

BEFORE THE POLLUTION CONTROL BOARD
OF THE STATE OF ILLINOIS

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NOV 24 2004

STATE OF ILLINOIS
Pollution Control Board

IN THE MATTER OF:)
)
REVISIONS TO RADIUM WATER)
QUALITY STANDARDS: PROPOSED) R04-21
NEW 35 ILL. ADMIN. CODE 302.307) Rulemaking - Water
AND AMENDMENTS TO 35 ILL. ADMIN.)
CODE 302.207 AND 302.525)


NOTICE OF FILING

To: See Attached Service List

Please take notice that on November 24, 2004, we filed with the Office of the Clerk of the Illinois Pollution Control Board an original and ten copies of the attached ***WATER REMEDIATION TECHNOLOGY LLC'S RESPONSE TO DOCUMENTS REQUESTED DURING FOURTH PUBLIC HEARING***, a copy of which is served upon you.

Respectfully submitted,

WATER REMEDIATION TECHNOLOGY, LLC

By: 
One of its Attorneys

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THIS FILING IS BEING SUBMITTED ON RECYCLED PAPER

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CODE 302.207 AND 302.525)

**WATER REMEDIATION TECHNOLOGY LLC'S RESPONSE
TO DOCUMENTS REQUESTED DURING FOURTH PUBLIC HEARING**

Water Remediation Technology LLC ("WRT") through its attorneys, submits the documents requested to be submitted by WRT during the October 21 and 22, 2004, public hearings in this proceeding. A summary of these requests and documents follows:

REQUEST ONE: Illinois Environmental Protection Agency ("IEPA") requested references for the statement by Mr. Theodore G. Adams concerning 1 pCi/L as representing background for total radium in surface waters. The following documents are responsive to this request:

- Agency for Toxic Substances and Disease Registry ("ATSDR") toxicity summary on radium. (See October 22 Hearing Exhibit 16.)
- C. T. Hess, J. Michel, T. R. Horton, H. M. Prichard and W. A. Coniglio, The Occurrence of Radioactivity in Public Water Supplies in the United States, Health Physics Vol. 48, No. 5 (May), pp. 553-586 (1985). (See Attachment A hereto.)
- Jacqueline Michel and C. Richard Cothorn, Predicting the Occurrence of ²²⁸Ra in Ground Water, Health Physics Vol. 51, No. 6 (December), pp. 715-721 (1986). (See Attachment A.)

REQUEST TWO: Counsel for the City of Joliet requested a copy of the permit applications submitted to the Illinois Department of Nuclear Safety (“IDNS”) for the Village of Oswego. Copies of those permit applications (which were marked as Exhibit 17 during the October 22 hearing) were previously submitted to counsel for IEPA and the City of Joliet. (See October 22 Hearing Exhibit 17.)

REQUEST THREE: Counsel for the City of Joliet requested copies of WRT contract with the Village of Oswego and/or the Village of Elburn. WRT has been informed by the Village of Oswego that it has provided the requested contract to counsel for the City of Joliet.

Respectfully submitted,

WATER REMEDIATION TECHNOLOGY, LLC

By: _____

One of its Attorneys

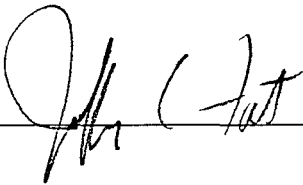
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CERTIFICATE OF SERVICE

The undersigned, an attorney, certifies that he/she has served upon the individuals named on the attached Notice of Filing true and correct copies of *WATER REMEDIATION TECHNOLOGY LLC'S RESPONSE TO DOCUMENTS REQUESTED DURING FOURTH PUBLIC HEARING* by First Class Mail, postage prepaid, on November 24, 2004.



SERVICE LIST

R04-21

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Exhibit A

THE OCCURRENCE OF RADIOACTIVITY IN PUBLIC WATER SUPPLIES IN THE UNITED STATES†

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Abstract—Examination of the collected data for radionuclide concentration measurements in public water supplies in the United States show more than 51,000 measurements for gross α -particle activity and/or Ra, 89,900 measurements for U, and 9,000 measurements for Rn. These measurements were made as part of national and state surveys of radionuclide concentrations in utility water supplies for Ra and Rn; and the National Uranium Resource Evolution (NURE) survey for U which included non-utility water supplies.

Surface water has low values for Ra and Rn but levels comparable to ground water for U. Separate isotope measurements were not taken for much of the Ra and U data. Because ^{226}Ra to ^{228}Ra ratios and ^{238}U to ^{234}U ratios are not fixed in water, further measurements are needed to establish the specific isotopic concentrations by region. Analysis of the state average values in geological provinces shows the highest provincial areas for Ra are the Upper Coastal Plain, the glaciated Central Platform, and the Colorado plateau. For U, the highest areas are the Colorado plateau, the West Central Platform, and the Rocky Mountains. For Rn, the highest provinces are New England and the Appalachian Highlands-Piedmont. Regional hydrogeological and geochemical models are suggested for guiding the formulation of regional standards and monitoring strategies. Utility supplies serving small populations have the highest concentration for each radionuclide and have the lowest fraction of samples measured, which shows a need for further measurements of these small population water supplies. Risk estimates for the average concentration of Ra in utility ground water give about 941 fatal cancers per 70.7-yr lifetime in the United States. Risk estimates for the average concentration of U in utility surface and ground water give about 105 fatal cancers per 70.7-yr lifetime in the United States. Using 1 pCi/liter in air for 10,000 pCi/l in water, the Rn in utility water risk estimate is for 4,400–22,000 fatal cancers per 70.7-yr lifetime in the United States.

