USGS
science for a changing world

# A model for radionuclides in glacial and bedrock aquifers of the United States

**Joseph D. Ayotte[1], Sarah M. Flanagan[1], Laura Hayes[1], and William S. Morrow[2]** – U.S. Geological Survey, [1]New Hampshire – Vermont Water Science Center, [2]Illinois Water Science Center

## Introduction

Uranium and $^{222}$radon are naturally occurring radionuclides found in ground water throughout the United States and can be associated with adverse health outcomes. This study uses a geologic model to explain the variation in uranium and $^{222}$radon concentrations in three of the principal aquifers identified by the USGS's National Water Quality Assessment Program (NAWQA) **(Figure 1).**

Data from over 1,400 domestic, public-supply, and monitoring wells sampled by the NAWQA program from 1991-2003 were analyzed. Because of the size and complexity of the Glacial aquifers, we developed a model to differentiate sub aquifers based on source materials, glacial lobe delineation, and directions of Wisconsinan ice flow from Canada.

**Figure 1. Locations of NAWQA Regional Assessments in Principal Aquifers**



## Uranium and $^{222}$radon in principal aquifers

Radionuclide concentrations differ by Principal Aquifer **(Table 1)**. The median uranium concentration in the three Principal Aquifers in this study are below 1 microgram per liter (µg/L), but the 75th and 90th percentiles vary. Viewed this way, uranium is highest in New England Crystalline aquifers (Table 1) and lowest in the Cambrian – Ordovician aquifer. The median $^{222}$radon concentration is near the USEPA proposed MCL of 300 Pico curies per liter (pCi/L) in the glacial and Cambrian – Ordovician aquifers but is 7 times higher (2,144 pCi/L) in New England Crystalline aquifers where 25% of wells sampled exceed the proposed USEPA alternative MCL (AMCL) of 4,000 pCi/L **(Table 1)**.

**Table 1. Summary of uranium and $^{222}$radon by Principal Aquifer in the glaciated northern United States**

| Principal Aquifer | N | Maximum | 90th | 75th | Median | 25th | 10th | Minimum |
|---|---|---|---|---|---|---|---|---|
| **Uranium, µg/L** | | | | | | | | |
| New England Crystalline | 113 | 428.9 | 12.8 | 2.9 | < 1 | <1 | < 1 | < 1 |
| Glacial | 874 | 162.3 | 7.6 | 2.1 | < 1 | <1 | < 1 | < 1 |
| Cambrian – Ordovician | 73 | 5 | 3 | 1 | < 1 | <1 | < 1 | < 1 |
| **$^{222}$Radon, pCi/L** | | | | | | | | |
| New England Crystalline | 107 | 215,210 | 12,000 | 4,899 | 2,144 | 1,200 | 573 | < 80 |
| Glacial | 609 | 8,056 | 888 | 523 | 313 | 208 | 150 | < 80 |
| Cambrian – Ordovician | 70 | 2,700 | 920 | 616 | 370 | 230 | 181 | < 80 |

## Sources of uranium and $^{222}$radon

Variation of uranium and $^{222}$radon in ground water depends in part on the source materials of the aquifers. The source of glacial materials depends on factors that include 1) the direction of glacial ice lobe movement; 2) existence of older glacial deposits; and 3) the underlying bedrock geology. Ice that flowed into the upper Midwest, United States from Canada came from various directions and carried materials from along its flow path. This material is a mix of existing glacial deposits (which may have originated elsewhere) and materials from the local geology, creating a composite mixture that can affect contaminant concentrations, such as uranium and $^{222}$radon.

## Incorporating a geologic model




**Figure 2. Geology of the upper Midwest, United States**

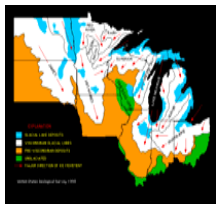


**Figure 3. Wisconsinan glacial lobes and ice directions**

To analyze regional variation of radionuclides in ground water in the glaciated U.S., the glacial principal aquifer was divided into sub-aquifers. Different classifications of the glacial aquifers are available (Meinzer, 1923; Heath, 1984; USGS, 2003) but are mostly based on age and stratigraphy. Other studies have used generalized geologic regions or political boundaries (Hess and others, 1985; Michael, 1987; Focazio and others, 2001; Longtin, 1988).

Maps of generalized bedrock geology **(Fig. 2)**, Wisconsinan glacial lobes **(Fig. 3)**, their advance directions, and their composition, were used to delineate important sub-aquifers within the glaciated terrain of the U.S. Analysis of the bedrock lithology and the glacial lobe data identified groups that might relate to radionuclide occurrence in ground water (King and Beikman, 1974; Schumann, 1993).

