HYDROGEOCHEMICAL CONTROLS ON U IN A SANDSTONE AQUIFER

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ABSTRACT
Elevated U in Jacobsville sandstone aquifers in Michigan's Upper Peninsula was characterized. Approximately 26% of the 271 wells tested had U above 30 µg/L, generally occurring in clusters. The hydrogeochemical controls on groundwater U concentrations were characterized using water sampling and geophysical logging. Enrichment of U in the sandstone may make wells in its oxidized aquifers at risk for U concentrations above maximum contaminant levels. Wells were logged with a gamma-ray spectrometer to determine whether groundwater U anomalies could be explained by the heterogeneous distribution of U in the sandstone. While some wells showed a correlation between aqueous U concentrations and the U concentration in the rock, other wells did not, indicating that other factors are also important. $^{235}U/^{238}U$ isotope ratios were consistent with naturally occurring U. $^{234}U/^{238}U$ ratios showed characteristic fingerprints for groundwater from different aquifer conditions. Groundwater age dating may help explain variations in well water U concentrations.

RÉSUMÉ
L'U élevé dans Jacobsville sandstone aquifers dans le Peninsule Superieure de Michigan a été caractérisée. Approximativement 26% des 271 puits essayés avait l'U au-dessus de 30 µg/L, généralement dans les groupes. Le hydrogeochemical contrôle sur les concentrations d'U d'eau souterraine ont été caractérisé utilisant de l'eau mesure et noter géophysique. L'enrichissement d'U dans le sandstone peut faire des puits dans son aquifers oxydé au risque pour les concentrations d'U au-dessus des MCL. Les puits ont été logged avec un gamma-ray spectrometer pour déterminer si les anomalies d'U d'eau souterraine pourraient être expliquées par la distribution hétérogène d'U dans le sandstone. Pendant que quelques puits ont montré une corrélation entre les concentrations d'U aqueuses et la concentration d'U dans le sandstone, les autres puits n'ont pas fait, indiquant que les autres facteurs sont aussi importants. $^{235}U/^{238}U$ proportions étaient conforme à U naturellement actuel U. $^{234}U/^{238}U$ proportions d'isotope ont montré empreintes digitales à la caractéristique pour l'eau souterraine de aquifer différent conditionne. Dater d'âge d'eau souterraine peut aider explique des variations dans les concentrations d'U le puit d'eau.

1. INTRODUCTION

U is a naturally occurring radioactive element and a heavy metal in groundwater. In December 2003 the EPA began regulating U in community water supplies in order to reduce the risk of cancer and kidney disease. The EPA estimates that approximately 500 Community Water Systems (1%) will be affected by the new 30 µg/L (ppb) maximum contaminant level (EPA 2000). Other regulatory agencies that have recommended limits on U in drinking water include: the World Health Organization (2 µg/L), Health Canada (20 µg/L), Australian Drinking Water Guidelines (20 µg/L), and Bavarian water authorities (5 µg/L) (Kinze 2002).

U is highly soluble in shallow oxidized groundwater (Ingebritsen 1998). Granite and gneiss aquifers tend to have low U concentrations in the water even though the host rocks are enriched in U. Sandstones typically have lower U concentrations than granites but may produce high groundwater U concentrations (Merkel et al. 2002). Understanding the controls on groundwater U concentrations might help to avoid areas with the potential for elevated U or allow for managing the well system in a manner that negated the need for removing elevated U concentrations to facilitate compliance with the new U regulation. A preliminary characterization of the hydrogeochemical controls on the naturally occurring U isotope distribution in aquifers of the Jacobsville sandstone located in Michigan's Upper Peninsula was conducted over the past three years. Prior to this study, all previous studies of U in these sandstone aquifers were focused on identifying deposits of economic importance for energy generation, with no attention towards the impacts of U on drinking water quality.

Previous studies have examined the effects of redox conditions, host rock U concentration, and groundwater residence time on groundwater U concentrations. Kinze (2002) concluded that areas with high redox and elevated U concentrations in the rock are at risk for high U concentrations in groundwater. In general, case studies have qualitatively shown that elevated U concentrations occurred in oxidized groundwater associated with U-rich formations (*e.g.*, Sinha et al. 1997; Ayotte et al. 2001).

