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Geologic Studies to Identify the Source for High Levels of Radium and Barium
in Illinois Ground-Water Supplies: A Preliminary Report

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ABSTRACT

Analyses of water from municipal wells in Illinois by the Illinois Environmental Protection Agency showed that more than 300 wells exceeded the upper limit, 3 picocuries/liter (U.S. Public Health Service, 1962), for gross alpha radiation in drinking water. More than 30 wells exceeded the upper limit, 1 milligram/liter (U.S. Public Health Service, 1962), for barium in drinking water. High levels of radiation in ground water were more extensive in areal distribution than the high levels of barium. All of the affected wells were finished in bedrock, primarily in rocks of the Cambrian and Ordovician Systems of northern Illinois. The geologic settings in which the high levels of radiation and barium were documented indicated that the problem was not restricted to Illinois.

The source of the radiation in ground water was thought to be the natural occurrence of the radioactive elements uranium-238 and thorium-232 in the aquifer rocks. Analyses of a limited number of rock samples indicated that uranium and thorium concentrations were highest in fine-grained sediments in the aquifer systems; the highest concentration was in shales that confine the aquifer.

The occurrence of natural radioisotopes in ground water was thought to be complex, involving source rocks, ground water chemistry, and the hydraulic stress placed on the aquifer.

Chemical analyses of rock samples indicated that high concentrations of barium were widespread in rocks of the Cambrian and Ordovician Systems. The concentration of barium in ground water was controlled by solubility equilibria reactions with sulfate ion. A map showing sulfate ion concentration in the Cambrian-Ordovician Aquifer could be used to delimit regions where barium might occur at concentrations exceeding 1 milligram/liter.

KEY WORDS

aquifer properties/barite/barium/chemical properties/ground water/radioactivity/radioisotopes/radium/sedimentary rocks/solubility/sulfate/thorium/uranium.

INTRODUCTION

For a year we have investigated the natural occurrence of radium and barium in ground water of Illinois at levels of concentration greater than those recommended in health standards; research will continue through 1979. An inventory of analyses of ground water from municipal wells in Illinois was provided by the Illinois State Environmental Protection Agency. These analyses showed that the high concentrations of radiation in the drinking water were essentially restricted to the Cambrian-Ordovician Aquifer in northern and central Illinois. High concentrations of barium were also found to be generally limited to this aquifer in northern Illinois; highest concentrations were restricted to an area in northeastern Illinois. The geologic settings in which the high levels of radiation and barium have been documented indicate that the problem was probably not restricted to Illinois.

The Cambrian-Ordovician Aquifer is composed of thick sections of sandstone and dolomite interbedded with shale. In downward order, the aquifer consists of the Galena-Platteville Dolomite, Glenwood-St. Peter Sandstone, and Prairie du Chien Group of Ordovician age, followed by the Trempealeau Dolomite, Franconia Formation, and the Ironton-Galesville Sandstone of Cambrian age. The Ironton-Galesville Sandstone is underlain by shales of the Eau Claire Formation that have very low permeability (Walton, 1965). The Eau Claire Formation is underlain by the Mt. Simon Sandstone. Available data indicate that regionally the entire sequence from the Galena-Platteville Dolomite through the Ironton-Galesville Sandstone behaves hydraulically as one aquifer (Walton, 1965).

In northern Illinois, some municipal wells were finished in the Mt. Simon Sandstone as well as in the overlying Cambrian and Ordovician strata. No wells receive ground water only from the Mt. Simon Sandstone, however. Wells in the Cambrian-Ordovician bedrock are usually cased to dolomites in the upper unit of the aquifer and are then open-hole to their total depth, receiving water from a large section of rock. Liners have been placed against incompetent strata in some wells. In northeastern Illinois wells may have open intervals greater than 300 meters (1000 ft) and total depths greater than 450 meters (1500 ft). Wells finished in the Mt. Simon Sandstone may have total depths greater than 610 meters (2000 ft).

In northwestern and north-central Illinois, where the uppermost unit of the aquifer is the Galena-Platteville Dolomite, which directly underlies the glacial drift, recharge of the aquifer is produced from vertical leakage of water through glacial drift. In northeastern Illinois, where the Maquoketa Formation overlies and confines the aquifer, much recharge is produced from rocks of Silurian age or the glacial drift by vertical leakage through the Maquoketa Formation (Walton, 1965). Southward, the Cambrian-Ordovician bedrock dips into the Illinois Basin, and the aquifer is overlain and confined by a thick sequence of fine-grained rocks, including shales of the Pennsylvanian System. In general, water in the aquifer becomes more highly mineralized as the rock strata deepen into the basin. In central Illinois, sandstones in the aquifer are productive, but the ground water is of marginal quality because of its mineralization.

RADIUM

The source of radiation in ground water is apparently the natural occurrence of the radioactive elements uranium-238 and thorium-232 in the aquifer rocks. Uranium and thorium, as minor constituents in the lithosphere, are present in trace amounts in almost all geologic materials. The calculated

average composition of uranium and thorium in sedimentary rocks of different lithologies is shown in table 1. Each atom of thorium and uranium decays through discrete transformations and characteristic half-lives by the emission of several radioactive alpha and beta particles and gamma rays (figs. 1, 2, and 3).

Radium-226, a naturally occurring radioactive daughter of uranium-238, is a potential health hazard when present in ground water. Radium-226 is of special concern, because it has a relatively long half-life (1622 years) and high specific activity. The human body metabolizes radium in much the same way that it metabolizes calcium—radium-226 will become concentrated in bones if it is ingested over a long period. The relationship between exposure to radium-226 and health risk is not well established. In 1962, the United States Public Health Service recommended that gross alpha activity in drinking water not exceed 3 picocuries/liter (pCi/l). One picocurie of radium-226 equals 1×10^{-12} grams. An inventory of gross alpha activity in raw ground water from municipal wells in Illinois by the Illinois Environmental Protection Agency revealed that more than 300 wells in Illinois exceeded the gross alpha radiation limit of 3 pCi/l. This is a conservative limit, chosen to allow for the possibility that all alpha activity might be caused by radium-226, although the gross alpha activity may include other, less hazardous radioisotopes. In 1976 the United States Environmental Protection Agency enacted regulations to limit the average annual concentration of gross (total) alpha radioactivity in drinking water to 15 pCi/l and the average annual concentration of radium-226 and/or radium-228 to 5 pCi/l. The regulations state that the average annual concentration of gross (total) beta radioactivity should not exceed a concentration of 50 pCi/l.

The identity of radioisotopes in ground water of the Cambrian-Ordovician Aquifer and the form in which they are present, in solution or sorbed on colloids or finely divided particulate matter, is not well understood. Bennett, Markwood, and Bell (1976) studied municipal water supplies in northeastern Illinois and found that water treatment processes such as ion-exchange softening, lime

softening, and filtration to remove iron significantly reduced gross alpha activity, gross beta activity, and radium-226 concentrations in ground water. The investigators also found in all cases that the gross alpha activities were much greater than could be supported by the concentration of radium-226 in the water sample.

