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RADIOACTIVITY IN GROUND AND SURFACE WATER

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ABSTRACT. Values for the uranium content and total radioactivity of some underground and surface waters are presented for samples from 77 localities, largely in the United States. The uranium content for these samples ranges between 0.02ppb and 460ppb. The uranium content and total radioactivity of water from carnotite-bearing beds in the Grand Junction, Colorado area are spectacularly high when contrasted with values for water from non-mineralized beds.

Uranium content and total radioactivity was determined from residues obtained by evaporation of water samples. The total radioactivity of these residues is shown to change through time.

INTRODUCTION

General statement.—A portion of the geochemical cycle of uranium includes the transportation of the element by water, both on and beneath the surface. Little, however, is present in the literature on the uranium content of such natural waters. For instance Kuroda (1953) in a compilation of data on the radioactivity of natural waters lists uranium values for one locality in Japan and two in Portugal. Furthermore, most references to the radioactivity of natural waters emphasize sea water, and mineral and thermal springs. For these waters values for radium, radon, thoron, and total activity expressed in radium equivalent are most common. As an exception to this, however, Adams (1954) has listed the uranium content of some surface waters in Wisconsin.

The investigations here reported are primarily concerned with the uranium carried in ground water and secondarily with that in surface water. Brief consideration is given to the nature of other radioactive elements found in these waters.

Purpose of investigation.—This study was begun to determine the amount of uranium carried in underground waters and, if possible, the nature of additional radioactive elements. The investigations were also pursued to establish whether or not the radioactivity of ground water might be useful in prospecting for radioactive deposits.

The investigation has produced some values for the uranium content of underground and surface waters from various environments, and some rudimentary speculations on other radio elements in these same waters. Furthermore it seems feasible to use the radioactivity of water in geochemical prospecting under certain conditions.

Acknowledgments.—This study is part of a general program supported by funds granted by the U. S. Atomic Energy Commission to the University of Wisconsin under University contract No. AT (11-1)-178. The project since its inception in 1951 has been under the general direction of Professor Farrington Daniels, Chairman of the Department of Chemistry, University of Wisconsin. Uranium analyses in this report were made in the analytic section of the project under the supervision of Dr. J. A. S. Adams, now of the Department of Geology, Rice Institute. Dr. Adams has also criticized this report in manuscript form. The bulk of the alpha count measurements were made in the Department of Chemistry at Wisconsin by Mr. John Ockerman, now of the Ray-O-Vac Co., Madison, Wisconsin. A few of the alpha counts were

made by Miss Joyce Bauer, also of the Wisconsin Chemistry Department. Dr. M. Starr Nichols, Assistant Director of the Wisconsin State Laboratory of Hygiene at Madison kindly provided analyses of the fluorine content of certain waters.

The writers are indebted for assistance in the collection of samples to Mr. William A. Oesterling, Alcoa Mining Company, and Mr. E. A. Brecke, Ozark-Mahoning Co., both of Roseclare, Illinois, to Dr. Henry Faul, and Mr. David Phoenix, both of the U. S. Geological Survey.

METHOD OF INVESTIGATION

Collection of samples.—Samples were collected by the authors from various underground and surface localities in Wisconsin, Illinois, Kentucky, Texas, New Mexico, and the Colorado Plateau states of Utah and Colorado.

A total of 77 water samples from various underground and surface localities are reported on in this paper. Their general distribution is given in table 1. Of these 68 were 7.6-liter (2-gallon samples), 5 were 8-liter samples and 2 were 7-liter samples. The sample was evaporated to dryness in a steel pan and the residue, usually amounting to a few grams, was removed by scraping. It is estimated that more than 95 percent of the residue obtained from each sample was recovered. In all cases evaporation was carried on within a few days of the collection of the sample, and in many instances only a few hours elapsed between collection of the sample and evaporation. The residues were later analyzed in the laboratory.

TABLE 1

General Distribution of Water Samples Discussed in this Report

Location	No. of Samples
Wisconsin	50
Southern Illinois-Western Kentucky	8
Colorado Plateau in vicinity of Grand Junction, Colorado	8
Texas Panhandle	6
Canada	3
Others	2
Total	77

At the time of collection the approximate pH of the water was determined with a colorimetric field test using phenyl red. Samples which appeared turbid were filtered before evaporation.