Szabo (1997) directly measured the radionuclide concentrations of different lithologies in the Newark Basin to characterize the distribution of dissolved radionuclides. Szabo concluded that, along with other factors, anomalously radioactive strata were required to produce elevated radioactivity in groundwater. Szabo used borehole-geophysical logging to identify strata with elevated radioactivity, but did not specifically log wells with elevated groundwater U concentrations.

Tricca (2000) used a mathematical model to suggest that groundwater U concentrations should increase linearly with groundwater age. Szabo and Zapecza (1991) found that given sufficient residence time and favorable geochemistry, elevated U could occur in groundwater even without U-rich rocks. Our study investigated whether wide variations in groundwater U concentrations could be attributed in part to differences in groundwater age as well as differences in U concentrations in the host rock.

$^{234}U/^{238}U$ activity ratios are used for converting between U activity (pCi/L) and mass concentration (µg/L) for compliance with the MCL (Wong et al. 1999). Understanding U isotope ratios has broader ramifications because of their potential for tracing groundwater from different aquifer conditions (Porcelli and Swarzenski 2003). Osmond and Cowart (1992) summarized U disequilibria in groundwater based on activity ratios. Where $^{234}U/^{238}U$ activity ratios are near 1 (*i.e.,* secular equilibrium) in the host rock, simple weathering will result in groundwater $^{234}U/^{238}U$ in equilibrium. Elevated $^{234}U/^{238}U$ activity ratios in groundwater occur, in part, due to alpha recoil (Osmond and Cowart 1992). When U-238 decays by alpha decay to Th-234 the Th nucleus may be recoiled out of the mineral into the groundwater. Th-234 then decays via Pa-234 to U-234, resulting in an excess of U-234 in the groundwater. $^{234}U/^{238}U$ activity ratios are used in this work to "finger print" groundwater and investigate whether elevated U concentrations might be derived through different mechanisms, *e.g.*, strong dissolution versus long residence time, in different regions of the study area.

Spatial variations in U concentrations in aquifers of the Jacobsville sandstone in the Upper Peninsula of Michigan were investigated in relation to geochemical conditions, the heterogeneous distribution of U in the host rock, and the increase in groundwater age along flow paths. The distribution of $^{234}U/^{238}U$ activity ratios was also considered to distinguish different mechanisms for elevated U concentrations. The characterization activities in this work included: isotopic U analysis of water samples from 271 wells, detailed analysis of selected wells, spectral gamma-ray logging of wells with U concentrations above and below the MCL, and CFC age dating of wells along a flow path.

2. METHODS

2.1 Study Area

This study focused on aquifers of the Jacobsville sandstone located in Michigan's Upper Peninsula (Figure 1). The Middle Proterozoic Jacobsville sandstone is a red to mottled fluvial-deltaic-lacustrine sequence of sandstones, conglomerates, siltstones and shales deposited as basin fill in the 1.1 Ga old Mid-Continent rift (Hamblin 1958; Kalliokoski 1982). The region has a history of U prospecting, with particular interest in an unconformity-related U deposit at the base of the Jacobsville sandstone, however no economical U deposits were found (Kalliokoski 1976; Johnson 1977). During the 1970s, residential wells were analyzed for U as part of the National Uranium Resource Evaluation (NURE) (Arendt 1980). During that time, there was no consideration of the health risks of U in drinking water. In 2001, local wells were analyzed for (unadjusted) gross alpha content and found to have elevated radioactivity, which combined with the EPA's announcement of a U MCL, provided initial motivation for this study.

2.2 Selection of Wells

Approximately 270 wells were analyzed for isotopic U. Initially wells were selected based on existing elevated gross alpha tests. Subsequently samples from wells were solicited based on areas of interest and availability of well logs. Also some wells that had been part of the National Uranium Resource Evaluation (NURE) were reanalyzed for U. Numerous wells were analyzed by request of the well owners.

2.3 Collection of Samples

Jacobsville sandstone wells are uncased in the sandstone and well water represents a mixture of groundwater from numerous permeable intervals. Because samples were taken from residential wells in daily use, it generally was not necessary to purge the wells of stagnant water. Samples were typically representative of drinking water, however samples from water treatment systems were avoided, or taken in duplicate with untreated samples for comparison. Filtration of select samples with 1 µm size filter did not reduce the U concentration indicating that U was soluble in the water. Samples were collected in polyethylene bottles with size requirements dependant on the lab, preservation with HNO_3 also depended on the lab.

