

SIR: Uranium and Radon in Groundwater from Glacial and Bedrock Aquifers in the Northern United States: Occurrence, Related Factors, and Implications

Authors: Joe Ayotte, Sarah Flanagan, and William Morrow

Refs to add:

Warner – gapt

Nure – national

I. ABSTRACT

II. INTRODUCTION

A. Radionuclide rule

Uranium in drinking water is an issue of concern ever since the U.S. Environmental Protection set a drinking-water standard of 30 micrograms per liter for this naturally occurring radioactive element (U.S. Environmental Protection Agency., 2000). High levels of radon-222 in certain regions of the country have long been known to be a major water quality issue of concern (U.S. Environmental Protection Agency, 1999).

B. Purpose and scope

The purpose of this study is to identify the occurrence and distribution of uranium and radon-222 gas in groundwater in three aquifer systems: glacial aquifers in the northern United States, Cambrian/Ordovician (CO) aquifers in Minnesota and Michigan, and in crystalline-rock aquifers in New England, New York, and New Jersey (NYNE). Further, this study seeks to explain the variation in uranium concentrations within the three aquifers systems by use of related chemistry data and ancillary data. Ultimately, factors related to U and Rn occurrence and distribution will be discussed for areas of high concentrations.

C. Acknowledgements

Thanks to Terri Arnold of the GAPPT team for compiling the original water-quality datasets. Will add others as the report progresses.

D. Previous investigations

Describe findings from previous studies that characterize occurrence of radionuclides in drinking water and other aquifers; this section will be divided up into 1) national studies, 2) regional or local studies of importance to this report, and 3) geochemistry and controls related to U and Rn in ground water.

National studies of U and Rn in groundwater or that are related to ground water are numerous (Aieta and others, 1987; Alley, 1993; Duval, 1990; Focazio and others, 2001; Hess and others, 1985; Longtin, 1988; Michael and Cothorn, 1986; Michael and Jordana, 1987; Otton, 1992; Smith, 2001a; Smith, 2001b; U.S. Environmental Protection Agency., 2003; Wanty and Nordstrom, 1993; Zapecza and Szabo, 1988). In a study of radioactivity in public water supplies of the United States {Hess, 1985 #36}, the importance of geologic controls on the occurrence of radionuclides.

Local and regional studies may include but not be limited to the following list: (Aieta and others, 1987; Asikainen and Kahlos, 1979; Bruce and McMahon, 1998; Brutsaert and others, 1981; Dennehy and others, 1998; Dixon and Lee, 1989; Fenlon and Moore, 1996; Focazio and others, 2001; Hall, Boudette, and Olszewski, 1987; Hall, Donahue, and Eldridge, 1985; Hess and others, 1985; Kay, 1999; King, Michael, and Moore, 1982; Lindsey and Ator, 1996; Longtin, 1988; Loomis, 1987; Maloney, 1988; Michael and Cothorn, 1986; Michael and Jordana, 1987; Morrissey, 1988; Morrow, 2001; Norton, Hess, and Brutsaert, 1989; Rama and Moore, 1984; Robinson and others, 2004; Schlottmann and Breit, 1992; Seiler, 2004; Stanton and others, 1996; Szabo and dePaul, 1998; Szabo and Zapecza, 1991; Wanty and Gunderson, 1988; Wanty and others, 1992; Wanty, Lawrence, and Gunderson, 1992; Wanty and Nordstrom, 1993; Wood, Kraemer, and Shapiro, 2004; Zapecza and Szabo, 1988; Zelt and Frankforter, 2003).

Describe previous significant findings for the more pertinent publications that support our own findings and conclusions for the high concentrations of U in the glacial west, such as Robert Zielinski's (GD in Central Region) report on the impact of land use and geology on the uranium concentrations in the Arkansas River in Colorado, Zielinski's journal papers on uranium and phosphate fertilizers and their impact on surface-water quality (Zielinski and others, 1997; Zielinski, Asher-Bolinder, and Meier, 1995; Zielinski and Meier, 1988; Zielinski, Simmons, and Orem, 2000).