Alpha counts.—The radioactivity of the residues was measured in terms of alpha counts per hour using scintillation counters.[1]

A circular sample dish 2 inches (5.08 cm) in diameter contains a "thick source" of the sample. The distance between the counting surface of the sample and the phosphor screen is 0.78 cm. The counting geometry has been estimated by Ockerman as about 33 percent.

[1] The alpha scintillation counters used were of the type described by Reed (1950). Some of the counters were constructed by Mr. John Ockerman at the Department of Chemistry, University of Wisconsin. Others were commercially manufactured units purchased from the Nuclear Instruments and Chemical Corp., Berkeley, California.

Samples were counted long enough to obtain a standard deviation of less than 10 percent. Most of the alpha counts reported in tables 1-4 were obtained on residues from 6 to 9 months after evaporation of the water.

The number of alpha counts per hour of a sample is a measure of the radioactivity of the residue only at the time of counting. As discussed later the radioactivity of the sample changes with time, and it is therefore impossible to obtain a check of the alpha counts of any one sample by later counting (table 2). Furthermore, because the sample counted was a residue obtained by evaporation, any radioactive gases such as thoron or radon which may have been present in the original water sample would not be present in the residue at any time, being driven off by the process of evaporation. Despite these facts the total alpha counts give at least a relative measure of the intensity of radioactivity in the residue and hence in the water from which it was derived.

TABLE 2

Change of Total Radioactivity of Water Residue with Time

Sample No. 5-5-25. Eagle Hutson mine sump, Salem, Ky.			
U p.p.b. (water) = 0.5 p.p.b. Wt. residue = 2.7 g.			
Time (months)	½	9	29
U-eq. p.p.b.	34	13	15
U/U-eq. (%)	1½	4	3
Sample No. 9-3-1. Cactus Rat Mine seep, Colorado Plateau			
U p.p.b. (water) = 100 p.p.b. Wt. residue = 11.4 g.			
Time (months)	2	6	23
U-eq. p.p.b.	370	520	380
U/U-eq. (%)	27	19	26

Uranium content of the residue.—The uranium content of the individual samples of residue was determined under the supervision of Dr. Adams using the fluormetric method described by Grimaldi, May and Fletcher (1952). Repeated analyses were found to fall within the range of a standard error originally estimated to be between 10 and 15 percent.

Uranium content of the water.—The uranium content of the original water sample was obtained by the following formula:

$$\underset{\text{(water)}}{\text{Uppb}} = \underset{\text{(residue)}}{\text{Uppm}} \times \frac{\text{gms. residue}}{\text{gms. water evaporated}} \times 10^3$$

Total radioactivity of the water.—Uranium accounts for only a portion of the radioactivity of the original water. An attempt was made to determine the total radioactivity of the water in terms of uranium equivalent using the following formula:

$$\underset{\text{(water)}}{\text{U-equiv. ppb}} = \underset{\text{(residue)}}{\text{alpha cph}} \times 2 \times \frac{\text{gms. residue}}{\text{gms. water evaporated}} \times 10^3$$

The factor of "2" was determined empirically in the laboratory where it was found that standard water samples gave one alpha cph for 2 ppm of uranium.

The values of uranium-equivalent, however, are at best an approximation of the total radioactivity of the original water. In addition to the errors

inherent in residue recovery in alpha counting and in uranium recovery other considerations limit the precision of the final figures of uranium-equivalent. Thus radioactivity of the residue changes through time as already mentioned. and thus the alpha counts represent only a measure of the radioactivity of the residue at time of counting. Secondly, radioactive gases in the original water would be lost in evaporation and cannot be represented in the resulting residue.

Therefore the values of uranium-equivalent given for the original water sample are approximations only. The same is true of the percentage of the radioactivity of the original water due to uranium. This value is obtained by dividing the uranium content of the water by the uranium equivalent of the radioactivity of the water in ppb and multiplying by 100. Nevertheless the figures probably have a qualitative value although their degree of precision is difficult to judge.