2.4 Uranium Analysis

Most samples from this study were analyzed by ICP-MS. The majority of samples were analyzed for isotopic U at the Laboratory of Isotope and Trace Element Research, at Old Dominion University. Blanks, duplicates, and a blind sample prepared from a standard were included with the well samples. Water samples from the Chassell city test wells were analyzed at Radiation Safety Engineering (Chandler, AZ). In the later stages of the study, requests for U analysis were sent to the Michigan Department of Environmental Quality (MI DEQ) Drinking Water Laboratory, which is certified to test for U in drinking water, but does not report U isotope ratios. The reported U concentrations from these laboratories were generally consistent (deviations of less than 10%) for a given well, even for water samples collected in different years and seasons.

2.5 Redox and Geochemical Analysis

Detailed geochemical analysis for selected wells were conducted at the MI DEQ Lab. A flow cell (FC4000, QED Environmental Systems, Ann Arbor, MI) was used at wells to measure oxidation-reduction potential (ORP), dissolved oxygen, conductivity, pH, and temperature.

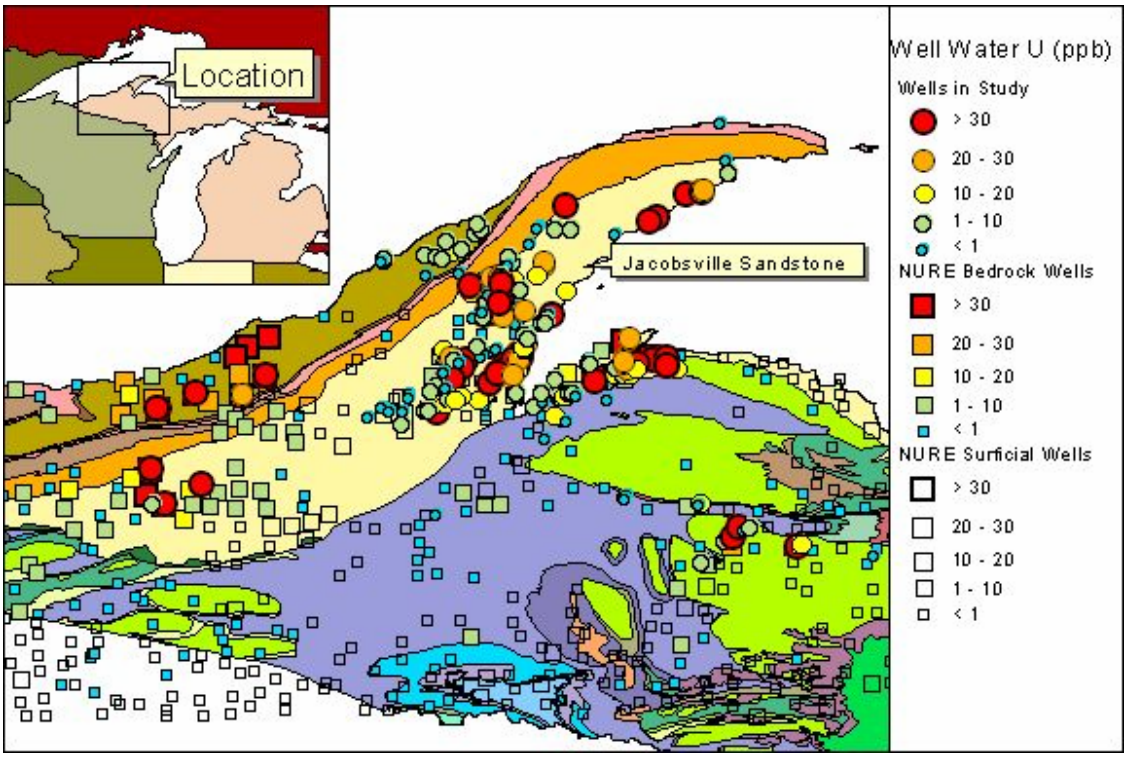

Figure 1. Spatial distribution of well water U (ppb) in relation to bedrock geology.