We determined gross alpha activity and gross beta activity for unfiltered and filtered raw ground-water samples collected from five municipal supplies (table 2). Where gross alpha activity or gross beta activity occurred at high levels, filtration may have removed radioisotopes sorbed on finely divided particulate or colloidal matter in the ground water. Filtration had little effect on the radiation of ground-water samples with low activity levels (table 2).

The membranes used to filter samples of ground water collected at Herscher, Lostant, and Lynwood were analyzed at Argonne National Laboratory by means of the radon emanation technique (Lucas, 1964) to determine the concentration of radium-226 on the filters. The filters contained little radium-226 (the measured concentrations were essentially zero), so the radium-226 in these samples must have been in solution. Therefore, the observed decrease in gross alpha activity from unfiltered to filtered samples must be related to the presence of other radioisotopes in the ground water.

To identify the source for radioisotopes present in ground water, selected rock samples were analyzed for radium-226 and radium-228 at Argonne National Laboratory with a germanium-lithium detector. The concentrations shown in figures 4 and 5 are equivalents of uranium-238 and thorium-232 calculated with an assumption of equilibrium between the parent-daughter nuclides. The limited number of analyses performed indicated that concentration levels are highest in the fine-grained lithologies in the aquifer rocks and in the shales that confine the aquifer.

Concentrations of thorium-232 (radium-228) in the dolomites were generally much greater than the calculated average composition in carbonates (table 1 and figure 4). Concentrations measured in sandstones were in agreement with the average composition (3.9 ppm), although a concentration of 22.7 ppm thorium-232 (radium-228) was detected in a sample of Mt. Simon Sandstone (lower Cambrian). The composition of most shales agreed with the calculated average composition; however, shales of the Maquoketa Group, and especially the Pennsylvanian System, had higher concentrations of thorium-232 (radium-228).

The highest concentrations of uranium-238 (radium-226) were in shales of the Pennsylvanian System; these concentrations were much greater than the calculated average composition of 4.5 ppm. Shales in the Cambrian System contained slightly less uranium-238 (radium-226) than the average composition in shales. The concentration of uranium-238 (radium-226) in dolomites ranged from .35 ppm in dolomites of the Platteville Group to 4.75 ppm in shaley dolomites of the upper Cambrian System; the average composition in carbonate rocks is calculated to be 2.2 ppm. The concentration of uranium-238 (radium-226) in sandstones ranged from 0.75 ppm to 2.8 ppm; the concentration in most samples correlated closely with the calculated average composition of 1.0 ppm.

Whereas thorium-232 decays directly to radium-228, uranium-238 decays to radium-226 in five steps (fig. 1). Disequilibrium between uranium-238 and radium-226 may result from differences in decay energies and chemical behavior of the daughter nuclides in aqueous systems. To evaluate disequilibrium, splits of 25 rock samples were analyzed directly for uranium-238 (radium-226) at Argonne National Laboratory and for uranium-238 at the Illinois State Geological Survey by instrumental neutron activation. Results of analyses indicated that disequilibrium exists in some samples. Sixteen of the 25 samples were in equilibrium. For rock samples of the Cambrian and Ordovician Systems, two sandstone samples were slightly enriched in radium-226, whereas one sandstone sample, one dolomite sample, and two shale samples were found to be slightly depleted in radium-226. Further investigation will

be made of disequilibrium in aquifer rocks and in shales that confine the aquifer.

Increased concentrations of radium-226 and other radioisotopes in ground water may have resulted from the interaction of several mechanisms. The source of the radiation might be stratigraphically or regionally restricted rock strata; however, source rocks might occur throughout the aquifer, and the presence of radioisotopes in solution might be controlled by physico-chemical interactions at the aquifer rock-water interface. The Eh, pH, ionic strength, and chemical composition of the ground water may affect the activity of radioisotopes in solution.

Using available information (Krause, 1960, and Bennett, Bell, and Markwood, 1976) we studied the control of radium-226 activity by solubility equilibria reaction with sulfate ion. Radium sulfate ($K_{sp} = 4.25 \times 10^{-11}$) is the most insoluble of the alkaline earth sulfates and is also the most insoluble radium salt known (Kirby and Salutsky, 1964). The maximum activities of radium-226 measured in ground water are several orders of magnitude less than those allowable by the K_{sp} of radium sulfate, however. Also, the lowest activities of radium-226 (<1 pCi/l) occur in recharge areas where sulfate ion activity is commonly less than 10 mg/l. Down-gradient in the aquifer where high radium-226 activities occur (≥ 30 pCi/l), sulfate ion activities may be greater than 700 mg/l. Apparently other mechanisms limit the activities of radium in solution to levels much lower than would be maintained by solubility equilibria reactions.

The chemistry of barium in aqueous systems may affect the concentration of radium in solution. Where barium is present in solution at high levels of concentration in the Cambrian-Ordovician Aquifer, coprecipitation by isomorphous replacement with a salt such as barium sulfate may remove radium ions from ground water.

Cation exchange sites on the aquifer rock may have an important effect on the concentration of radium-226. Emrich and Lucas (1963) noted a good correlation between increase in radium-226 activity and increase in mineralization of

ground water. Ionic strength varies from less than 0.007 molar in recharge areas to greater than 0.028 molar down-gradient in the basin. Although it has not been carefully investigated, this relationship suggests that cation exchange may be an important mechanism in controlling the concentration of radium-226 in solution.

A phenomenon that is related to cation exchange sites and that is unique to radioisotopes is the "Szilard-Chalmers" or "hot atom" effect. This phenomenon is the momentum imparted to an atom during decay by emission of an alpha particle. The recoil energy caused by this process may be sufficient to expel radium-226 atoms from exchange sites occupied by thorium-230 parents. This mechanism may be more effective as the ionic strength of the ground water increases.

The high levels of radiation in ground water from the Cambrian and Ordovician Systems in northeastern Illinois may be related to hydraulic stress on the aquifer. The Cambrian-Ordovician Aquifer is an important source of ground water and is heavily pumped in northeastern Illinois. Estimated pumpage from the aquifer increased from 200,000 gallons per day in 1864 to 78 mg/d (million gallons per day) in 1958, 165.7 mg/d in 1975, and 181.4 mg/d in 1976 (Robert T. Sasman, personal communication, 1978). Large volumes of ground water have been withdrawn in northeastern Illinois; in the Chicago region, the piezometric surface of the aquifer has changed more than 210 meters (700 ft) in some places (fig. 6). The withdrawal of ground water from fine-grained rock strata in the aquifer as a result of this stress may have caused increased concentrations of radiation; however, a positive relation between pumpage and radiation levels on a regional basis in northeastern Illinois has not yet been found because of problems with the available data. Nevertheless, an evaluation of the available radiation analyses has shown that hydraulic stress on the aquifer is not the only mechanism responsible for the high levels of radiation. High radiation levels were also present in samples of ground water from new wells finished in the Cambrian-Ordovician Aquifer in regions where the aquifer is not heavily pumped.