Describe previous significant findings related to U and Rn in New England to set the stage for support of findings in this study (Brutsaert and others, 1981; Focazio and others, 2001; Hall, Boudette, and Olszewski, 1987; Hall, Donahue, and Eldridge, 1985; Maloney, 1988; Michael and Cothorn, 1986; Michael and Jordana, 1987; Morrissey, 1988; Norton, Hess, and Brutsaert, 1989; Robinson and others, 2004; Szabo and Zapecza, 1991; Wood, Kraemer, and Shapiro, 2004).

III. APPROACH AND METHODS

A. Study design

This study includes aquifers within the glaciated terrain of the United States. The aquifers include glacial and glacio-fluvial aquifers, the Cambrian-Ordovician aquifers of the upper mid west and fractured crystalline bedrock aquifers of the New England-New York region.

1. Description of study area

FIGURE: Map of study area

2. Regional occurrence and distribution

3. Water use of selected aquifers

B. Well selection

1. Shallow monitoring wells (agr and urban land use)

2. Domestic supply wells (variable land use)

C. Sampling and analysis

Water samples were collected and analyzed for major ions, trace elements, chlorofluorocarbons (CFCs), and other constituents by multiple study units, from the period 1992 – 2003 (See Supporting Information), in accordance with NAWQA protocols (Koterba, Wilde, and Lapham, 1995; Lapham, Wilde, and Koterba, 1995). Before collection of any samples, wells were pumped until water temperature, dissolved oxygen, specific conductance, and pH were stable (Koterba, Wilde, and Lapham, 1995). Cleaned fluorinated ethylene polypropylene (FEP) tubing with stainless steel fittings was connected to either a submersible pump or to the water line as close as possible to the wellhead. Samples were collected at a flow rate of 0.05 L/min in a portable sampling chamber. Samples for uranium and other trace elements were collected through a 0.45- μ m disposable polypropylene capsule filter and were acidified with ultra pure nitric acid to a pH of less than 2. The analyses for uranium (and other trace elements)

were made at the USGS National Water Quality Laboratory (NWQL) using ICP/MS methods with laboratory reporting levels (LRL) of 0.2 - 1 µg/L (Faires, 1993; Ivahnenko, Szabo, and Hall, 1996). Analyses of radon-222 were done by liquid scintillation methods (Mullin and Wanty, 1991; Prichard and Gessell, 1977).

E. Data values below the laboratory reporting limit

All data used in this study have a percentage of samples for which the concentration of the analyte is below the LRL. For this reason, each constituent was examined and the lowest common LRL was determined and then all other below LRL data was set to that higher LRL (Helsel and Hirsch, 1992). This facilitates use of all the data in nonparametric statistical analyses (see section on Statistical analyses below) without making assumptions about the distribution of the data below the LRL. The lowest common LRL for U is ____ and for radon-222 is _____. For this data set, ____percent of the U data and ____ percent of the radon-222 data is below the LRL.

F. Ancillary data and explanatory variables

Specific Geographic Information Systems (GIS) and ancillary data were compiled for the analysis of Uranium and Radon-222 occurrence. Many of these data sets were generated by the USGS's National Water Quality Assessment Program (NAWQA) These include (1) aquifer type—bedrock or glacial unconsolidated, aquifer texture, geographic location and source material, well depth, and well type; (2) population statistics (U.S. Bureau of the Census, 2000); (3) National Land Cover Data set (U.S. Geological Survey, 2000); (4) Soil Survey Geographic (STATSGO) database (U.S. Dept. of Agriculture Natural Resources Conservation Service, 1994; Wolock, 1997); (5) lithology and lithochemical character of near surface bedrock in New England (Robinson, 1997; Robinson and Kapo, 2003); (6) National Uranium Resource Evaluation Stream Sediment Chemistry data (Smith, 2001a; Smith, 2001b); (7) proximity to mapped lineament data (Moore and others, 2002); (8) index of recharge to ground water and hydrologic landscape models (Wolock, 2003a; Wolock, 2003b); and (9) water-sample temperature, pH, specific conductance, and dissolved oxygen data.