2.6 Gamma-Ray Spectroscopy

A GRS 2000 gamma-ray spectrometer was rented from the manufacturers, GF Instruments (Brno, Czech Republic). The gamma-ray spectrometer utilizes the characteristic gamma energies of the U-series, Th-series, and ^{40}K to calculate equivalent U (eU), equivalent Th (eTh), and K in wells and at outcrops. The eU concentrations are the same as actual U concentrations if the U-series is in equilibrium. Equilibrium analysis of samples from a Jacobsville sandstone core indicated that the U-series was in equilibrium (Sherman 2004). However, an outcrop sample showed disequilibrium, probably due to weathering effects. Gamma-ray spectroscopy is used routinely in the petroleum industry to identify U-rich fractures and clay type in boreholes (Gearhart Industries 1986). Gamma-ray spectroscopy has been used in groundwater studies to correlate layers (Buckley and Oliver 1990) and is used at outcrops to characterize facies in terms of K (%), eU (ppm), and eTh (ppm) and their ratios (Bristow and Myers 1989; Davies and Elliot 1996; North and Boering 1999; Ehrenberg and Svana 2001).

2.7 CFC Groundwater Age Dating

Cholorfluorocarbons (CFCs) are stable compounds of anthropogenic origin used as refrigerants, solvents, and aerosol propellants. CFCs have accumulated in the atmosphere and recharged into the groundwater at known concentrations since the 1940s. CFCs have been used to age date groundwater since the 1970s (Busenberg and Plummer 1992). CFC samples were collected in glass bottles that were flushed and capped with foil-lined caps underwater in a beaker (USGS 2003). Samples were analyzed by gas chromatograph at the University of Utah Dissolved and Noble Gas Laboratory.

3. RESULTS

3.1 Distribution of U in Groundwater

Approximately 26% of 271 wells analyzed in this study produced water with U concentrations above the MCL (Figures 1 and 2). Approximately 7% of wells produced water with U concentrations over 100 µg/L. The average well water U concentration was 25 µg/L and the median was 8 µg/L. While our study focused on bedrock wells in the Jacobsville sandstone, elevated U was also found in wells in surrounding formations, including the Freda sandstone and Archean granites and gneiss. Water samples collected from surficial wells as part of the NURE program had U concentrations below the MCL, which supported our observations that elevated U was generally associated with bedrock wells. Repeated sampling of wells in this study and sampling of wells from the NURE program indicated that well water U concentrations are generally consistent over time, even when sampled in different years and seasons. Wells drilled through the unconformity at the base of the Jacobsville sandstone did not produce U above the MCL.

