Elevated radium (Ra) and barium (Ba) concentrations are found in deep bedrock aquifers in parts of northern Illinois, and are the most important water quality issues for public water suppliers in most of northern Illinois. In addition to health considerations, the presence of Ba can cause precipitation of barite (BaSO₄) in well bores, pumps, and discharge pipes, necessitating expensive maintenance. Because most wells drilled into deep bedrock aquifers are open to multiple aquifers, allowing waters to mix within the borehole, it is difficult to establish if one or more specific aquifers are primarily responsible for the elevated Ra and/or Ba. Indirect evidence from previous studies suggested that the Ironton-Galesville aquifer may be the major source of contamination. The objective of this project was to identify and sample wells open only to individual deep bedrock aquifers in the Chicago region, and analyze for radium isotopes (²²⁶Ra, ²²⁸Ra), Ba, and complete inorganic chemistry. A better understanding of the occurrence of Ra and Ba may help public water suppliers in siting future deep bedrock wells.

A total of 25 wells open to only one of four deep bedrock aquifers in five geographic groups in northern Illinois were sampled in August 2007. The four aquifers are: (1) Galena-Platteville; (2) Ancell (St. Peter); (3) Ironton-Galesville; and (4) Mt. Simon. Groups were selected based on proximity of wells open to different aquifers. All groups were located in areas known to have Ra activities above its drinking water standard (5 pCi/L); two groups were located in the region with Ba concentrations above its standard (2 mg/L). In addition to Ra isotopes and Ba, samples were analyzed for major ions, minor and trace elements, dissolved organic carbon, hydrogen sulfide, and methane.

Radium activities greater than 5 pCi/L were found in all groups, while Ba concentrations greater than 2 mg/L were found only in northeastern Kane County and one well in western Lake County. Ironton/Galesville wells tended to have higher total Ra activities than samples from other aquifers, being most visibly obvious within the geographic groups (Figure 1). Although the highest Ra activities generally were from Ironton/Galesville wells, the highest total Ra activity was found in a Mt. Simon well, although two other Mt. Simon wells sampled did not have particularly high Ra levels.

²²⁶Ra was detected in all samples and ²²⁸Ra was detected in all but four samples (> 1.0 pCi/L). ²²⁸Ra was the primary source of radioactivity in all the Mt. Simon wells and most of the Ironton/Galesville wells. In contrast, ²²⁶Ra was the primary source of radioactivity in all the Ancell and Galena/Platteville wells.

Conditions were reducing throughout the study area. Sulfide was detected (> 0.1 mg/L) in about half of the wells, as was methane. Sulfate concentrations were generally lowest in group 2 wells, which had the highest Ba concentrations and relatively high Ra levels. Barium concentrations were related to redox-sensitive species, particularly SO₄²⁻ and Fe. Samples with elevated Ba concentrations had low concentrations of SO₄²⁻ and Fe, while samples with relatively elevated
SO$_4^{2-}$ or Fe had low Ba concentrations. Samples that had elevated Ba had low concentrations of SO$_4^{2-}$, Fe, and H$_2$S and were supersaturated with respect to barite; these were primarily wells in group 2.

The drinking water standard for Ra was exceeded in wells from all aquifers. Differences in $^{226}$Ra/$^{228}$Ra ratios as a function of aquifer identity suggested there may be some differences in the source and mechanism of release of Ra into solution. The smaller percentage of $^{226}$Ra in the Ironton/Galesville and Mt. Simon aquifers compared to the shallower aquifers may indicate less U in the deeper aquifer materials and/or that a significant fraction of the $^{226}$Ra that has been produced has been transported away.

Barite appeared to be the primary control of both Ra and Ba concentrations. In areas where SO$_4^{2-}$ reduction has removed SO$_4^{2-}$ from solution (i.e., group 2), Ba can accumulate in solution. This in turn disables a mechanism for Ra removal (i.e., co-precipitation in barite). Three of the wells with Ba concentrations above the drinking water standard had the three greatest Ra activities.

It appears that geographic location exerts a stronger control on overall groundwater chemistry, specifically Ra and Ba, than does the identity of the particular deep bedrock aquifer. Previous sampling indicated that Ra and Ba concentrations are low west of our study area where the Maquoketa Formation is absent, groundwater is relatively young and dilute, and conditions are not strongly reducing. Down-gradient of this area in our study area, conditions become SO$_4^{2-}$-reducing, and Ra and Ba can accumulate in solution. Further down-gradient where SO$_4^{2-}$ is reintroduced into solution, Ra and Ba concentrations decrease. It appears that each aquifer (at least from the Ancell and deeper) is following similar geochemical evolutionary pathways with similar effects on Ra and Ba chemistry. An additional factor to consider is that there are a large number of wells penetrating through all the deep bedrock aquifers in this region, which may allow for enhanced leakage between the aquifers. This may have the effect of reducing geochemical differences among aquifers.

Figure 1. Total Ra ($^{226}$Ra + $^{228}$Ra), $^{226}$Ra, and $^{228}$Ra activities as a function of sample group and source aquifer.