Procedural checks and conclusions.—Procedural checks were made in the laboratory with standard and "spiked" water samples to determine (1) the proportion of residue recovered after evaporation of the water, (2) the proportion of the uranium recovered from the residue, and (3) the alpha counts produced by a given quantity of uranium in the residue. These checks showed (1) that at least 95 percent of the residue is recovered by scraping. (2) that at least 70 percent of the uranium is recovered from the residue sample, and (3) that 2 ppm of uranium yields one alpha count per hour.

On the basis of these facts and in view of the fair consistency of the values for the uranium content for different samples of waters of the same general type, it is felt that the uranium content of residue and original water as given in the tables to follow have quantitative significance. The validity of these values is further emphasized by the fact that Adams (1954) obtained values for the uranium content of Wisconsin surface waters similar to those reported in table 9. Adams obtained his data by the fluormetric analysis of water samples rather than by the analysis of residues as in this study.

The values in uranium equivalent for total radioactivity of the original water and the values for the percentage of uranium of total radioactivity in the original water are appproximations only, as previously indicated. Despite this these values are probably of the correct order of magnitude.

The data in the following sections are reported with the full appreciation of the limitation imposed by the method of the investigation here reported.

URANIUM CONTENT AND RADIOACTIVITY OF WATER

Data for 73 of the 77 water samples collected are listed in tables 3 through 9. The four samples not tabulated are discussed in the text.

Ground water from the Colorado Plateau.—Table 3 lists data on waters from aquifers in the uranium mining district of the Colorado Plateau in the vicinity of Grand Junction, Colorado. Five samples were collected from mineralized (carnotite) aquifers and three from non-mineralized aquifers. The table clearly shows that the waters from the mineralized areas are more radioactive than those from the non-mineralized aquifers. In uranium content waters from mineralized aquifers range from 100 to 460 ppb, whereas

TABLE 3
Data on Samples from Localities
on the Colorado Plateau in Colorado and Utah

Sample No.	Location	Source	Aquifer	pH	Residue				Water		
					Weight	Uranium ppm	Alpha cph	Time elapsed to count (months)	Uranium ppb	Uranium equiv. ppb	U/U-eq. (%)
			Mineralized Aquifer								
9-3-1-	Cactus Rat Mine, Thompson, Utah	mine seep	Morrison fm.	8.3	11.4	66	173	6	100	520	19
9-4-1-	Yellow Cat Camp, Thompson, Utah	spring	Morrison fm.	>8.4	4.1	468	664	6	250	720	35
9-5-1	Yellow Circle Camp, Moab, Utah	spring	Morrison fm.	>8.4	2.5	640	454	6	200	300	67
9-8-1	45-90 Mine, Uravan, Colorado	mine seep	Morrison fm.	>8.4	3.7	950	627	6	460	610	75
9-9-1	Henry Clay mine, Uravan, Colorado	mine sump	Morrison fm.	>8.4	2.0	1040	1767	6	270	930	29
			Non-mineralized Aquifer								
9-6-1	Joe Davis Canyon, Egnar, Colorado	spring	Morrison fm.	>8.4	7.4	2.1	9	6	2.0	17	12
9-7-1	Gypsum Gap, Naturita, Colorado	spring	Dakota fm.	>8.4	9.4	7.2	25	6	9.0	62	15
9-10-1	Dolores River Canyon, Gateway, Colorado	spring	Wingate fm.	>8.4	1.9	10.9	19	6	2.7	10	27

in non-mineralized aquifers the range is from 2 to 9 ppb. The total radioactivity and the percentage of radioactivity due to uranium are also significantly higher in the mineralized than in the non-mineralized zones. All waters were alkaline having pH values of 8.3 or more.

Ground water from northeastern Wisconsin.—Ground-water samples from Cambrian sandstone in northeastern Wisconsin are listed in table 4, and water from Ordovician, Silurian and late Pleistocene aquifers of the same area are listed in table 5.

The waters from the Cambrian sandstone were obtained from wells which were drilled far into the aquifer and which, in some cases, reached to the Precambrian basement. The Paleozoic rocks of the area dip to the southeast off the Wisconsin arch or dome of Precambrian rocks. Therefore the wells increase in depth toward the southeast and bottom between 200 feet and 1000 feet beneath the surface.