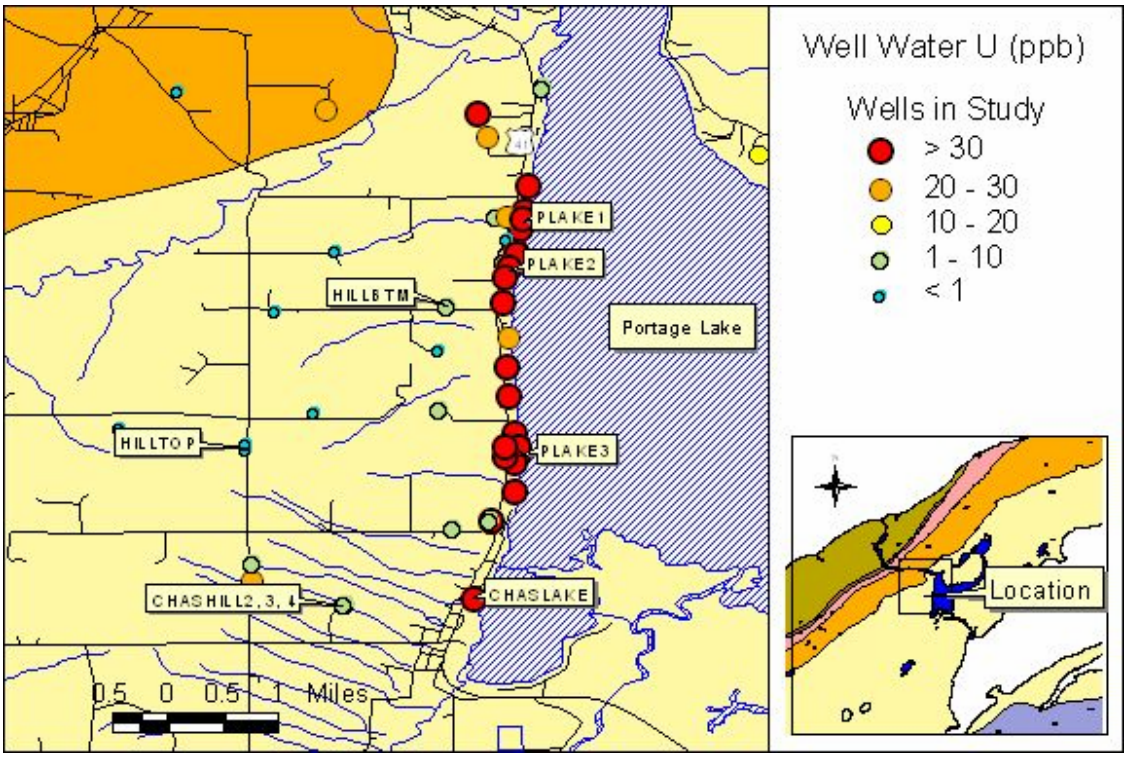

Figure 2. Wells drilled approximately 100-ft (30-m) below lake level produced U above the MCL. Wells that were logged with the spectral gamma-ray probe are denoted with labels.

U concentrations above the MCL frequently occurred in clusters so that neighboring wells tended to have similar U concentrations. A noted exception was found in the Houghton-Chassell area where wells near Portage Lake produced U above the MCL, while wells across US 41 on the hill produced U below the MCL (Figure 2). This apparent anomaly might be explained by differences in well bottom elevations. Wells drilled deeper than ca. 500 ft (150 m) amsl produced water with U concentrations above the MCL.

3.2 U Isotope Ratios

The $^{235}U/^{238}U$ ratios of well water samples were consistent with naturally occurring U (mean mass ratio of 0.0072). $^{234}U/^{238}U$ activity ratios varied from near secular equilibrium to as high as 16, with an average of 2.25 and median of 1.75. The Houghton-Chassell area wells that produced water with U concentrations above the MCL had low activity ratios (<2), perhaps indicating strong dissolution rates that would make recoil relatively less important (Figure 3). In other areas of the study, however, wells that produced U concentrations above the MCL had high activity ratios (>2), perhaps indicating a difference in the rock-water interaction. In areas with high-density data, neighboring wells plotted in distinct fields on charts of $^{234}U/^{238}U$ as a function of the reciprocal of U concentration, which highlights the potential for using U isotope data to trace groundwater from different aquifer conditions.

3.3 Geochemistry

Variations in redox conditions can exert strong control on groundwater U solubility. Well water in this study was consistently oxidized with Eh values of 0.3 to 0.5 V, indicating the potential for high U solubility. Preliminary analysis and modelling suggested that U was generally complexed with carbonates and unsaturated in groundwater. Vanadium may limit U solubility in some areas through precipitation of carnotite and tyuyamunite (which have positive saturation indices) though at U concentrations above the MCL (Sherman 2004).

3.4 Spectral gamma-ray logs of water wells

Nine wells were logged with a spectral gamma-ray probe to characterize the U concentration in the host rock in the near vicinity of the well bore. The locations of logged wells are denoted with labels in Figure 2. In some wells there appears to be a correlation between well water U concentrations and the U concentration of the host rock, as determined by spectral gamma-ray logging of water wells. Figures 4a, b and c show eU logs for 3 wells, PLAKE1, PLAKE2 AND PLAKE3, that produce water with U above the MCL and are located near Portage Lake between Houghton and Chassell. These wells all show eU

