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The accumulation of radioactive contaminants in drinking water distribution systems

Darren A. Lytle [a,*], Thomas Sorg [a], Lili Wang [b], Abe Chen [c]

[a] U.S. Environmental Protection Agency, ORD, NRMRL, WSWRD, 26 W. Martin Luther King Dr., Cincinnati,OH 45268, United States

[b] U.S. Environmental Protection Agency, OW, GWDW, SRMD, Washington, DC 20460, United States

[c] ALSA Tech, LLC, Powell, OH 43065, United States

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ABSTRACT

The accumulation of trace contaminants in drinking water distribution system sediment and scales has been documented, raising concerns that the subsequent release of the contaminants back to the water is a potential human exposure pathway. Radioactive contaminants are of concern because of their known health effects and because of their persistence within associated distribution system materials. The objective of this work was to measure the amount of a number of radioactive contaminants (radium, thorium, and uranium isotopes, and gross alpha and beta activity) in distribution solids collected from water systems in four states (Wisconsin, Illinois, Minnesota, and Texas). The water utilities chosen had measurable levels of radium in their source waters. In addition, 19 other elements in the solids were quantified. Water systems provided solids primarily collected during routine fire hydrant flushing. Iron was the dominant element in nearly all of the solids and was followed by calcium, phosphorus, magnesium, manganese, silicon, aluminum and barium in descending order. Gross alpha and beta radiation averaged 255 and 181 pCi/g, and were as high as 1602 and 1169 pCi/g, respectively. Total radium, thorium and uranium averaged 143, 40 and 6.4 pCi/g, respectively. Radium-226 and -228 averaged 74 and 69 pCi/g, and were as high as 250 and 351 pCi/g, respectively.

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1. Introduction

The tendency for some trace contaminants in drinking water to accumulate in the distribution system has been reported (Lytle et al., 2004a,b; Friedman et al., 2010; Valentine and Stearns, 1994; Snoeyink et al., 2002; Schock, 2005; Schock et al., 2008). Lytle et al. (2004a,b) reported on the elemental composition of 13 metals, including iron and arsenic, of 67 internal pipe wall solids and hydrant flush solids collected from several Midwestern water utilities. Measurable levels of all 13 elements were found in all samples with iron being the largest component in all but five samples, where it was the second most abundant. Because arsenic has a strong affinity for iron surfaces, it was found in all the solid samples, with the highest amounts occurring in the hydrant flush solids. The arsenic content of the solids ranged from 10 to 13,650 μg As/g solid (as high as 1.37% by weight). Relatively large amounts of arsenic (>500 μg As/g solid) were even found in solids exposed to relatively low concentrations of arsenic in the water (<10 μg/L). Friedman et al. (2010) reported on the composition

* Corresponding author.
E-mail address: lytle.darren@epa.gov (D.A. Lytle).

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of over 70 pipe sections and hydrant flush solids from 20 water utilities. They focused on regulated inorganic contaminants and radionuclides including antimony, arsenic, barium, chromium, nickel, lead, radium, selenium, thallium, and uranium. Others (Valentine and Stearns, 1994; Snoeyink et al., 2002; Schock, 2005; Schock et al., 2008; Fisher et al., 2000) reported on the accumulation of radium, vanadium, aluminum, and other elements on distribution system materials.

The concern with the accumulation of contaminants in the distribution system is the potential release of contaminants back to the drinking water, which would result in consumers being potentially exposed to elevated contaminant levels. Reiber and Dostal (2000) were the first to document a major release of contaminants into a mid-sized Midwestern distribution system following the introduction of chlorination. Shortly after chlorination began, many complaints of colored water were received, and the follow-on investigations found high levels of iron, copper, and arsenic in the colored water samples. Most surprisingly, the colored water arsenic levels were found to be in mg/L even though the arsenic level in the ground water was below 7 μg/L. Lytle et al. (2010) analyzed water samples collected from a number of home taps of a water system that had approximately 23 μg/L of arsenic at the entry point, but as high as 299 μg/L of arsenic in the distributed water. Arsenic concentrations in the first draw water samples were higher than those in water entering the distribution system in 29 percent of the tap water samples collected, which were attributed to iron solids found in the distribution system.