## Incorporating terrestrial gamma surveys

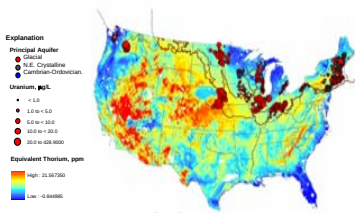



**Figure 4. Equivalent Thorium (ppm) and concentrations of uranium in wells**

In addition to the glacial and bedrock geology, national data on the equivalent uranium (eU) and thorium (eTh) content of near-surface sediments from terrestrial gamma surveys (Duval, 1990) was used to help define areas of potential high concentrations of radionuclides in groundwater **(Fig. 4).** The 75th percentiles of uranium and $^{222}$radon concentrations in ground water for each NAWQA study network are plotted against the associated 75th percentile of eU and eTh from the gamma survey. Above 2.0 ppm eU and 6 ppm eTh, ground water $^{222}$radon concentrations generally increase **(Fig. 5a, b).** Above 5 ppm eTh, uranium concentrations increase but are highly variable **(Fig 5d).**

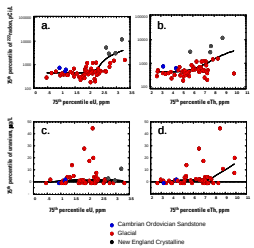



**Figure 5a-d. Relation of the 75th percentile concentration of ground water (a, b) $^{222}$radon and (c, d) uranium to the 75th percentile eU and eTh in near-surface sediments.**

## A framework for regional analyses

The Principal Aquifer and sub-aquifers are shown on the maps below (Fig. **6a-b**). The maps also show locations of wells used in this study and concentrations of uranium **(Fig 6a)** and $^{222}$radon **(Fig 6b)**.



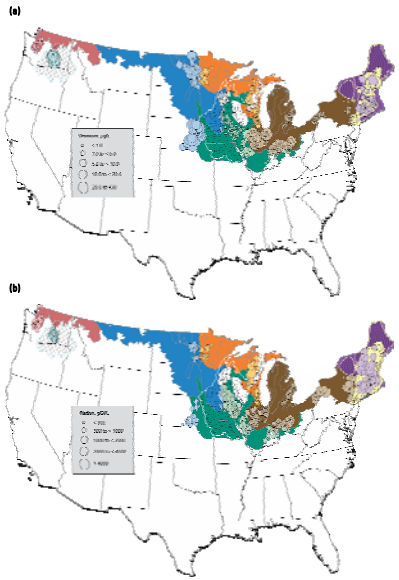



**Figure 6a-b. Principal Aquifer and sub-aquifer groups with (a) uranium and (b) $^{222}$radon in ground water. New England Crystalline underlies Northeastern Glacial aquifer. The Cambrian – Ordovician Sandstone aquifer underlies the Midwest Glacial aquifer. The Columbia Plateau Glacial aquifers are outside the extent of Pleistocene glaciation.**

## Source-water guideline exceedences

**Table 2. Percent of drinking water and monitoring wells with uranium and $^{222}$radon greater than the USEPA Maximum Contaminant Level (MCL or AMCL), by aquifer category and well type. --, no data.**

| Principal Aquifer or sub aquifer | Drinking water wells: Uranium N | Uranium Percent > 30 µg/L | $^{222}$radon N | $^{222}$radon Percent > 4,000 pCi/L | $^{222}$radon Percent > 300 pCi/L | Monitoring wells: Uranium N | Uranium Percent > 30 µg/L | $^{222}$radon N | $^{222}$radon Percent > 4,000 pCi/L | $^{222}$radon Percent > 300 pCi/L |
|---|---|---|---|---|---|---|---|---|---|---|
| PMG | 31 | 0 | 57 | 0 | 46 | 5 | 0 | 17 | 0 | 65 |
| CPG | -- | -- | 44 | 0 | 89 | 46 | 9 | 39 | 0 | 72 |
| WCG | 52 | 12 | 35 | 0 | 40 | 116 | 11 | 41 | 0 | 73 |
| NCG | 76 | 0 | 74 | 0 | 34 | 93 | 0 | 31 | 0 | 32 |
| ECG | 70 | 0 | 123 | 0 | 45 | 209 | 0 | 209 | 0 | 53 |
| PWG | 59 | 0 | 88 | 0 | 41 | 8 | 0 | 83 | 0 | 66 |
| COS | 73 | 0 | 74 | 0 | 66 | -- | -- | -- | -- | -- |
| NEG | 67 | 0 | 61 | 2 | 92 | 55 | 0 | 29 | 0 | 93 |
| NEC | 113 | 4 | 107 | 30 | 94 | -- | -- | -- | -- | -- |

## Correlations with uranium and $^{222}$radon

Spearman correlation coefficients (rho) between uranium and other water chemistry data vary by aquifer and sub aquifer. Analysis of correlations may help identify processes affecting concentrations of uranium in ground water **(Table 3)**.