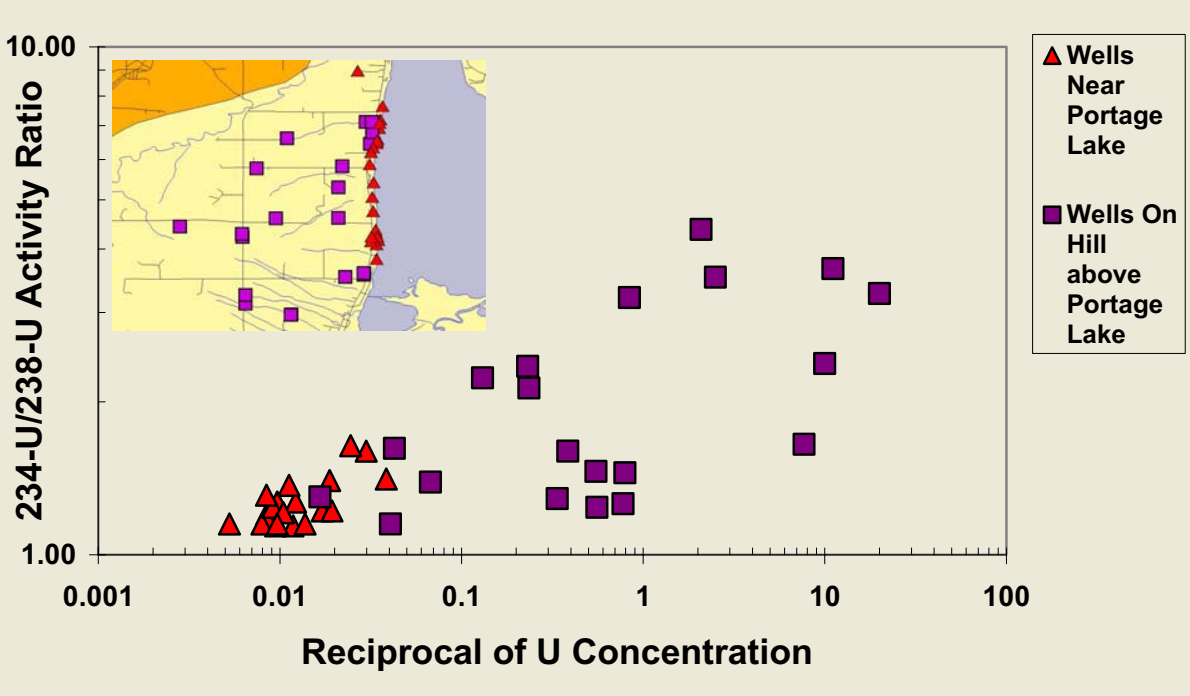

Figure 3. U isotopes have the potential to “finger print” groundwater from different aquifer conditions.

values above 5 ppm and the PLAKE2 well shows a spike of 22 ppm.

The HILLBTM well, which is 140 ft (43 m) above Portage Lake, produced water with low U concentration (1.19 µg/L) and had low U (eU is consistently below 5 ppm) in the host rock (Figure 5a). The HILLTOP well, approximately 300 ft (90 m) above Portage Lake, produced water with extremely low U concentration (0.05 µg/L) and, except for a small spike, shows generally low eU (Figure 5b).

Not all wells, however, show a correlation between well water U concentration and U of the host rock. CHASHILL2, 3, and 4 produced water below the MCL, but show elevated eU on their logs (Figures 6a, b and c). Conversely, CHASLAKE, located down hill from the other test wells, was abandoned based on U levels in the water but did not show high U on the log (Figure 5c), though the groundwater flow path likely passed through U-enriched strata.

3.5 CFC age dating of groundwater

CFC based apparent ages may help explain spatial trends in groundwater U concentrations. The PLAKE2 well, located where groundwater discharges into Portage Lake, produced U above the MCL (82 µg/L) and shows CFC derived apparent groundwater ages (>50 years) near and exceeding the older limit of the dating method. The HILLTOP well, which is located near the top of the topographic divide, produced water with extremely low U (0.05 µg/L) and showed CFC derived apparent groundwater ages near modern (<2 years). Age dating of the HILLBTM well, which is in between the other two along the assumed direction of regional groundwater flow, was inconclusive and may have been contaminated with atmospheric CFC.

4. DISCUSSION

Selection of some wells based on elevated gross alpha tests, elevated NURE results, and word of mouth, may have biased the study toward areas with elevated U. Because wells are uncased in the Jacobsville sandstone well water U concentrations represent a mixture of groundwater from numerous intervals. If relatively U-rich strata have low permeability these intervals may not contribute significant water to the well. The oxidation-reduction potential of well water was measured using a flow cell to isolate the probe from the atmosphere, however, the measured redox of well water may not accurately reflect redox conditions of the aquifer as groundwater may mix with air in the borehole. Groundwater ages of some wells in this study area are near the older limit of the CFC dating method. Additionally, the apparent ages derived from the CFC-11, CFC-12 and CFC-113 were not always consistent.