Contamination of drinking water from naturally occurring radionuclides and anthropogenic sources affects water utilities across many regions of the United States. The U.S. Environmental Protection Agency (EPA) established maximum contaminant levels (MCLs) for radionuclides in water due to an increased cancer risk from the long-term ingestion. Specifically, alpha emitters of 15 pCi/L, beta and photon emitters of 4 mrem/yr, radium (^{226}Ra/^{228}Ra) of 5 pCi/L and uranium of 30 μg/L are established drinking water standards (EPA, 2000), although uranium's MCL was independently determined based on kidney toxicity.

Water treatment technologies that can effectively remove radionuclides from water include lime softening, reverse osmosis, iron removal/coagulation, green sand filtration, and preformed hydrous manganese oxide filtration. The same mechanisms that are responsible for the ability of these technologies to remove radionuclides in the treatment plant may also increase the likelihood that they may accumulate in the distribution system. For instance, calcium, iron, and manganese mineral surfaces are in abundance in many areas of distribution systems.

Few reports exist on radionuclide levels in distribution system solids, and the focus has been limited to only radium (^{226}Ra) and radon gas resulting from its decay or resulting from other decay chains. Valentine and Stearns (1994) reported the ^{226}Ra content of 10 pipe deposits and hydrant flushing solids from three communities in Iowa. Extractable ^{226}Ra levels ranged from 13 to 66 pCi/g (or 56–800 pCi/g, when normalized to dissolvable fractions). Radon release rates from distribution system solids and increased radon levels in actual drinking water distribution systems were also reported. The authors concluded that radium-containing deposits in the distribution system contributed to the increase in radon levels in the distribution system. Fisher et al. (2000) compared ^{226}Ra concentrations in waters entering the distribution system or point-of-entry (POE) and at locations in the distribution system or the point-of-use (POU) in a rural community in Iowa. POE ^{226}Ra levels ranged between 5 and 10.3 pCi/L, and POU ^{226}Ra levels ranged from 0.4 to 12.9 pCi/L. Homes connected to older water mains had significantly higher mean ^{226}Ra levels than those connected to recently installed water mains (8.3 vs. 5.3 pCi/L). Friedman et al. (2010) reported on the ^{226}Ra content of 58 pipe deposits and hydrant flush samples. The median ^{226}Ra concentration was 9.1 pCi/g, and values ranged from 1.1 (10th percentile) to 84 pCi/g (90th percentile). The ^{228}Ra solid content was estimated from the ^{228}Ra aqueous concentration and based on the assumption that it behaved similarly to ^{226}Ra with regard to surface adsorption characteristics. The median ^{228}Ra concentration was estimated to be 4.9 pCi/g, and the total median radium (^{226}Ra and ^{226}Ra combined) concentration was estimated to be 14 pCi/g.

These reports of radium accumulation, taken together with documented cases of accumulation and release of inorganic substances in distribution systems, suggest a need to ascertain the extent to which other radionuclides also accumulate in drinking water distribution systems to better understand potential exposure. The objective of this work was to determine the radioisotope composition of distribution system solids collected from a number of water systems in Illinois, Wisconsin, Minnesota, and Texas that reported measurable levels of radium in their finished waters. Specifically, 25 solids samples (e.g., fire hydrant flushed solids, sediments, pipes, etc.) were collected, and the solids were concentrated, separated, and analyzed. The solids were analyzed for total radium, uranium, and thorium isotopes, gross alpha and beta radioactivity, and a number of major and trace elements including Fe, Mn, Ca, Mg, Ba, P, and others.

2. Experimental

2.1. Study sites

Sampling sites were identified with input from the respective state primacy agencies. The objective was to identify community water systems that had or have measurable levels of radium in their source water, and were interested in knowing more about their system, hence making their assistance likely. The water systems were requested to provide pipes, hydrant flush and water samples, and water treatment history and water quality records.

Utilities were encouraged to send iron-based pipe sections of any reasonable diameter or length, although any pipe material was accepted. The internal surface of corroded metal pipe sections was thought to represent corrosion by-product solids, even though iron hydroxide, calcium carbonate, and other precipitated solids could also be incorporated into the solid matrix or the surface of the corrosion solids.

Hydrant flushed samples were requested from distribution zones that historically had colored water complaints. Hydrant flushed water generally contains loosely bound solids that are susceptible to transport through the distribution system by