Division of Science, Research and Technology

Research Project Summary

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Northern New Jersey Radionuclide Investigation: Determination of Uranium, Radium and Radon in Ground Water with High Gross Alpha-Particle Activity, and Implications for Future Monitoring Efforts

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Abstract

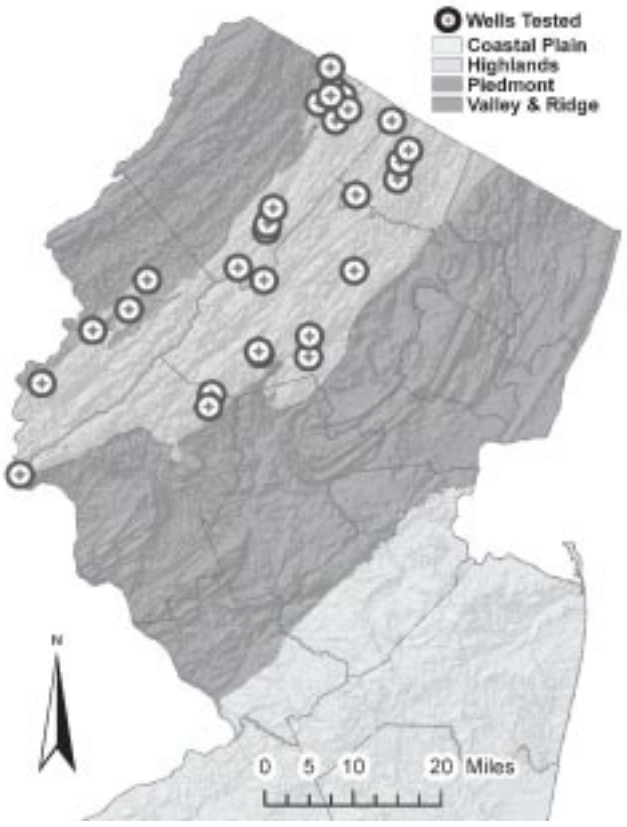
While a great deal is known about radioactivity in southern New Jersey ground water, the amounts of radioactivity in the ground water in Northern New Jersey ground water are not as well defined. The purpose of this study was to evaluate the presence of gross alpha, radium, uranium, and radon in ground water in the Highlands Physiographic Province. A total of 31 wells were targeted for sampling. The wells selected were known to have high levels of gross alpha-particle activity or were near wells that had previously been shown to have radiological problems.

The ground water samples from the targeted wells were collected between June, 2007 and November, 2007. The results of the analysis can be summarized as follows:

- Gross Alpha Particle Activity: Water from 11 of 31 wells exceeded the (uncorrected) gross alpha standard of 15 pCi/L. (Because the presence of Uranium affects the gross alpha-particle activity measurement, corrections need to be applied to evaluate the gross alpha compliance with the 15 pCi/L MCL.)
- Radium: Water from 1 well exceeded the combined Radium 226/228 MCL of 5 pCi/l.
- Uranium: Samples from 7 of the 31 wells exceeded the Uranium MCL of 30 ug/l, with 4 samples exceeding 60 ug/l.
- Radon 222: Samples from 12 of the 31 wells had levels of Radon-222 ranging from 10,000 to 40,000 pc/l Radon-222 and 20 of 31 had > 4,000 pCi/L (the proposed USEPA Alternative MCL).

These results indicate that elevated concentrations of radionuclides are a potential health issue for residents using ground water in the Highland Physiographic Province. The data presented here will be subsequently evaluated to determine the vulnerability of wells in the Highlands region to contamination by uranium or gross alpha-particle activity. This evaluation should allow a ranking of wells into low, medium, and high vulnerability categories based on statistical relations (NJDEP, 2004a).

Figure 1: Location of wells sampled in this study



Introduction

Radioactivity in Southern New Jersey Coastal Plain ground water has been studied extensively (Szabo et al., 2005). The situation in Northern New Jersey ground water, like much of the Appalachian region in the eastern United States (Szabo et al., 2001), is not as well defined. It is known that certain geological units in Northern New Jersey contain radioactive elements such as uranium (U), and radium (Ra), and some ground water samples have been collected with high gross alpha-particle activity (Serfes, 2004). The high activity is presumably due to the presence of Ra or U, both of which are regulated in public drinking water (USEPA 2000a). Furthermore, radon (Rn) is considered a major issue in northern New Jersey, and occurrence data would be helpful to NJDEP's efforts to encourage testing and remediation of Rn in private homes [http://www.nj.gov/dep/rpp/radon/index.htm].