BARIUM

The Illinois Environmental Protection Agency inventory of municipal wells showed that more than 30 municipal wells exceeded the standard for maximum concentration of barium in drinking water—1 mg/liter (U.S. Public Health Service, 1962). The maximum barium concentration detected in these wells was 18 mg/l. The occurrence of such high concentrations of barium in potable ground water is uncommon and not well documented. All of the municipal wells except one are finished in the Cambrian-Ordovician Aquifer in northern Illinois. The exception, a municipal well in western Illinois, was finished in limestone bedrock of the Devonian System. Barium concentrations measured in this well range from 1.0 to 2.6 mg/l. Barium concentrations as great as 5 mg/l have been reported in chemical analyses of domestic wells which are finished in bedrock younger than Ordovician; however, water samples from domestic wells are so rarely analyzed for barium that it is impossible to determine the extent of barium in these less-productive bedrock aquifers.

Barium concentrations were determined for unfiltered and filtered raw ground-water samples from selected wells (table 3). Most of the barium was present in solution. Where the concentration of barium was greater than approximately 5 mg/l, filtration reduced the concentration somewhat, indicating that part of the barium was present on finely divided colloidal or particulate matter. The barium removed by filtration might have been present sorbed on metal oxides or hydroxides or as barium sulfate.

To evaluate the bedrock as the source of barium in ground water, rock samples were selected and analyzed for barium by X-ray emission spectroscopy (fig. 7). The calculated average composition of barium in sedimentary rocks is presented in table 4. Barium concentrations in the Cambrian and Ordovician Systems ranged from less than 25 ppm to greater than 1100 ppm. The lowest concentrations were in dolomites of the Galena and Platteville Groups. The highest concentrations were in

fine-grained sediments of the Glenwood (upper Ance11) and Eau Claire Formations (lower Cambrian). Barium concentrations were higher in finer-grained sediments and in older sediments. Many samples of shale and sandstone contained concentrations of barium that were significantly greater than the calculated average composition.

Because barium was widely present in the bedrock but restricted in its regional occurrence in ground water of the Cambrian and Ordovician Systems, the chemical controls of barium solubility were investigated. Barium sulfate is highly insoluble, and, in aqueous systems, equilibria reaction with sulfate ion is a likely control on barium ion concentration. The isocon lines in figure 8 show sulfate ion concentration in the Cambrian-Ordovician Aquifer in northeastern Illinois; the stippled pattern indicates regions where barium concentrations greater than 1 mg/1 have been detected. Much of the Cambrian-Ordovician Aquifer in northern Illinois has sulfate ion concentrations less than 10 mg/1. Down-gradient in the aquifer, sulfate ion concentration increases rather suddenly along a northeast-to-southwest-trending belt in northeastern Illinois. Where sulfate ion concentration is greater than approximately 50 mg/1, barium is rarely present at significant levels of concentration; however, the highest barium concentrations in the Cambrian-Ordovician Aquifer were detected where the sulfate ion concentration starts to increase.

Figure 9 is a graph of barium ion activity as a function of sulfate ion activity in samples of ground water collected from selected wells in northern Illinois. The negative log of barium ion activity is plotted on the vertical scale, and the negative log of sulfate ion activity is plotted on the horizontal scale. The diagonal lines on the graph represent the solubility product for barium sulfate calculated from free energies, for which the value is $10^{-8.9}$ (Garrels and Christ, 1965) and as determined experimentally, for which the value is 10^{-10} (Sil1én and Martell, 1964). The graph shows that for most water samples, the barium

ion activity is controlled by solubility equilibria with sulfate ion activity.

The samples represented by the symbol X were collected from dolomites present at the base of the Maquoketa Group. These samples have anomalously low concentrations of calcium, magnesium, and strontium and high concentrations of sodium and bicarbonate. For these samples the control on concentration of barium is apparently cation exchange on fine-grained shale membranes in the Maquoketa Group.

Membrane effects do not explain the barium ion activity present in the cluster of three water analyses shown on the graph below the 10^{-10} solubility product line. These samples may simply reflect a lack of available barium in the source rocks to go into solution.

Two separate analyses for three wells (fig. 8) illustrated that barium ion activity and sulfate ion activity do not necessarily remain constant at some sampling locations. Figure 9 shows that water samples from these wells were occasionally supersaturated with respect to the $10^{-8.9}$ solubility product. Of particular interest is the precipitation of barium sulfate on the pumps and in the columns of these wells which causes difficulty in maintenance. (Here we have a situation in which theoretical calculations have successfully predicted a phenomenon.) These wells receive water laterally—sulfate-rich water from the east and low-sulfate water that is barium-rich from the west. Mixing these waters in the well column caused occasional conditions of supersaturation, as shown by barium sulfate solubility equilibria, and, hence, precipitation.

In the region where the sulfate concentration in ground water undergoes significant lateral changes, it would be possible to restrict the barium concentration in municipal ground-water supplies to comply with the drinking water standard by locating wells where the ground water contains sulfate ion at a concentration of approximately 50 mg/l or greater. In some regions there may be a vertical variation in sulfate ion concentration in the aquifer whereby casing off

strata that contain low-sulfate ground water may restrict the concentration of barium in ground water pumped from the well. The distribution of sulfate ion in the aquifer system is under further investigation.

SUMMARY

The occurrence of high levels of radiation in ground water of Illinois is essentially restricted to the deep Cambrian-Ordovician Aquifer in northern and central Illinois. High levels of barium were generally limited to this aquifer in an area of northeastern Illinois.

The natural radioisotopes in ground water were thought to be daughter nuclides from the decay of uranium-238 and thorium-232 in bedrock of the aquifer system. Analyses of a limited number of rock samples by using a germanium-lithium detector determined that the concentration of uranium in most samples was in general agreement with the calculated average composition in rocks of these lithologies; highest concentrations were in shales and shaley sandstones, and lowest concentrations were in dolomite. The thorium concentration in sandstone and shales of the aquifer system correlated closely with the average composition in rocks of these lithologies, whereas concentrations of thorium in some dolomites of the aquifer system were significantly higher than the average composition in carbonate rocks. In general, the higher thorium concentrations were in shaley sediments. From the limited analytical data available, it was impossible to identify specific stratigraphic units as sources for radioisotopes in ground water. The high level of radiation in ground water was thought to be a complex phenomenon, possibly involving the aquifer rock, ground-water chemistry, and hydraulic stress on the aquifer.

The analyses of rock samples by X-ray emission spectroscopy determined that the barium concentration in shales and sandstones widely present in the Cambrian-Ordovician Aquifer were significantly greater than the average composition

for rocks of these lithologies. Barium ion concentration in ground water was controlled by the concentration of sulfate ion; the measured concentrations were in good agreement with the solubility product of barium sulfate. An isoconcentration map of sulfate ion in ground water of the Cambrian-Ordovician Aquifer defined regions where barium might occur in ground water. For some municipalities the map could be used to locate wells where the ground water contains sufficient sulfate ion to prevent the presence of barium ion at concentrations greater than the standard for maximum concentration of barium in drinking water.