Compared with the Colorado Plateau waters, particularly those from mineralized aquifers, these Wisconsin waters are much lower in uranium content, ranging from 0.1 to 1.7 ppb. The total radioactivity expressed as uranium-equivalent is much lower than that of the water from the carnotite-bearing beds of the Colorado Plateau but is similar to the total radioactivity

TABLE 4

Data on Samples from Deep Wells in Northeastern Wisconsin

Aquifer in all instances, Cambrian sandstone.
Well depths range between 200 and 1000 feet and average about 800 feet.

Sample No.	Location	Well	pH	Fluorine	Residue: Weight	Residue: Uranium ppm	Residue: Alpha cph	Residue: Time elapsed to count (months)	Water: Uranium ppb	Water: Uranium equiv. ppb	Water: U/U-eq. (%)
10-1-1	Appleton	Outagamie Asylum	7.6	0.5	2.06	1.6	39	6	0.4	20	2
10-2-1	Kaukauna	City Well #4	7.4	2.0	7.90	1.6	54	6	1.7	112	1½
10-3-1	Wrightstown	Fox River Dairy	7.6	2.6	8.44	1.0	9	6	1.1	20	5
10-8-1	Point Sauble	Village Well	7.8	2.6	4.93	0.13	108	6	0.1	140	0
10-9-1	Pulaski	City Well	7.8	0.4	2.14	1.2	67	6	0.3	32	1
10-10-1	Suamico	Suamico Dairy	7.8	1.7	1.81	21.0	93	6	5.0	44	9
10-13-1	Bonduel	City Well	8.0	0.1	1.80	1.1	75	6	0.3	36	1
10-14-1	Seymour	City Well #2	7.8	0.4	2.29	1.6	46	6	0.5	33	1½
10-17-1	De Pere	East Bank Well	7.8	2.2	2.13	1.2	126	6	0.3	71	½
10-18-1	De Pere	Armour & Co.	7.8	2.2	1.79	1.5	162	6	0.4	76	½
10-19-1	Green Bay	Ashwaubenon Sanitary District	7.8	2.6	1.82	2.1	151	6	0.5	72	½
10-20-1	Green Bay	Gray Street Well	8.0	2.0	1.97	0.8	118	6	0.2	61	½
10-21-1	Green Bay	Boland Road Well	7.6	2.0	1.91	4.2	192	6	1.1	50	2
10-22-1	Preble	Deckner Ave. Well	7.8	2.8	2.69	2.4	126	6	0.9	89	1
10-23-1	Preble	Town Well #2	7.8	2.4	5.54	1.7	130	6	1.2	190	½
10-24-1	Green Bay	Northern Paper Mills	7.8	2.3	2.89	1.7	143	6	0.7	110	½
10-25-1	Hortonville	City Well	7.4	0.1	2.38	3.7	36	6	1.2	23	5
10-26-1	Marinette	Marinette Paper Mills	7.6	1.2	5.44	1.7	93	6	1.2	132	1
10-27-1	Peshtigo	Peshtigo Paper Mills	8.2	0.7	1.43	0.9	119	6	0.2	42	½
10-29-1	Lena	Village Well	7.8	0.1	1.43	1.7	102	6	0.3	38	1
10-30-1	Oconto Falls	City Well #3	8.0	0.1	1.44	1.8	91	6	0.3	34	1
10-31-1	Oconto	City Well #6	8.0	1.6	1.67	1.2	109	6	0.3	48	½
10-32-1	Coleman	Village Well	7.6	0.1	2.00	2.5	67	6	0.7	35	2

of water from non-mineralized beds of that area. The total radioactivity of the water due to uranium is low when compared to Colorado Plateau waters from both mineralized and non-mineralized aquifers.

The pH of the waters from the Wisconsin Cambrian rocks ranges between 7.4 and 8.2. There is some indication that the total amount of uranium carried in these waters decreases with increasing pH. In this regard we may note that Phair and Levine (1953) report that uranium in the form of UO_3 is preferentially leached in H_2SO_4 solutions at the expense of UO_2, Ra and Pb and further that increasing concentration of H_2SO_4 increases the proportion of UO_3 leached.