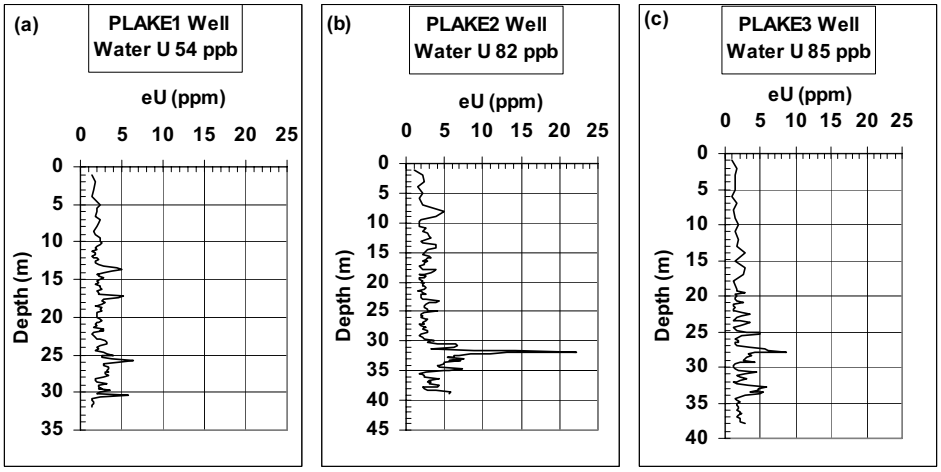

Figure 4. Spectral gamma-ray logs of wells showing variable eU enrichment. These wells produce water with U concentrations above the MCL.

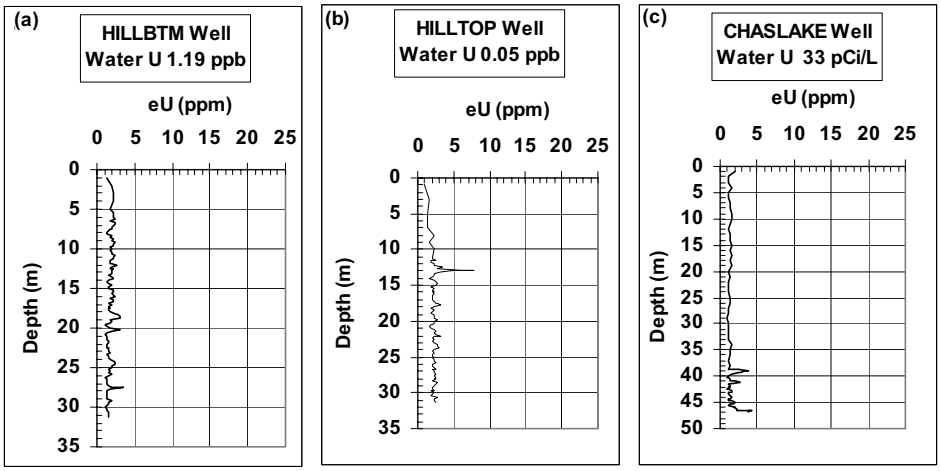

Figure 5. Spectral gamma-ray logs of wells with low eU. These well produce water with low, extremely low and high U.

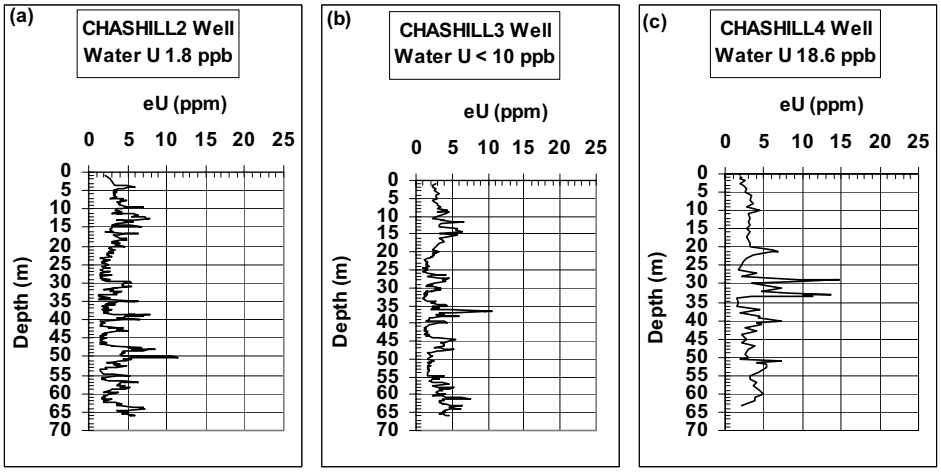

Figure 6. Spectral gamma-ray logs of wells with high eU but well water U concentrations below the MCL.