G. Statistical analyses

Nonparametric group comparison tests were used to examine the relations of U and Rn to other chemical environmental data because the data are skewed—that is, a large proportion of the samples have either U, Rn, or other chemical data concentrations that are reported as “less than the LRL.” For this study, all statistical analyses included the data reported as below the LRL, and the significance level (α) was 0.05.

Contingency table analysis (Pearson's Chi Square test for independence) were used to measure the association U and Rn (presence or absence) with

a nominal grouping variable of two or more categories (Helsel and Hirsch, 1992). Group comparison tests such as the Wilcoxon and the Kruskal-Wallis tests were used to determine if the means of the ranks of the data from two or more groups were significantly different (Helsel and Hirsch, 1992; SAS Institute, 1990). Where multiple comparison tests showed independent populations exist, the Tukey test was used on the ranks of the data to identify group means that were significantly different (Helsel and Hirsch, 1992; SAS Institute, 1990). Although other methods exist for estimating the distribution of missing data and comparing groups, such as substitution and distributional methods, nonparametric methods offer several benefits including 1) no assumption of normally distributed data; 2) better statistical power; and 3) data below the LRL are used without fabrication, resulting in an accurate portrayal of the “less-than the LRL” information (Helsel, 1990; Helsel and Cohn, 1988; Helsel and Hirsch, 1992).

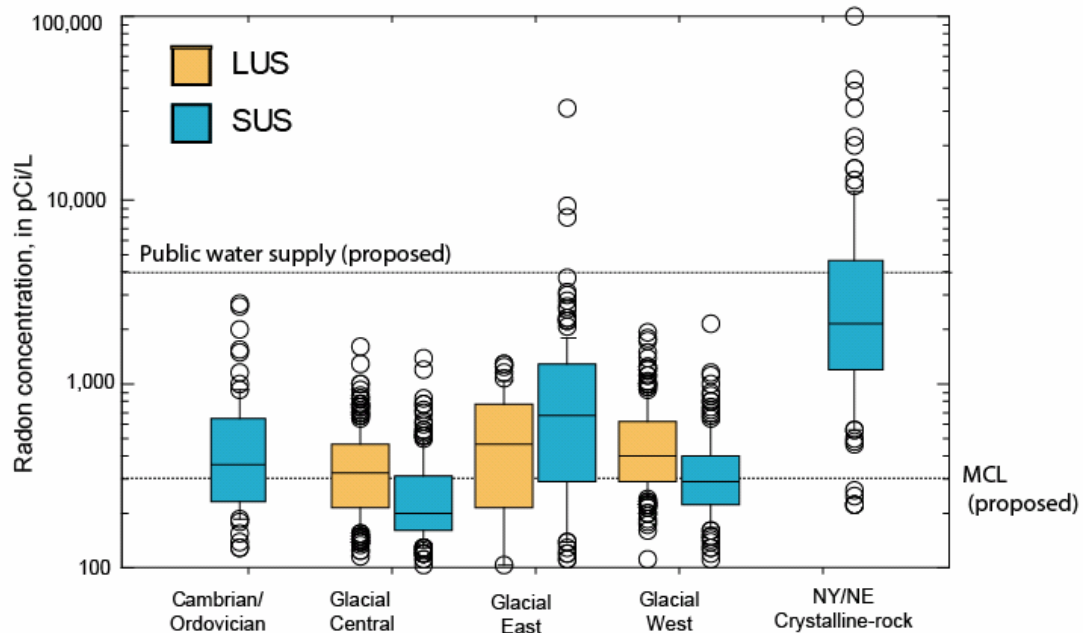
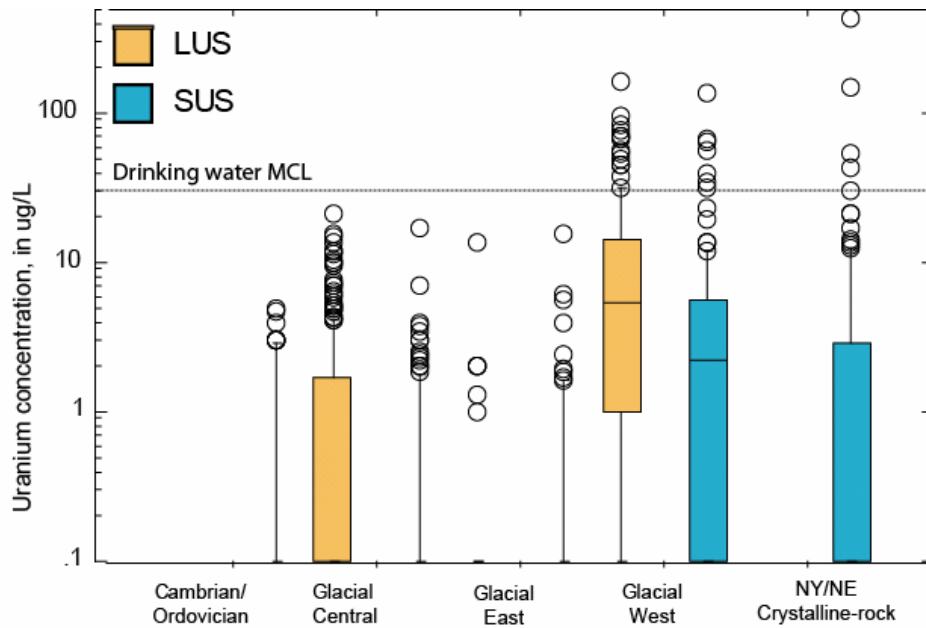
For analysis of environmental and other factors related to U and Rn concentrations, Spearman correlation coefficients were computed to measure the strength of the relation to explanatory variables (Helsel and Hirsch, 1992).

