 1 pCi/L, with a lifetime cancer risk of more than 1 in 10,000. These are conservative estimates of risk because MDH assumed the level of Po-210 remains the same over time (which is currently unknown) and that higher than average amounts of tap water are consumed daily, for many decades. Additionally, the samples with the highest Po-210 levels were found in source water before it is treated for removal of radionuclides. Over 90% of the samples taken in this study were collected before water treatment that removes radionuclides. While the specific effects of treatment on Po-210 are unknown, it is generally accepted that the commonly used treatment techniques should reduce Po-210 levels in finished drinking water.

Po-210 health risks are specific just to the amount of Po-210 in the groundwater. Other naturally occurring radionuclides present in the groundwater, such as radium and lead-210, would add to this risk. Individual alpha emitting radionuclides other than radium are not routinely measured in groundwater, and the results of this study suggest that more specific measurement of individual radionuclides may be important in aquifers where Po-210 is present.

This pilot study does not show how effectively water treatment removes Po-210 and lead-210. However, based on very limited lead-210 sampling in source water (8 samples) and treated water (2 samples), lead-210 may be slightly concentrated by some forms of treatment that bind radium in solid material as a way to reduce radium concentration in water. Excessive radium loading and/or poor flushing of radium from the filter bed could lead to eventual radium buildup and production of radionuclides such as radon and lead-210 from radium decay. As radium decays into the soluble radionuclide radon, this radionuclide could enter the treated water flow following pressure filtration. Paired source water/treated water samples are needed to assess how treatment processes and operation and maintenance of plants may be affecting lead-210 or Po-210 levels in finished water. Discovering new information on treatment effects will require a dedicated follow-up study. The limited number of treated water samples in this pilot study does not allow any conclusions to be drawn regarding possible effects of water treatment.

An objective of this pilot study was to examine many aquifers. The results suggest that the Mt. Simon aquifer, long known for high levels of gross alpha and radium, should be the subject of future investigations to understand why Mt. Simon aquifer community wells demonstrate such variable Po-210 activities. Mt. Simon wells near the Minnesota River in south-central Minnesota contained 1-5 pCi/L of Po-210 activity while Mt. Simon wells in southeast Minnesota contained Po-210 near or below detection limits. Differences in depth, recharge, redox, and other factors may determine Po-210 mobilization to groundwater in Minnesota. As information continues to be generated on radionuclide occurrence in Minnesota, an important application of this growing knowledge base would be to target domestic wells finished in aquifers with known radiological issues (such as the Mt. Simon and Jordan aquifers). In the same way that radon is

now commonly measured in homes, the need for assessment of alpha emitting radionuclides in domestic wells may also grow. Currently, Minnesota private well owners rarely test for alpha emitters in their water supply even though many wells draw source water that is likely to contain some alpha activity.

## Conclusions

While only a pilot project, this study represents the largest collection of Po-210 and lead-210 drinking water well occurrence data gathered in the state of Minnesota. This study targeted wells with known radiological issues to better understand the impact of Po-210 on Minnesota's important groundwater resources. This project has demonstrated the importance of Po-210 surveillance in Minnesota's groundwater and the feasibility and affordability of conducting these measurements. While levels of Po-210 were, overall, below levels associated with a 1 in 10,000 cancer risk, a few source water samples in the Mt. Simon aquifer exceeded the acceptable health risk range for Po-210 and also the gross alpha MCL. While the gross alpha MCL is an important screening limit for general alpha particle activity in water, gross alpha cannot be used to quantitatively address cancer risk from individual radionuclides in a water supply. It is possible that gross alpha can overestimate true risks, leading to unneeded treatment costs, or significantly underestimate risks leading to unanticipated health risks.

Further research of factors that enable Po-210 to get into groundwater and possible treatment-related increases in lead-210 are important public health research areas. MDH plans to analyze additive risks from radionuclides to determine if a group of radionuclides presents a greater health risk even if all levels of each individual radionuclide in the group are below health risk thresholds.