The purpose of this study was to analyze ground water samples for concentrations of U, Ra, and Rn in areas where, previously, high gross alpha-particle activities have been found. Specifically, the Highlands Physiographic Province of northern New Jersey was chosen for the initial occurrence assessment. This assessment is the first detailed study of radiological occurrence in the Highlands. The occurrence data collected from this study will be evaluated using additional statistical and mathematical assessment, which might help in identifying geological and water quality parameters associated with the presence of U, Ra, and Rn in ground water. Additionally, it is important to understand the relation of gross alpha-particle activity with U, Ra, and Rn, since gross alpha-particle activity is the first test result on radioactivity that a homeowner, a school principal, or a water-utility manager would receive if the water were tested for radioactivity.

Remediation options are widely different for U and Ra. Thus, in northern New Jersey where high gross alpha-particle activity is present, it is critical to determine what portion of the alpha activity is from U and from Ra to assess treatment options. There is sparse information concerning the effectiveness of existing treatment units utilized in northern New Jersey for removing the radionuclides present in the water. The initial survey of U occurrence could provide future guidance to homeowners on the topic of treatment options that are most effective. Additionally, the ratios of U to gross alpha-particle activity and other water-quality indicators could be used to develop guidance as to whether residents/utilities could complete the less expensive U mass analysis (in ug/L) at most sites, or whether they need the more expensive isotopic radioactivity analysis (in pCi/L) (Osmond and Cowart, 1976), and Ra analysis. High laboratory costs and equipment costs in the future may be avoided if the occurrence data can be collected and used for guidance in recommending the most suitable laboratory analyses and treatment options.

Methods

This study involved sampling 31 wells (see Figure 1) located in the Highlands Physiographic Province, a region with complex geology (Drake and others, 1996). The selected wells approached or exceeded the gross alpha standard of 15 piC/L, or were located in the same neighborhood as a previously sampled well with radiological concern. Both private wells and non community wells (with a focus on schools) were included. Effort was made to sample wells with water that had a wide range of pH and dissolved oxygen, two factors considered critical for understanding radionuclide chemistry and occurrence. The wells were analyzed for gross alpha, Ra 226/228, and U mass. Field parameters such as pH, dissolved oxygen (DO), specific conductance (SC), and turbidity were also determined. Rn was determined in the field using a field research (alpha-spectrometry) method. The samples with the highest Rn concentrations were also checked in the laboratory by the USEPA approved liquid-scintillation method. To assist in evaluating treatment options, both raw (untreated) and finished (treated) samples were collected at the 7 sites where treatment was already installed.

The study consisted of two phases completed cooperatively between US Geological Survey (USGS) and the NJ Department of Environmental Protection (NJDEP):

1) existing data were reviewed to identify wells with high gross-alpha activities, and
2) wells were sampled and analyzed for the suite of radioactive parameters described above (and for ancillary parameters detailed below).

This approach provides environmental radionuclide occurrence information for ground water from the Highlands Physiographic Province in northern New Jersey. A follow-up assessment phase will further evaluate the occurrence information, and will be used to predict the vulnerability of water from wells to contamination by natural U. Possible regional sampling and analytical recommendations for northern New Jersey can be developed as part of a follow-up assessment phase that would also identify additional data gaps.

Samples were collected following the modified version of the USGS low-level trace-element sampling protocol as applied to ground water sampling by Ivahnenko and others (1996). Samples were filtered using a 0.45-micron disposable polysulfone capsule filter within a portable glove-bag chamber (Horowitz and others, 1994). Samples for analysis of radionuclides, selected trace elements, silica, and major cations were preserved within a separate portable glove chamber onsite immediately upon collection with laboratory-grade nitric acid. Monitoring of field-determined properties pH, dissolved-oxygen (DO) concentration, specific conductance, water turbidity, and water temperature were completed in a flow-through cell after instrument calibration (Wilde and Radtke 1999). One field-equipment blank was collected before sampling was initiated. About 10 field replicate samples were collected for radionuclides.