H. QA/QC data

Quality-control (QC) samples, including replicates and field blanks, were prepared according to NAWQA protocols (Koterba, Wilde, and Lapham, 1995). ____ blanks (____ percent) and ____ replicates (____ percent) were collected for trace elements for the ____ wells sampled. ____ replicates for radon were collected for the ____ wells sampled. Analysis of all blank samples showed no inherent bias (we presume) in the method of analysis for the trace elements and major ions of interest. Replicate samples were collected immediately after the routine samples in the field using the same collection methods and equipment. All differences measured in concentrations between replicate pairs were well within the precision of the method for all trace elements and for radon-222 (again, we presume). Regardless, we may report the percent difference in the replicates for radon and for U, perhaps by study type.

IV. RESULTS

A. Occurrence and distr. section by principal aquifer type (PA), aquifer source material, well (network) type



Distribution of uranium and radon in ground water by principal aquifer and well network in the GAPT area.

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1. Highest U in study area in glacial west & New England
 2. Glacial (East, Central, West), Crystalline (NENY, CO)
- B. Factors related to uranium and radon by aquifer type – correlation, xy, etc.
1. New England NY crystalline
 - U, Rn by rock granites and metasediments

FIGURE: Boxplots by rock type, other things

TABLE: Correlation coefficients of highest and most probable
Correlation of U, Rn w/ explan. vars

Relevant x-y plots

Rn and U

Rn and Pb

FIGURE: radon v U and radon v Pb; others?

2. Glacial west

U, Rn by survey or crop or irrig. Properties...

FIGURE: Boxplots by ag type, other things

TABLE: Correlation coefficients of highest and most probable
Correlation of U, Rn w/ explan. Vars

Relevant x-y plots

U by SC, ions

U by ag factors

FIGURE: U v SC and U v. B,K,Ca,Na, SO₄; others?

V. DISCUSSION

- A. Relate occurrence findings to new regulations based on aquifer/water use
- B. Mechanisms for mobility, association with Pb
- C. Mechanisms for mobility, concentrating U
- D. Relate explanatory factors from anomalous areas rest of study area, if appropriate; discuss implications

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