The ground water in northeastern Wisconsin is relatively high in fluorine ranging from 0.1 ppm to 2.8 ppm. The correlation between uranium and

TABLE 5

Data on Samples from Wells of Intermediate Depth in Northeastern Wisconsin. Aquifers as Indicated

Sample No.	Location	Well	pH	Residue				Water		
				Weight	Uranium ppm	Alpha cph	Time elapsed to count (months)	Uranium ppb	Uranium equiv. ppb	U/U-eq. (%)
Aquifer: Ordovician ss. Depth: 200 to 1300 feet; Average, 500 feet										
10-4-1	Denmark	Chi. & N.W. RR Depot	8.4	8.14	4.3	4	6	4.6	9	50
10-5-1	Brillion	City Well	7.6	2.15	1.8	10	6	0.5	6	8
10-6-1	Algoma	City Well #2	7.4	3.82	2.4	41	6	1.2	41	3
10-11-1	Suamico	Carl Jenkins	8.2	1.66	9.0	42	6	2.0	18	11
10-12-1	Mill Center	Walter Kimbs	8.4	0.87	0.3	6	6	0.3	1½	2
10-15-1	Kaukauna	Rapid Crocke Powerhouse	7.6	6.30	0.8	54	6	0.7	90	1
10-16-1	Little Rapids	Brown Co. Sanitarium	8.0	2.80	0.5	18	6	0.2	14	1½
Aquifer: Ordovician limestone. Depth: 30 to 150 feet.										
10-37-1	De Pere	Harry Liebergen	8.4	1.68	0.4	6	6	0.1	2½	4
10-38-1	Little Rapids	Roy Wellmer	8.2	3.19	0.8	6	6	0.3	5	6
10-47-1	Seymour	George Sieserman	7.4	2.36	0.6	4	6	0.2	2½	8
Aquifer: Silurian ls. Depth: 70 to 106 feet.										
10-7-1	Casco Junction	Green Bay and Western RR Well	7.4	2.56	4.6	35	6	1.6	24	6
10-35-1	Algoma	Louis Manly	7.8	2.56	1.5	6	6	0.5	4	13
10-36-1	Dykesville	John Collin	7.8	1.90	1.5	7	6	0.4	3½	11
10-39-1	Shirley	George Treml	7.6	7.98	1.5	16	6	1.6	33	5
Aquifer: Glacial Drift. Depth: 50 to 100 feet.										
10-40-1	Green Bay	Clarence Greatens	8.2	3.06	0.1	3	6	0.05	2½	2
10-42-1	Pound	Leo Cudnoski	8.4	1.59	6.5	24	6	1.4	10	14
10-43-1	Oconto Falls	Walter Birr	7.6	2.09	0.6	8	6	0.2	4½	4
10-45-1	Embarrass	Anthony Anton	8.4	0.75	0.4	11	6	0.05	2	2½
Aquifer: fractured granite and glacial drift. Depth: 350 feet.										
10-46-1	Embarrass	Wedig Krubsach	8.0	1.42	10.0	29	6	1.9	11	17

fluorine is not marked, but in general the total radioactivity of the water in uranium-equivalent increases with an increase in the fluorine content of the water. The fluorine concentration is greatest in the deeper wells down the dip of the Cambrian sandstone with the highest values recorded along the Fox River Valley between Lake Winnebago and Green Bay.

The results of water measurements from shallower and stratigraphically higher aquifers than the Cambrian in northeastern Wisconsin are given in table 5. The aquifers are Ordovician sandstone and dolomite, Silurian dolo-

mite, and Pleistocene (Wisconsin) gravel. The waters from these aquifers are similar in uranium content to that from the Cambrian, but their total radioactivity is not so great. The proportion of radioactivity due to uranium is variable but averages about 5 or 10 percent.

Ground water from the Texas High Plains.—Table 6 lists the data on five water samples from the Ogallalah formation (Pliocene) of the High Plains of Texas. Their uranium content, 2 to 10 ppb, is intermediate between that of the Wisconsin waters and the waters from mineralized waters of the Colorado Plateau. They are similar in uranium content to the waters from the non-mineralized aquifers of the Colorado Plateau. Their total radioactivity is small but the percentage of activity due to uranium is between 25 and 63, similar to the water from the carnotite-bearing aquifers of Colorado and Utah. The source of uranium in these samples is thought to be volcanic ash which occurs in Pleistocene beds above the Ogallalah and probably in the Ogallalah itself. Ground-water recharge of the Ogallalah is local.