Table A-1. Analytical technique and method reporting level of chemical analyses performed for the northern New Jersey U occurrence study, 2007.

Constituent	Laboratory Reporting Level (LRL)	Method
Gross alpha, short term (48-hr)	3 pCi/L	Ba-sulfate co-precipitation & Low-background proportional count
Ra-226 Ra-228 Ra-224	0.3 pCi/L 0.5 pCi/L 0.5 pCi/L	Ba-sulfate co-precipitation & Gamma Spectroscopy
Uranium	0.3 ug/L	ICP-Mass Spectrometry
Manganese	0.2 ug/L	
Iron	10 ug/L	ICP - optical spectroscopy
Arsenic	0.1-0.3 ug/L	ICP-collision Mass Spectrometry
Sulfate, Chloride, Nitrate	0.3 mg/L	Ion Chromatography
Uranium isotopes	0.1 pCi/L	Alpha Spectrometry or ICP-MS
Arsenic species	0.1-0.3 ug/L	Field extraction; ICP-Mass Spectrometry
Total *coliform* bacteria Fecal *coliform* bacteria	1 CFU/ 100mL	Most Probable Number with defined substrate (Colilert)
FIELD MEASUREMENTS		
Temperature	0.1 ˚C	Field Meter (YSI or Hach, Calibrated)
pH	0.1 units	
Dissolved oxygen	0.1 mg/L	
Conductance	5 uS/cm	
Turbidity	0.1 NTU	
Alkalinity	0.1 mg/L	Titration
Rn-222	120 pCi/L 50 pCi/L	Alpha Spectrometry or Liquid Scintillation

Analytical techniques and method reporting levels for chemical constituents performed for the 31 ground water samples from the Highlands Province of northern New Jersey to determine the radionuclide occurrence are summarized in Table A-1.

Results

Radionuclide (gross alpha-particle activity and U, Ra-224, Ra-226, Ra-228, and Rn-222) and inorganic constituent concentrations are available for each well site. Where treatment systems were available, the data are denoted by sample type (raw, treated). Sampled well locations are illustrated on a map of northern New Jersey (Figure 1). The data files containing the water-quality characteristics and results linked with site identifiers will be published online in the USGS Annual Data Report [nj.usgs.gov/adr/WDR-NJ-06-3/index.html] (updated every spring), which allows all the data to be readily accessed by the public and USGS cooperators.

Discussions and Conclusions

A summary of the salient findings regarding radionuclide occurrence in the Highlands region of northern New Jersey include:

1. Water from 11 of 31 wells exceeded the (uncorrected) gross alpha of 15 pCi/L, since U does affect the gross alpha-particle activity, measurement corrections need to be applied to evaluate the gross alpha compliance with the 15 pCi/L MCL

2. Water from 1 well exceeded combined Ra MCL (5 pCi/L) and 1 well was near the MCL (3.3 pCi/L)

3. Water from 4 of 31 wells had > 60 ug/L U, 7 exceeded the MCL of 30 ug/L, and 11 were > 15 ug/L

4. Water from 12 of 31 wells had > 10,000 pCi/L Rn-222, and 20 of 31 had > 4,000 pCi/L (the proposed USEPA alternate MCL). Only 1 water sample of 31 had Rn-222 < 300 pCi/L, the proposed USEPA MCL.

5. In addition, two wells exceeded the NJ Arsenic MCL of 5 ug/L with values of 14.9 ug/L and 7.1 ug/L (NJDEP, 2002b); four wells had detectable levels of total coliform, with one well testing positive for fecal coliform.

6. At seven of the 31 sampled wells, there was some type of water treatment, and both pre- and post-treatment samples were collected. Six of the wells had a water softener (cation exchange resin), which removes Ra from water. One sample exceeded the Ra 226/228 MCL of 5 pCi/L, and the water softener reduced the level below the MCL. Two wells had anion exchange treatment to remove U, and both systems reduced the U concentration below the MCL. Only one system had treatment for Rn, and a treated sample could not be collected for Rn analysis at that well. Treatment is an issue that needs to be evaluated separately for each of the different radionuclides (U, Ra, Rn).