**Table 3. Spearman correlation coefficients for Uranium by principal aquifer and sub aquifer; --, no data; bold p < 0.05.**

| Aquifer | ANC | B | Ca | HCO3 | Li | Mn | Ni | Se | SO4 | Sr | TDS | $^{222}$Rn | alpha |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| PMG | -- | -- | -- | -- | -- | -- | -- | -- | -- | -- | -- | -- | -- |
| CPG | **0.88** | **0.42** | **0.31** | **0.77** | **0.47** | 0.22 | **0.31** | **0.37** | **0.51** | **0.52** | **0.88** | 0.18 | -- |
| WCG | **0.40** | **0.36** | **0.37** | **0.37** | **0.43** | **-0.31** | -0.01 | **0.60** | **0.27** | **0.32** | **0.36** | **0.06** | -- |
| NCG | 0.18 | 0.04 | **0.19** | **0.24** | **0.20** | **0.18** | **0.42** | **0.19** | **0.29** | 0.14 | **0.17** | 0.10 | 0.06 |
| ECG | **0.35** | **0.18** | **0.40** | 0.29 | 0.11 | **0.48** | **0.43** | **0.16** | **0.41** | **0.23** | **0.36** | -0.19 | -- |
| PWG | -0.16 | -0.45 | **0.47** | -- | -- | -0.04 | **0.45** | 0.13 | **0.58** | **0.61** | **0.29** | **0.41** | -- |
| NEG | **0.31** | -0.03 | **0.29** | **0.30** | 0.16 | -0.01 | -0.01 | -- | **0.37** | **0.22** | **0.19** | 0.01 | **0.54** |
| COS | **0.48** | -0.14 | **0.42** | -- | -0.22 | **0.36** | **0.47** | 0.06 | **0.32** | 0.02 | **0.27** | 0.09 | -- |
| NEC | 0.18 | -0.05 | **0.25** | -0.03 | 0.24 | -0.18 | -0.01 | -0.13 | -0.03 | 0.10 | 0.14 | **0.60** | **0.72** |

### New England Crystalline aquifers

In New England (NEC), uranium correlates with gross alpha (0.72) and $^{222}$radon (0.60) **(Fig 7a-c)**. This unusual result was also reported in Finland (Vesterbacka, 2005), in similar bedrock wells, where $^{222}$radon correlated with $^{238}$uranium (0.57). In addition, $^{222}$radon is weakly correlated with total lead (0.38, p =0.0005) in the NEC aquifer **(Fig 7d)**. $^{222}$radon and $^{210}$lead were also correlated (0.74) in the Finnish study. Where $^{222}$radon in ground water is high it is possible that some lead associated with $^{222}$radon decay remains dissolved (Focazio, 2001 ).

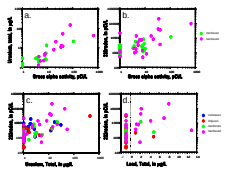



**Figure 7a-d. Gross alpha is related to (a) uranium and (b) $^{222}$radon. $^{222}$ radon is related to (c) uranium and (d) lead in New England Crystalline aquifers.**

### Columbia Plateau Glacial aquifers

The Columbia Plateau Glacial aquifers (CPG) are affected by agricultural irrigation (Embrey, 1988). TDS concentrations exceeding 500 mg/L **(Fig. 8d)** indicate irrigation effects (Whitehead, 2000).
Uranium correlates with bicarbonate (0.77), field specific conductance (0.71), sodium (0.70), TDS (0.68), acid neutralizing capacity (0.68), sulfate (0.51), and pH (-0.48) **(Fig. 8a-c)**. These suggest that complexation with bicarbonate and sulfate ions (Hem, 1985 ) is a control on uranium solubility.

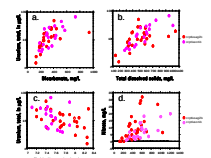



**Figure 8a-d. Uranium is related to to (a) bicarbonate, (b) TDS, and (c) pH; Nitrate is related to TDS (d) in the Columbia Plateau Glacial aquifer.**

## Summary and Conclusions

Uranium and $^{222}$radon concentrations vary significantly but are more predictable when analyzed by a sub-aquifer classification of the glacial deposits that emphasizes source materials of the sediment. Uranium concentrations exceed the USEPA MCL of 30 µg/L in source water to drinking water wells in the West-central Glacial aquifer, the bedrock wells of the New England Crystalline aquifers, and in monitoring wells in the West-central and Columbia Plateau Glacial aquifers. All sub-aquifers have groundwater with $^{222}$radon concentrations above the proposed MCL of 300 pCi/L. Drinking water wells in the New England Crystalline and Northeast glacial aquifers exceed the proposed AMCL of 4,000 pCi/L. Classifying the glacial aquifers with information on geology, glacial lobe direction, sources of sediment, and terrestrial gamma data, greatly improves the ability to explain regional radionuclide occurrence.

**Contact information:** Joseph Ayotte, USGS, 361 Commerce Way, Pembroke, NH 03275 jayotte@usgs.gov, 603.226.7810 or Sarah M. Flanagan, 603.226.7811, sflanaga@usgs.gov

May, 2006