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Table 1. Calculated average composition of uranium and thorium

Type of rock	U (ppm)	Th (ppm)	Th/U
Igneous	2.8	11	3.9
Sedimentary			
Sandstone	1.0	3.9	3.9
Shale	4.5	13	2.9
Carbonate	2.2	0.20	.09

SOURCE: After Horn and Adams, 1966.

Table 2. Effect of filtration^a on gross alpha and beta activity in raw ground-water samples from selected municipal wells finished in the Cambrian-Ordovician Aquifer

Location	Radiation (pci/l)			
	Alpha-		Beta-	
	Unfiltered	Filtered	Unfiltered	Filtered
German Valley well no. 1 (Sec. 32, T. 26 N., R. 9 E., Stephenson County)	4 ± 2	4 ± 2	8 ± 2	6 ± 2
Hanover Park well no. 4 (Sec. 36, T. 41 N., R. 9 E., Cook County)	7 ± 3	8 ± 4	19 ± 6	22 ± 6
Herscher well no. 5 (Sec. 29, T. 30 N., R. 10 E., Kankakee County)	13 ± 6	12 ± 6	23 ± 7	33 ± 8
Lostant well no. 4 (Sec. 24, T. 31 N., R. 1 E., La Salle County)	56 ± 12	12 ± 6	43 ± 4	5 ± 2
Lynwood well no. 2 (Sec. 7, T. 35 N., R. 15 E., Cook County)	140 ± 20	74 ± 17	91 ± 5	82 ± 5

SOURCE: Illinois Environmental Protection Agency.

^a0.2-micron Sartorius membrane filter.

Table 3. Effect of filtration^a on barium concentration in ground-water samples from selected wells finished in the Cambrian-Ordovician Aquifer

Location	Barium (mg/l)	
	Unfiltered	Filtered
West Dundee well no. 1 (Sec. 27, T. 42 N., R. 8 E., Kane County)	9.1	7.5
German Valley well no. 1 (Sec. 32, T. 26 N., R. 9 E., Stephenson County)	6.3	5.5
Hanover Park well no. 4 (Sec. 36, T. 41 N., R. 9 E., Cook County)	3.6	3.6
Illinois Tollway Garage (Sec. 3, T. 42 N., R. 6 E., Kane County)	2.6	2.5
Domestic well (Sec. 12, T. 40 N., R. 8 E., Kane County)	0.5	0.5
Buck Lake Subdivision (Sec. 28, T. 37 N., R. 5 E., De Kalb County)	0.1	0.1

^a0.2-micron Sartorius membrane filter.

Table 4. Calculated average composition of barium (ppm)

Type of rock	Ba
Igneous	595
Sedimentary	
Sandstone	193
Shale	250
Carbonate	30

SOURCE: After Horn and Adams, 1966.

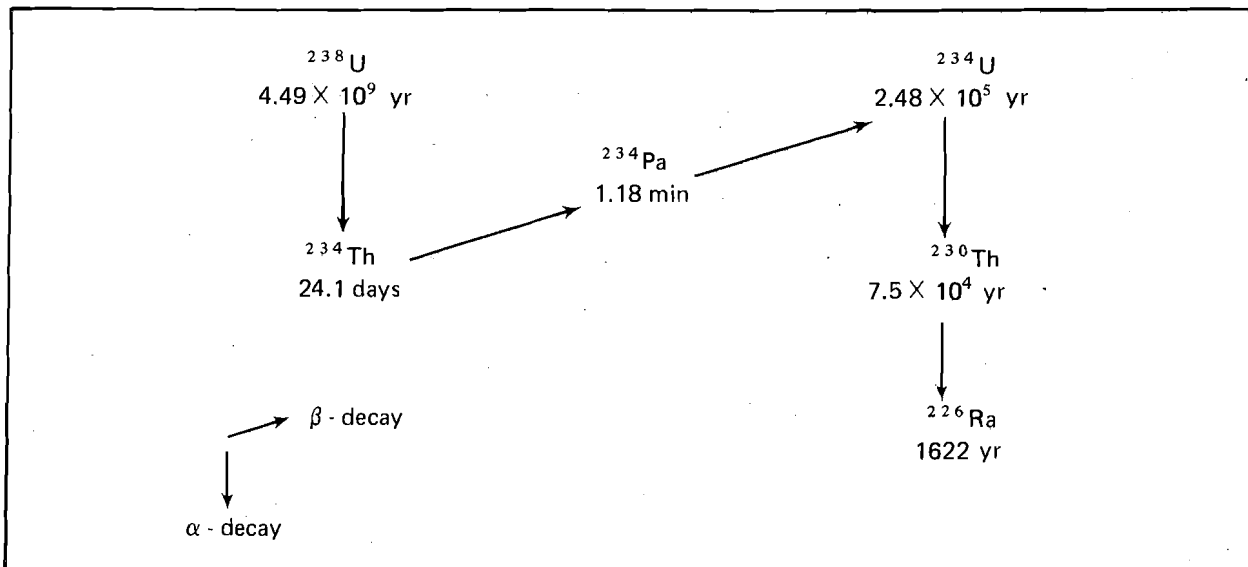


Figure 1. The radioactive decay series and half-lives of ^{238}U through its first five daughters.

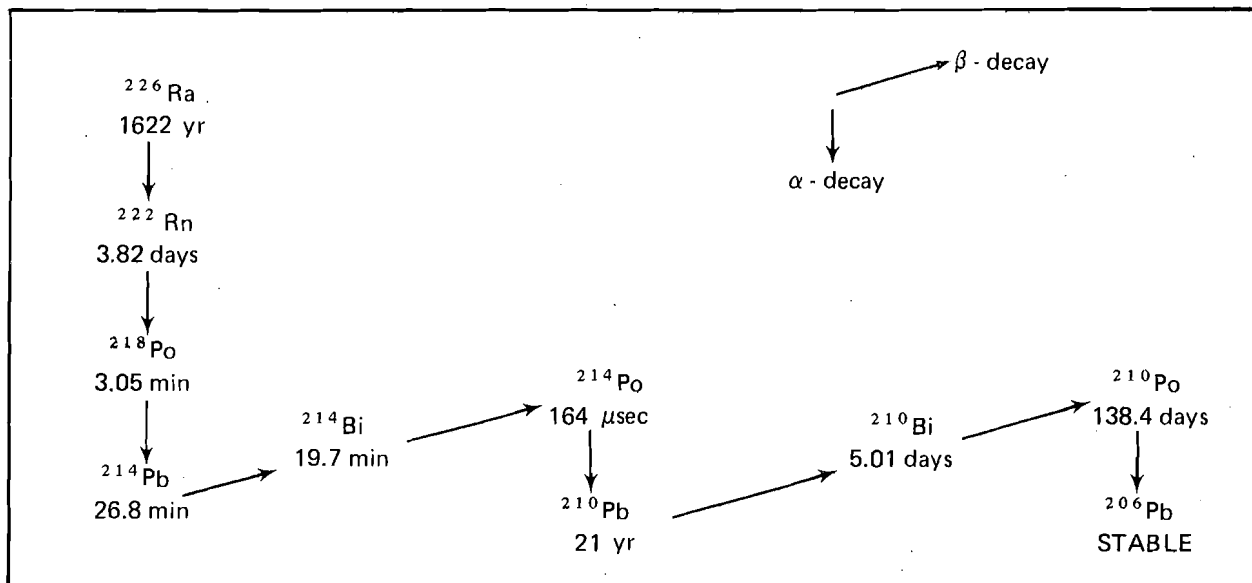


Figure 2. The radioactive decay series and half-lives of ^{238}U from ^{226}Ra to ^{206}Pb .