Minnesota's reliance on groundwater increases exposure to radionuclides present in many aquifers. Vigilant surveillance for Po-210 and lead-210 in public water supplies and private wells is important to protect, maintain, and improve the health of all Minnesotans.

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# Tables and Figures

**TABLE 1.** Overall summary of Po-210, lead-210, and gross alpha activity. Mean and median values derived from untreated source water sample locations.

Analyte	Mean (pCi/L)	Median (pCi/L)	Maximum (pCi/L)	Detection % (>0.1 pCi/L Po-210) (>0.5 pCi/L Pb-210)
Polonium-210	0.39	0.13	5.0	67%
Gross Alpha (Day 30)	30	26	88	97%
Lead-210	0.38	0.30	0.70	38%

**TABLE 2.** Analytical results for polonium-210, lead-210, and 30-day post-collection gross alpha activity. Locations listed by highest polonium activity.

Well Unique Number	Aquifer	Po-210, pCi/L (95% confidence interval)	Gross Alpha, 30-day (pCi/L)	Lead-210, pCi/L (95% confidence interval)
00430604	CMTS	4.99 (±0.748)	88	0.551 (±0.307)
00415943	CMTS	1.33 (±0.233)	29	0.326 (±0.180)
00241335	CMTS	1.23 (±0.213)	-- <sup>¥</sup>	0.702 (±0.315)
00151559	CMTS	0.528 (±0.129)	-- <sup>¥</sup>	--
00645355	CMTS	0.371 (±0.087)	41	0.631 (±0.262)
Entry Point E03_RC	*	0.334 (±0.092)	15	2.870 (±0.414)
00206456	CMTS	0.308 (±0.092)	15	0.120 (±0.171)
00625261	CMTS	0.299 (±0.099)	-- <sup>¥</sup>	--
00203614	CMTS	0.297 (±0.099)	22	0.209 (±0.173)
00654758	QBAA	0.279 (±0.091)	<3	0.249 (±0.182)
00227965	CJDN	0.242 (±0.081)	13	0.269 (±0.167)
00603076	PMHN	0.237 (±0.093)	23	--

POLONIUM-210 OCCURRENCE IN MINNESOTA'S AQUIFERS: A PILOT STUDY

Well Unique Number	Aquifer	Po-210, pCi/L (95% confidence interval)	Gross Alpha, 30-day (pCi/L)	Lead-210, pCi/L (95% confidence interval)
Entry Point_E01_H	*	0.232 (±0.079)	44	1.524 (±0.275)
00150359	CMTS	0.211 (±0.084)	31	--
00453792	CMTS	0.190 (±0.084)	37	--
00241352	CTCW	0.142 (±0.068) <sup>#</sup>	25	--
00200664	CJDN	0.139 (±0.069)	-- <sup>¥</sup>	--
00433254	CMSH	0.135 (±0.078)	-- <sup>¥</sup>	--
00201191	CMSH	0.133 (±0.063)	50	--
00559414	CMTS	0.118 (±0.065)	45	--
00538117	PMFL	0.118 (±0.057)	15	--
00207284	CJDN	0.104 (±0.064)	41	--
00213133	OSTP	0.102 (±0.071)	12	--
Entry Point_E02_D	*	0.065 (±0.062)	14	--
00150341	CMTS	0.061 (±0.040)	14	--
00206179	CJDN	0.053 (±0.051)	26	--
00557160	PMHN	0.051 (±0.162)	41	--
Entry Point_E01_IG	*	0.047 (±0.125)	15	--
00217122	DCOM	0.039 (±0.041)	16	--
00217116	DCOG	0.034 (±0.062)	29	--
00546911	CMTS	0.034 (±0.040)	9.1	--
00222588	PMSX	0.030 (±0.067)	20	--
00222547	PMSX	0.027 (±0.049)	39	--

POLONIUM-210 OCCURRENCE IN MINNESOTA'S AQUIFERS: A PILOT STUDY

Well Unique Number	Aquifer	Po-210, pCi/L (95% confidence interval)	Gross Alpha, 30-day (pCi/L)	Lead-210, pCi/L (95% confidence interval)
00433259	CMSH	0.024 (±0.064)	24	--
00207708	CJDN	0.011 (±0.060)	20	--
00617517	CJDN	-0.010 (±0.046) <sup>#</sup>	24	--

\* Entry point sample

--¥ not tested, sample taken by USGS staff, and filtered prior to collection

#pH>2 upon arrival at laboratory. Not included on sample results map, excluded from statistical calculations.