TABLE 6
Data on Samples from the Texas Panhandle
Aquifer: Ogallalah fm. (Pliocene)

Sample No.	Location	Source	ph	Residue				Water		
				Weight	Uranium ppm	Alpha cph	Time elapsed to count (months)	Uranium ppb	Uranium equiv. ppb	U/U-eq. (%)
8-3-3	Exell, Texas	Stock well	—	1.06	19	36	8	2.7	10	27
8-4-1	Exell, Texas	Stock well	8.1	1.11	16	30	8	2.3	9	25
8-7-1	Dawn, Texas	House well	—	2.08	25	44	8	6.8	24	27
8-7-3	Hereford, Texas	Irrigation well	—	1.56	21	20	8	4.3	8	54
8-8-3	Tule Canyon, Texas	House well	—	3.03	25	20	8	10.0	16	63

Ground water from southern Illinois-western Kentucky.—The results for seven water samples from the southern Illinois and western Kentucky Fluospar district are listed in table 7. Six of these samples were collected from mine workings, and one from an artesian spring. The uranium content is uniformly low, having values between 0.13 and 0.5 ppb.

Miscellaneous ground water samples.—A sample from the warm spring at Pagosa Springs, Colorado, and a sample from an artesian well at Newkirk, New Mexico, were collected. The results are given in table 8.

Surface waters.—Waters from surface streams were collected at nine localities, seven of them in Wisconsin. Data on these waters are given in table 9. The uranium content is generally low as is the total radioactivity in terms of uranium equivalent. The percentage of radioactivity due to uranium tends to be high.

Waters not tabulated.—The data for four additional samples are not included in tabular form. One of these was collected from a spring at the

TABLE 7
Data on Samples from Southern Illinois and Western Kentucky
Aquifer: Mississippian Limestone

Sample No.	Location	Source	ph	Residue				Water		
				Weight	Uranium ppm	Alpha cph	Time elapsed to count (months)	Uranium ppb	Uranium equiv. ppb	U/U-eq. (%)
5-3-2	Rosiclare, Ill.	Rosiclare Fluorspar Mine	7.8	1.61	1.2	10	9	0.25	4	6
5-6-9	Cave-in-Rock, Ill.	W. Green Fluorspar Mine	7.2	12.55*	1.1	4	9	0.18	13	1
5-7-1	Cave-in-Rock, Ill.	Deardorf Fluorspar Mine	8.2	2.84*	1.1	10	9	0.4	8	5
5-8-1	Rosiclare, Ill.	Alcoa Fluorspar Mine	8.3	2.81	1.9	13	9	0.7	10	7
5-5-25	Salem, Kentucky	Eagle Hutson Zinc Mine	8.1	2.70*	1.4	19	9	0.5	14	4
5-4-1	Cave-in-Rock, Ill.	"Rd. Spring" (artesian)	7.4	3.38*	1.4	68	9	0.6	58	1

* 8 liter sample.

Fission Mines, Wilberforce, Ontario; another from a pond in a pegmatite quarry, Madawaska, Ontario. The third, an 8-liter sample, was supplied by Mr. Alan Gregory from Beaverlodge Lake in the Lake Athabaska Area, Saskatchewan. Each of the samples was chosen because of proximity to a uraniferous source. In each instance, however, the amount of residue recovered was so small that the analytical results were not judged to be reliable. A fourth sample of water from Cambrian sandstone at Madison, Wisconsin is referred to later.

TABLE 8
Data on Miscellaneous Samples

Sample No.	Location	Source	ph	Residue				Water		
				Weight	Uranium ppm	Alpha cph	Time elapsed to count (months)	Uranium ppb	Uranium equiv. ppb	U/U-eq. (%)
8-9-1	Newkirk, N. M.	Housewell—	>8.4	9.88	1.7	4	8	2.2	10	22
9-1-1	Pagosa Springs, Colo.	(Triassic ss) Warm Spring	7.0	19.10	1.7	33	8	4.6	180	3

Conclusions.—On the basis of the data presented above the following conclusions can be drawn:

1. Both underground water and surface water carry uranium in measurable amounts. It cannot be said whether all of this uranium is in solution or whether it is in suspension associated in some way with colloidal-size particles.