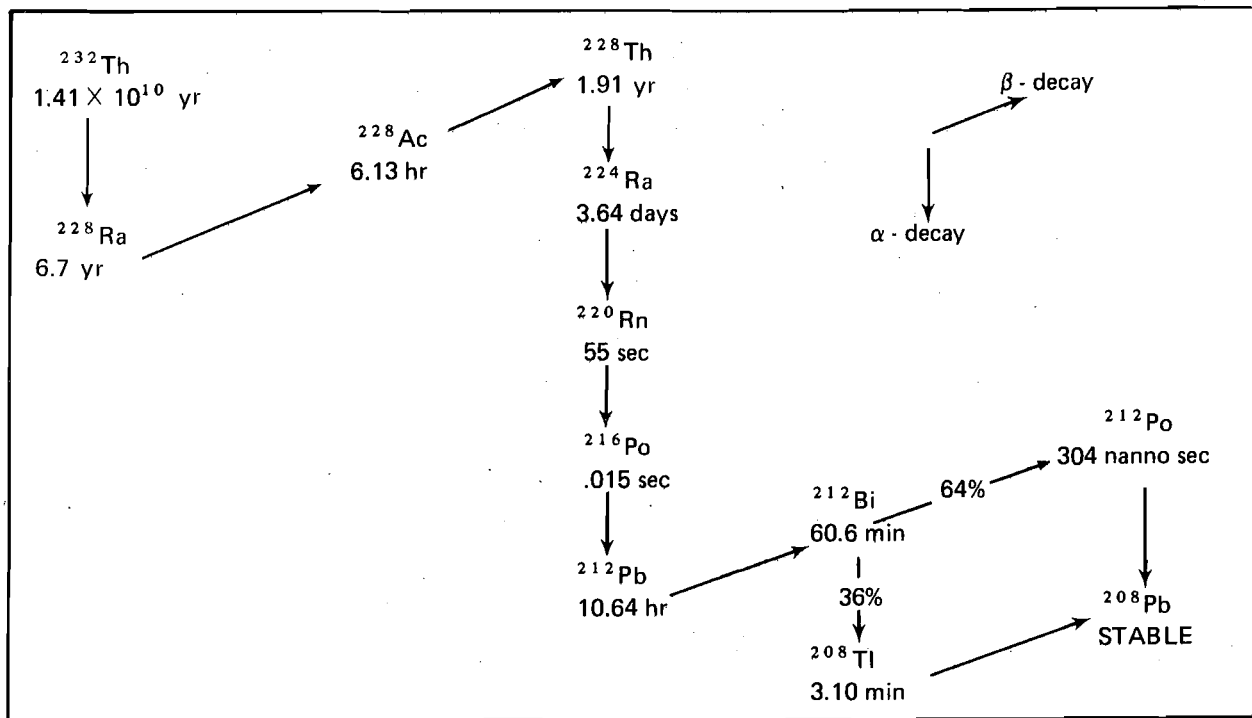


Figure 3. The radioactive decay series and half-lives of ²³²Th.

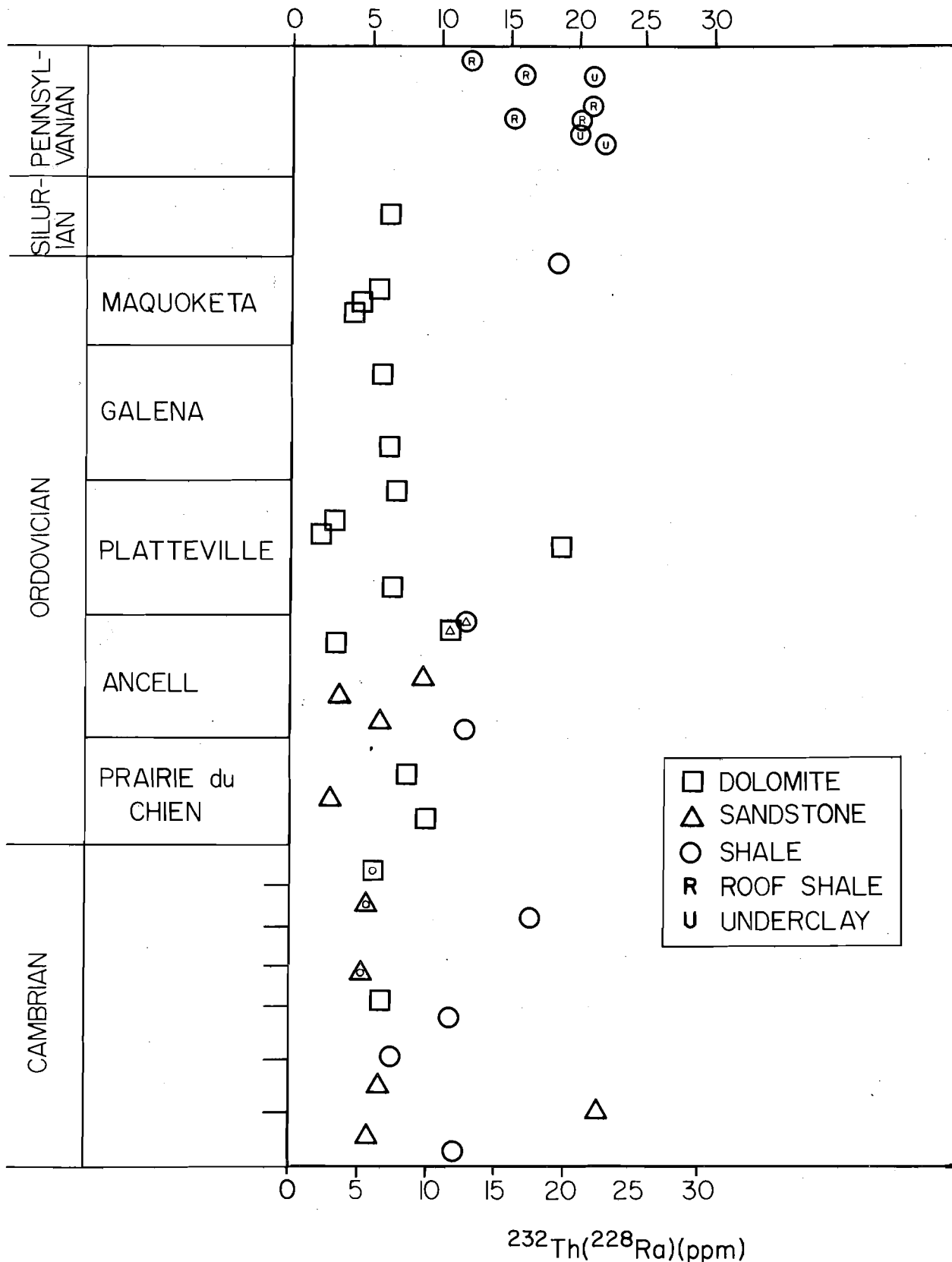


Figure 4. The concentration of ^{232}Th in sedimentary rocks of Illinois calculated from analyses for daughters of ^{228}Ra . Calculations assume daughter nuclides are in secular equilibrium.