**FIGURE 3.** Po-210 activity of individual source water samples shown grouped according to aquifer. Note broken Y-axis. Aquifer acronyms: CTCW (Tunnel City-Wonewoc), CJDN (Jordan), CMSH (Mt. Simon-Hinckley), CMTS (Mt. Simon), DCOG (Cedar Valley-Galena), DCOM (Cedar Valley-Maquoketa), PMFL (Fond du Lac Formation), PMHN (Mt. Simon-Hinckley), PMSX (Sioux Quartzite), OSTP (St. Peter), QBAA (Quaternary buried artesian aquifer)

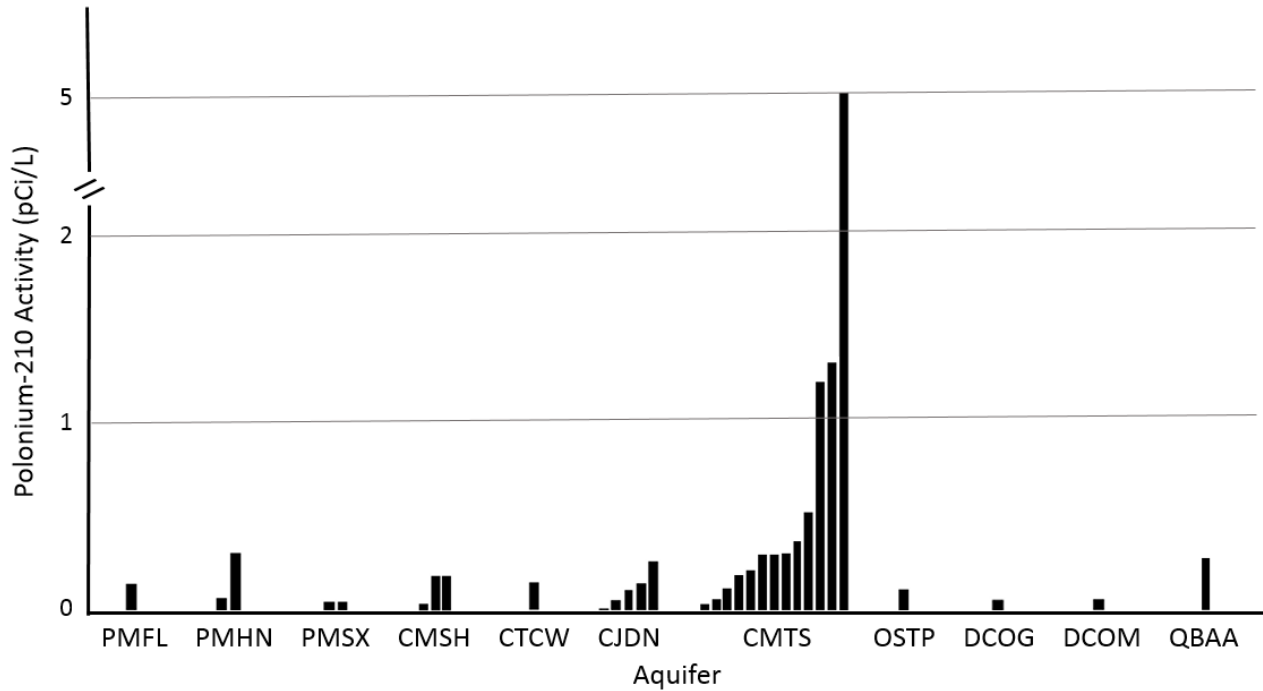


FIGURE 4. Polonium-210 activity in Minnesota.