2. Where underground waters pass through an aquifer containing carnotite, the total radioactivity and uranium content are high when compared with water from similar but non-mineralized aquifers. It cannot be said on the basis of the data collected whether the same relationships would hold for aquifers containing uranium-bearing minerals other than carnotite. On the other hand, the relatively high uranium content and radioactivity of waters from carnotite-contaminated beds suggests that geochemical methods might well be devised for prospecting for uraniferous deposits.

3. The residue technique used in this study appears to be practical only for those waters which carry much material in solution and thus produce a relatively large residue sample, say 1 gram per 2-gallon water sample.

4. The relation between pH and total uranium carried is not clear from the data at hand. Waters from Cambrian rocks in northeastern Wisconsin, however, decrease in uranium content as the pH increases from 7.4 to 8.0.

5. Incomplete data obtained from water in Cambrian rocks of northeastern Wisconsin also suggest that in these waters fluorine increases with increasing radioactivity but not necessarily with uranium.

CHANGES IN RADIOACTIVITY WITH TIME

It has already been stated that the alpha counts per hour for any one residue sample change with time. The effect of this change on the calculations of total radioactivity of the water in uranium-equivalent and of the percentage of radioactivity due to uranium has been illustrated in table 2. Successive

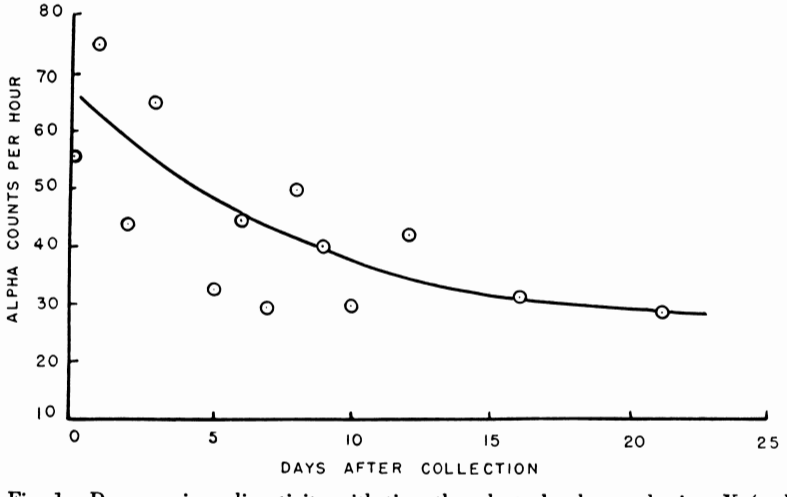

Fig. 1. Decrease in radioactivity with time thought to be due to thorium X (radium 224 in thorium series). Sample is a residue of water from the Cambrian sandstone at Madison, Wisconsin, taken at the main station well. The sample counted at successive intervals. Large scatter of points due largely to short counting periods.

recounts of a single sample showed that the change of radioactivity was greatest and least predictable in the first few months after evaporation. Samples counted 20 or more months after evaporation were found to be changing at a slower rate.

An excellent example of decreasing radioactivity in terms of alpha counts per hour is provided by a sample of residue obtained from the evaporation of water from a deep well in Cambrian sandstone at Madison, Wisconsin. The sample was counted repeatedly during the three-week period immediately following evaporation of the water. The activity decreased by one-half during this interval as shown in figure 1.

Table 10 lists 25 water samples for which the residue of each was counted two or more times at varying intervals. The data are not complete enough to warrant any definite conclusions or generalizations except (1) that the changes are real, (2) the radioactivity in terms of alpha counts may either increase or decrease with time, and (3) the direction of change appears to reverse itself in some instances.

Members of the 4n + 1 series cannot be cited as responsible for the change because they are not known to occur in nature. The actinium series is exceedingly rare in nature so must be excluded from consideration. This leaves members of the uranium and thorium series. Of these, elements with long half lives such as Uranium 238 and 234 and Thorium 232 and 230 disintegrate at too slow a rate to be reflected over the short time span of a few weeks, or months, or at most something over two years during which