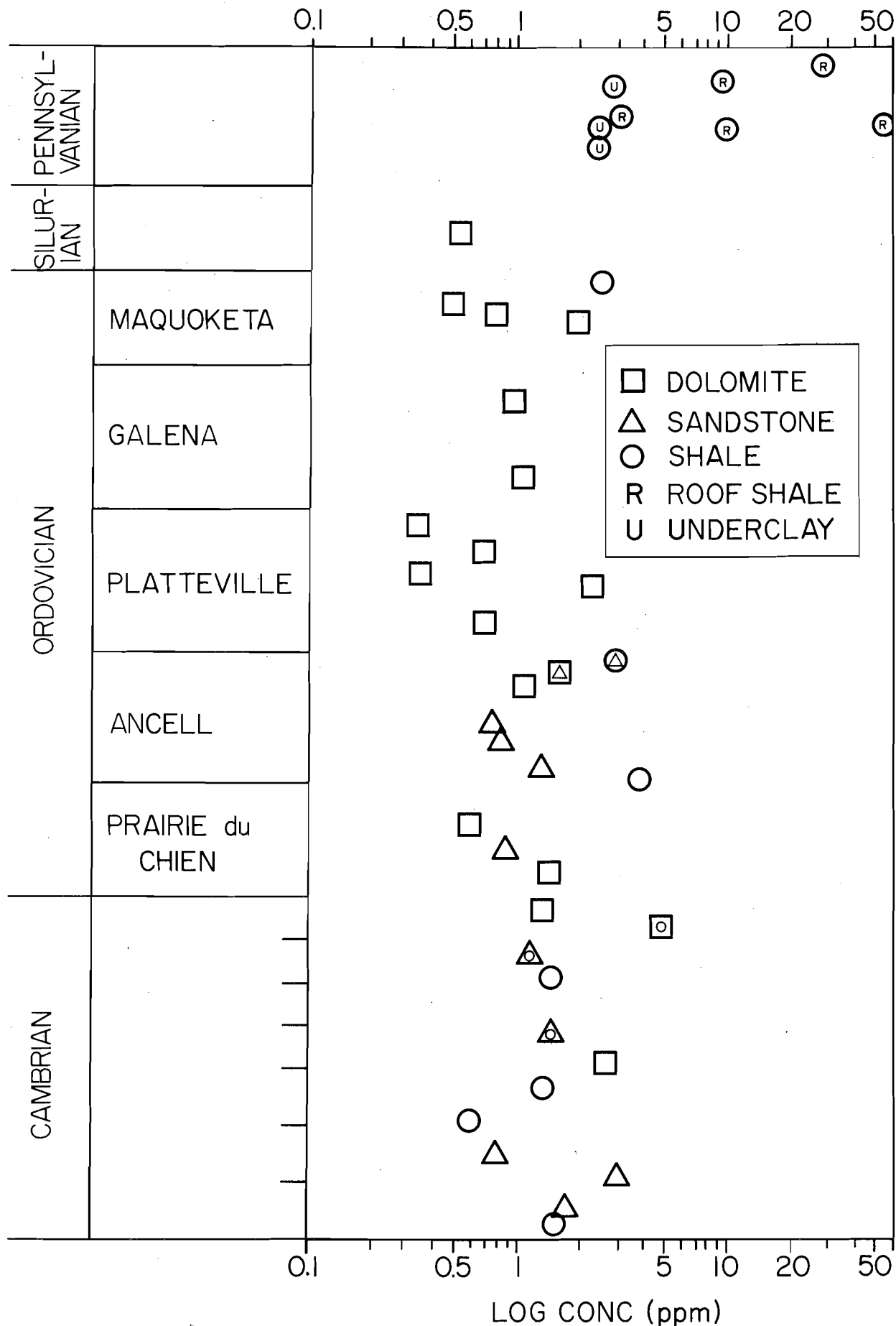
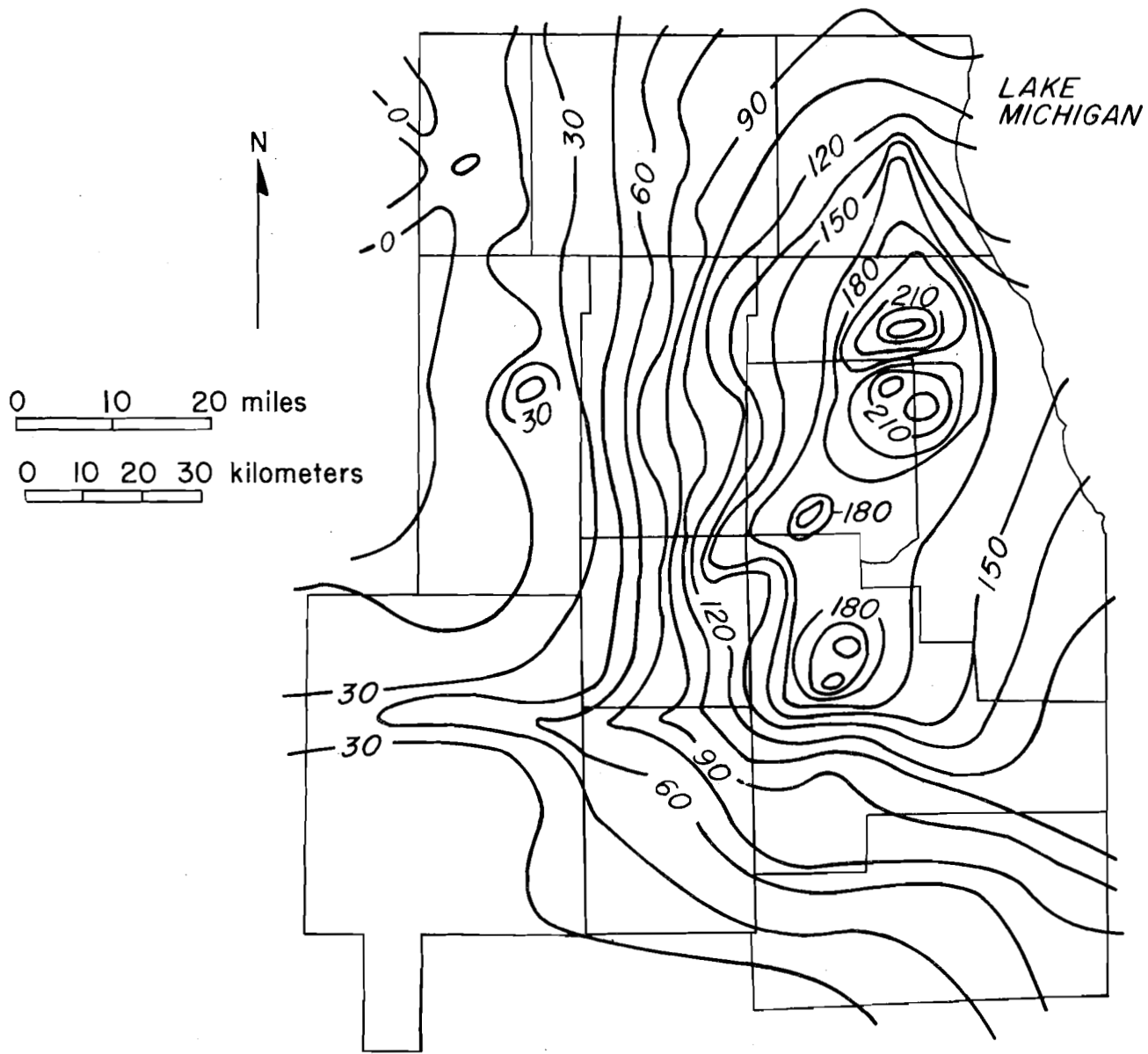