Recommendations

1. A statistical analysis of the data generated by this study should be carried out to determine the relationship between water quality parameters, field measured properties, gross alpha-particle activity, U, Ra, U isotope ratios, and various hydrogeologic variables.

2. This data should be utilized in updating and improving radon maps, and recommendations should be made about collecting radon samples in light of possible upcoming Rn regulations.

3. This study focused on the Highlands Physiographic Province. Additional sampling in northern New Jersey's other physiographic provinces where high gross alpha-particle activity has been detected, particularly the Piedmont (Newark Basin) and to a lesser extent the Valley & Ridge should be planned to clarify the occurrence of radionuclides in these regions where "Spatial" data gaps are found.

4. There is a data gap concerning the contribution of Rn from water into the indoor air of houses with existing radon-in-air remediation. This gap appears important because of the substantial prevalence of high-Rn well waters. The indoor air is already being treated for Rn entering from soil gas, but excessive exposure might occur in the living space from Rn degassing from water.

The magnitude of this increase has not been evaluated, nor is it known how long it takes after water use for the Rn levels to return to their previous levels.

5. The Private Well Testing Program should consider adding a gross alpha testing requirement in the counties in Northern New Jersey.

In summary, from these data it appears that the occurrence of radionuclides is an important issue for ground water in the Highland Physiographic Province, Further evaluation should consider techniques to determine the vulnerability of wells in the Highlands region to contamination by U, gross alpha-particle activity, and radon.

Appendix A. Analytical Methods - Detailed Description

The gross alpha-particle activities were analyzed by residue procedure in nitric acid matrix (residue on evaporation is collected on a stainless steel planchet) with low-background alpha counting in a low-background gas-flow proportional alpha-beta detector (Krieger and Whittaker, 1980; American Society Testing Materials, 1999) no more than 48-72 hours after sample collection using the Th-230 calibration curve (Parsa, 1998; USEPA, 2000b; NJDEP, 2002a; Szabo and others, 2005). The LRL (Laboratory Reporting Level) for gross alpha-particle is 3 pCi/L, although individual results can be occasionally lower than the LRL. The LRL is higher than that for the U isotopes because of the absence of sample purification steps and because the analytical equipment used is not as sensitive.

The field-extraction technique of Garbarino and others (2002) was used for determination of the concentration and speciation of As using improved chromatographic separation completed in the field thereby removing the concern of specie stability during storage. The As sample is extracted in the field in order to avoid the problems noted by Edwards and others (1996) and Hall and others (1999). The chemical compound EDTA is added to the sample for preservation of As(III) specie. The As(V) extracted from the water onto a resin cartridge in the field must be re-extracted from the resin in the laboratory. Typically dilute sulfuric or nitric acid is used. The extracts were analyzed by collision cell ICP-MS for As(V), and the liquid phase of the sample is analyzed for As(III).

Trace inorganic constituent concentrations were determined using inductively coupled plasma – mass spectrometry (ICP-MS) (Faires, 1993) or inductively coupled plasma – optical emission spectrometry (ICP-OES), and anions by chromatography (Fishman, 1993).

A portable RAD-7 (Durridge Co., Bedford, MA) alpha-spectrometer radon-in-air monitor was used to determine the Rn-222 concentrations in the water. For detection with the RAD-7, the Rn-222 was stripped from water and was circulated via air in the alpha-spectrometer counting cell. A small portion of the Rn-222 decays to Po-218, which is electrostatically deposited onto the semiconductor-based alpha-particle detector. The detector specifically records the decay of the Po-218, which is easy to detect because it occurs at high energy (6.00 MeV; Kim and others, 2001). A correction is made for the Bi-212 progeny, which has similar alpha energy. Because the Po-218 only has a 3-minute half life, it too rapidly decays, allowing both detection of the Rn-222 and maintaining a low alpha background (avoiding high bias) because of the rapid decay rate. (When concentrations exceed 10, 000 pCi/L, however, some high bias is possible because of interference in counting all the progenies). The concentration of Rn-222 was also determined both by the traditional liquid scintillation technique (USEPA, 1991), mostly for those samples with results over 10,000 pCi/L.

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