The results of this study are consistent with the assessment by Kinze (2002) that areas with high redox and elevated U in the rock are more prone to elevated U in the groundwater, at least on a formation or basin scale. The occurrence of U above the MCL in aquifers of the Jacobsville sandstone is likely related to U-enrichment in the formation. Furthermore, our results agree with Szabo (1997), who found that some wells penetrating anomalously radioactive strata did not produce water with elevated radioactivity, *e.g.*, a well penetrating a reducing mudstone with elevated U did not produce water with elevated U. In our study area the CHASHILL2, 3 and 4 wells showed anomalous U in the sandstone but aqueous U was below the MCL. Due to the red-bed characteristic of the Jacobsville sandstone it is not likely to have reduced beds, so other factors, *e.g.,* permeability of strata or groundwater residence time are likely to be important. Szabo (1997) also concluded that the presence of radioactive strata was required to produce elevated radioactivity in groundwater in the Newark Basin. Our study found that wells that produce water with elevated U do not necessarily have elevated U in the surrounding rock.

Ivanovich (1991) showed that $^{234}U/^{238}U$ activity ratios increase asymptotically along flow paths in shallow oxidized aquifers and jump to high values at redox fronts, where low U solubility results in precipitation of U on the aquifer grains which, in turn, enhances alpha recoil derived ^{234}U. Where redox fronts are absent, such as in our study area, another mechanism is needed to explain high $^{234}U/^{238}U$ activity ratios. Tricca (2000) showed that high $^{234}U/^{238}U$ activity ratios can also be achieved through low weathering rates. Dabous and Osmond (2001) showed that groundwater with high U concentration frequently has low activity ratios because alpha recoil derived ^{234}U is relatively less important under strong dissolution. This relationship was observed in Houghton-Chassell area where wells with high groundwater U concentrations have low $^{234}U/^{238}U$ activity ratios. Tricca (2000) showed that groundwater with both high U concentrations and high $^{234}U/^{238}U$ activity ratios may indicate an influence of the vadose zone or low weathering rates combined with long flow distances or low water velocities. Elsewhere in our study area, wells have both high groundwater U concentrations and high $^{234}U/^{238}U$ activity ratios (Sherman 2004). Porcelli and Swartzenski (2003) explained that changes in U host phase or geochemistry influence weathering without influencing recoil, which results in a change in the activity ratio. Thus, variations in $^{234}U/^{238}U$ in our study area may indication difference in U host phase, travel times, and/or influence from the vadose zone.

Osmond and Cowart (1992) developed plots of $^{234}U/^{238}U$ versus the reciprocal of U concentration and interpreted straight-line arrays as evidence of mixing or dilution of different groundwater. The Houghton-Chassell wells in our study area plotted in a linear array extending from the lower left corner to the upper right corner, which may be interpreted as a mixing of groundwater with low U concentration and a high activity ratio with groundwater with recently leached U and a low activity ratio. Though not common, very high $^{234}U/^{238}U$ activity ratios occurred in our study area. This is consistent with finding by Osmond and Cowart (1992) that activity ratios of 5, 10, and even 20 or higher have been measured in steady long-term flow systems of large confined aquifers.

5. CONCLUSIONS

Approximately 26% of 271 wells analyzed in this study produced water with U concentrations above the MCL. Elevated U was associated with bedrock rather than surficial wells. Well water U concentrations were generally consistent over time. U concentrations above the MCL frequently occurred in clusters and neighboring wells tended to have similar U concentrations except where topography resulted in significant differences in well bottom elevations. U-enrichment in the Jacobsville sandstone may make wells in its oxidized aquifers at risk for U concentrations above the MCL. For some wells groundwater U concentrations may be directly related to U concentrations in the surrounding strata. The presence of elevated U in the sandstone did not necessarily mean that wells would produce water with U concentrations above the MCL, indicating that other factors are also important. Groundwater age may help explain trends in groundwater U concentrations. $^{235}U/^{238}U$ ratios were consistent with naturally occurring U. $^{234}U/^{238}U$ activity ratios varied from near secular equilibrium to 16 with an average of 2.25. In areas with high-density data neighboring wells plotted in distinct fields or arrays on charts of $^{234}U/^{238}U$ versus the reciprocal of U concentration, which highlights the potential for using U isotope data to trace groundwater from different aquifer conditions.

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