Figure 5. The concentration of ^{238}U in sedimentary rocks of Illinois calculated from analyses for daughters of ^{226}Ra . Calculations assume daughter nuclides are in secular equilibrium.



—30— Contour; interval 15 meters (50 feet)

Figure 6. The change in the piezometric surface of the Cambrian-Ordovician Aquifer in northeastern Illinois from 1864 to 1975.

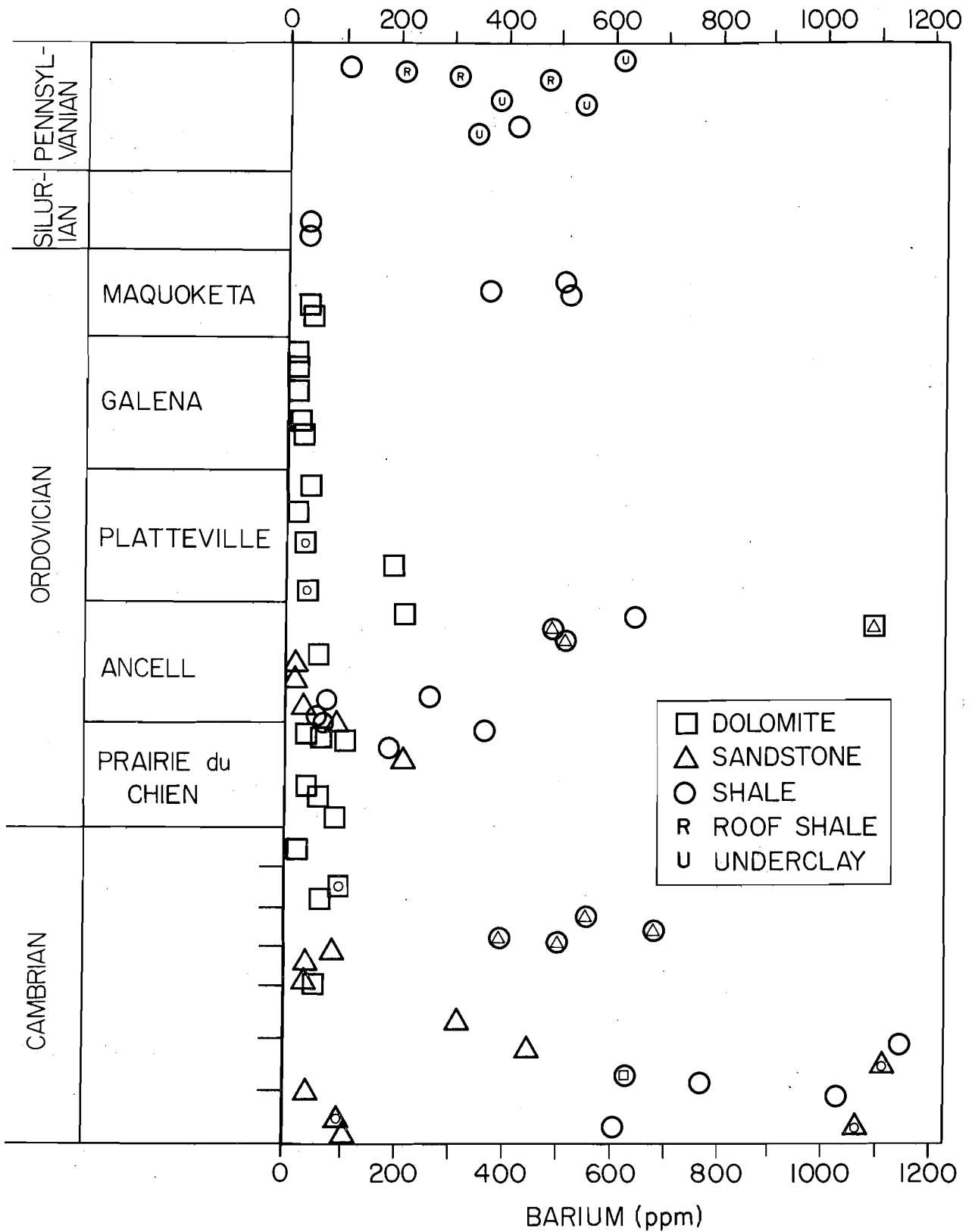


Figure 7. The concentration of barium, in parts per million, in sedimentary rocks of Illinois. Analyses by X-ray emission spectroscopy.