TABLE 9
Data on Surface Waters

Sample No.	Location	Source	pH	Residue				Water		
				Weight	Uranium ppm	Alpha cph	Time elapsed to count (months)	Uranium ppb	Uranium equiv. ppb	U/U-eq. (%)
5-7-3	Cave-in-Rock, Ill.	Deardorf Creek	7.2	3.09	0.2	4	9	0.13	3	4
5-2-4	Rosiclare, Ill.	Ohio River	7.8	2.65	1.1	9	9	0.5	6	9
8-6-1	N. of Amarillo, Texas	Canadian River	>8.4	4.67	5.7	11	0	3.5	14	25
10-28-1	Middle Inlet, Wis.	Granite Quarry Seep*	>8.4	1.50	4.3	8	6	0.85	3	28
10-33-1	Mountain, Wis.	Oconto River	>8.4	2.62	3.8	6	6	1.3	4	33
10-34-1	Clintonville, Wis.	Pigeon River	>8.4	1.41	16.0	16	6	3.0	6	50
10-44-1	Shawano, Wis.	Wolf River	>8.4	2.64	1.2	2	6	0.4	1½	27
10-41-1	Green Bay, Wis.	Fox River	7.8	1.22	2.9	5	6	0.5	1½	33
10-48-1	Appleton, Wis.	Fox River	>8.4	1.06	3.7	4	6	0.5	1	50
10-49-1	Madison, Wis.	Lake Mendota	>8.4	2.86	1.1	1	1	0.4	¾	50

* Water passes through calcareous drift of Wisconsin age before seeping through granite.

TABLE 10
Change in Radioactivity of Water Residue with Time in Terms of Alpha Counts per Hour

Sample No.	Location	Time elapsed to count (months)	Alpha cph	Time elapsed to count (months)	Alpha cph	Time elapsed to count (months)	Alpha cph	Time elapsed to count (months)	Alpha cph
5-3-2	Rosiclare, Ill.			7	10	9	10		
5-4-1	Cave-in-Rock, Ill.			7	121	9	68	18	48
5-5-25	Salem, Ky.	½	51			9	19	29	22
5-6-9	Cave-in-Rock, Ill.	½	5			9	4		
5-7-1	Cave-in-Rock, Ill.	½	20			9	10		
5-8-1	Rosiclare, Ill.			7	18	9	13		
8-3-3	Exell, Texas	0	98	8	36				
8-4-1	Exell, Texas	0	56	8	30			23	20
8-7-1	Dawn, Texas	0	50	8	44				
8-7-3	Hereford, Texas			8	20	27	25	29	26
8-8-3	Tule Canyon, Texas			8	20			29	32
8-9-1	Newkirk, N. M.			8	5	17	5		
9-1-1	Pagosa Springs, Colo.			8	33	17	31	29	44
9-3-1	Thompson, Utah	2	123	6	173			23	126
9-4-1	Thompson, Utah	2	638	6	664				
9-5-1	Moab, Utah	2	340	6	454			23	390
9-8-1	Uravan, Colo.	2	571	6	627				
9-9-1	Uravan, Colo.	2	1729	6	1767				
9-6-1	Egnar, Colo.	2	4	6	9				
9-7-1	Naturita, Colo.	2	18	6	25				
9-10-1	Gateway, Colo.	2	14	6	19				
10-10-1	Suamico, Wis.			7	108			23	41
10-14-1	Seymour, Wis.			7	50			23	44
10-18-1	De Pere, Wis.			7	162	15	168		
10-21-1	Green Bay, Wis.			7	192	15	147		

successive counts were made. The cause of the change then must lie among the members of the uranium and thorium series with shorter half lives. These would include those elements with half lives of Thorium X (radium 224) or less, but exclusive of radon and thoron which were not present in the original residue sample. Furthermore, we may exclude those elements of half lives less than three or four days because of the impracticability of measuring them by the alpha count method.

A series of activity curves for members of the uranium and thorium series was constructed. Theoretically the comparison of these curves with the changes in alpha activity of the water residues through time should permit speculations concerning the relative importance of some of the daughter elements of uranium and thorium. Unfortunately, data are insufficient for most samples to allow any precise plotting of the nature of the change of alpha activity through time. Only in the case of a sample obtained from waters in the Cambrian sandstone at Madison, Wisconsin are enough alpha determinations available to sketch the change in alpha activity through time. This

sample was counted at successive intervals over a three-week period and the results given in figure 1. It is presumed that the rapid decrease in activity during the first two weeks was due primarily to the presence of thorium X (radium 224 in the thorium series).

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