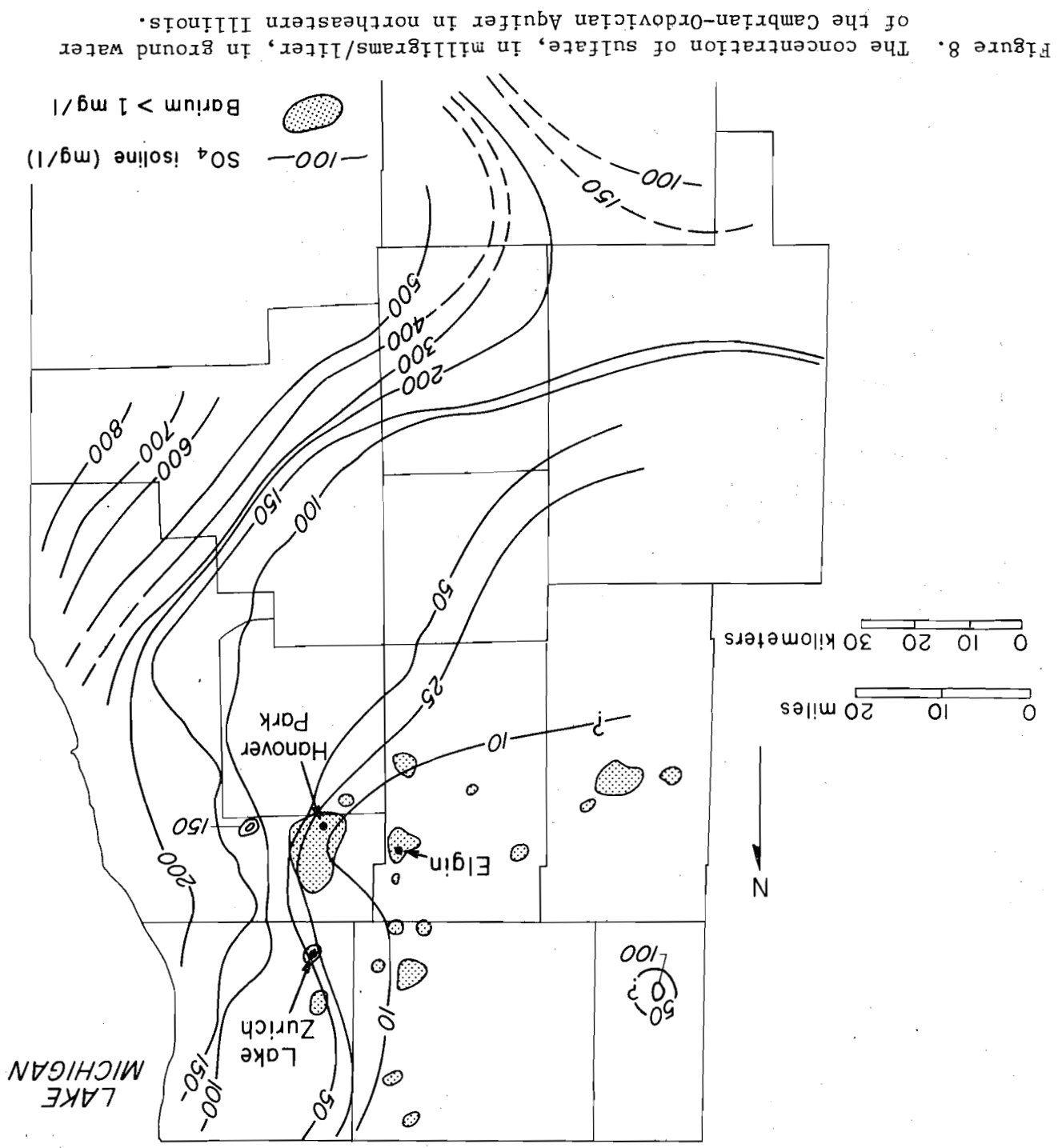


Figure 8. The concentration of sulfate, in milligrams/liter, in ground water of the Cambrian-Ordovician Aquifer in northeastern Illinois.

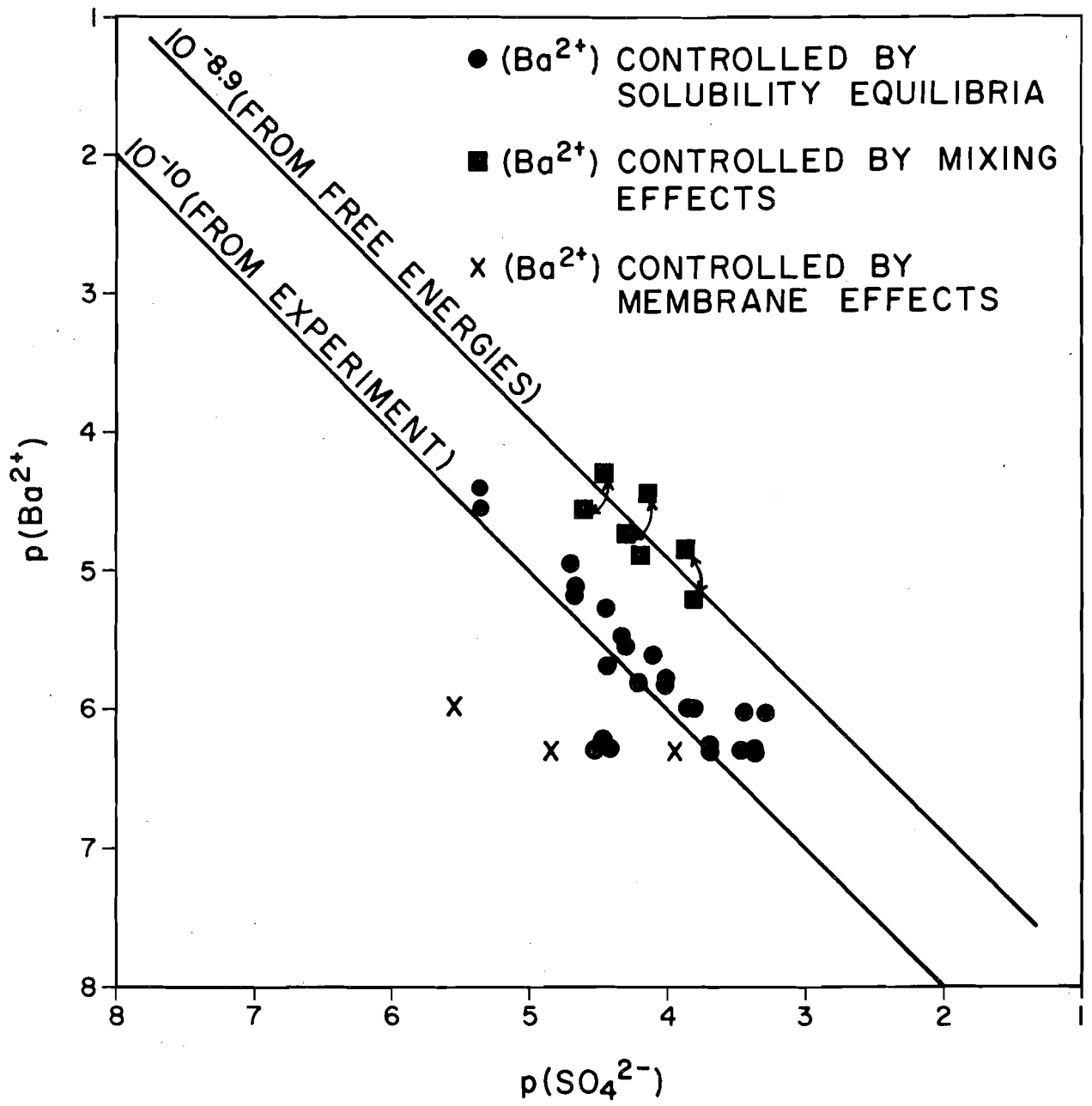


Figure 9. The relationship of (Ba²⁺) and (SO₄²⁻) activities to the solubility product of BaSO₄ in ground-water samples from the Cambrian-Ordovician Aquifer in northern Illinois. $K_{sp, BaSO_4} = 10^{-10}$ from experiment (Sillén and Martell, 1964) and $10^{-8.9}$ calculated from thermodynamic free energies (Garrels and Christ, 1965).