† This paper was prepared for and completed at the National Workshop for Radioactivity in Drinking Water under the sponsorship of the Office of Drinking Water, U.S. Environmental Protection Agency. The workshop was held in Easton, MD, 24–26 May 1983.

PREFACE

THE sections of this paper are arranged in the order of introduction, geochemistry and occurrence. A central theme of the report is that geological setting strongly influences the occur-

rence of natural radionuclides in drinking water. The observed concentrations of U, Ra and Rn in ground and surface water can be related to the rock types and the amount and distribution of U and Th in the materials which constitute the aquifer and surficial deposits. The United

States can be divided into 11 geological provinces, each of which is characterized by dominant types of rocks or deposits as well as ground-water flow systems, discussed in Table 1 and shown in Fig. 1 (Be81; Sc62). These provinces are discussed in all sections of the

Table 1. Summary of potential host rocks, geologic framework and nature of ground-water flow systems in the provinces of the coterminous United States (Fig. 1)

PROVINCE	GEOLOGICAL FRAMEWORK	NATURE OF GROUND-WATER FLOW SYSTEMS
1. New England. Adirondack Mountains	New England--complexly faulted metamorphic and metasedimentary rocks intruded by large masses of granite. Adirondacks--mountains composed of marble and schist intruded by granites, anorthosite, and gabbro.	Flow and head in bedrock of low permeability and overlying glacial aquifers greatly influenced by local topography and surface-water features.
2. Appalachian Highlands. Piedmont	Appalachian Highlands--mountain belt of granites and metamorphics thrust westward over Paleozoic rocks. Piedmont--non-mountianous belt of highly complex metamorphic rocks with abundant granites.	Flow and head in metamorphic and granite bedrock of low permeability, largely controlled by topography and surface water features; folded limestone locally cavernous and highly permeable at shallow depths supporting large springs; sandstone aquifers of moderate extent and permeability; flow systems generally related to local recharge in interstream areas and discharge to surface-water features.
3. Appalachian and Interior Plateaus	Appalachian and Interior Plateaus consist of gently dipping, gently folded sandstones, shales, carbonates, and evaporites. In southern Missouri exposes old crystalline rocks.	Regional flow in low-to-moderately permeable sandstones and carbonates; carbonates locally of high permeability at shallow depth due to fractures and solution channels support large springs.
4. Coastal Plain	Seaward dipping thickening wedge of sand, sandstones and shales with some evaporites and limestones; underlain by a basement of metamorphic rocks.	Regional flow in sand and limestone aquifers with intervening clay confining layers; predominant flow direction seaward; discharge upward through confining layers and to streams.
5. Glaciated Central Platform	Igneous and metamorphic rocks on the northwest overlain by sandstones, carbonates, shales, and evaporites; deep basin deposits in Michigan and Illinois.	Regional flow in sandstone and carbonate aquifers; highly mineralized water at depth in basins; glacial aquifers locally overlie bedrock.
6. Western Central Platform	Horizontal to gently dipping sandstones; deep sedimentary basins and structural high. Capped with sands and gravels.	Regional flow in layered sandstone and carbonate aquifers; thick confining beds of shale; deep basins contain highly saline water. Extensive fluvial deposit aquifers Nebraska south into Texas and glacial aquifers in North Dakota and South Dakota overlie sandstones.

Table 1. (Contd.)

PROVINCE	GEOLOGICAL FRAMEWORK	NATURE OF GROUND-WATER FLOW SYSTEMS
7. Rocky Mountain System	Igneous and metamorphic folded core rocks of Rocky Mountains and intermontane basins of shales, carbonates, evaporites, and sandstones. Intrusive and volcanic rocks.	Regional flow in layered sandstone and carbonate aquifers with shale confining beds in intermontane basins; local recharge and discharge controlled by topography and surface water features in fractured igneous and metamorphic rocks.
8. Colorado Plateaus	Flat-lying to gently warped layers of sandstones, shales, limestones and evaporites with volcanic rocks.	Regional flow in layered sedimentary rocks; chief aquifers are sandstones and carbonates; discharge to major streams; highly saline water at depth in deep basins and in association with salt beds.
9. Basin and Range	Elongated blocky mountains of faulted rock complexes; deep alluvium-filled intermontane basins; intrusive igneous stocks and plugs; extrusive ash-flow tuffs, rhyolites, and basalts.	Flow within closed basins; inter-basin flow between closed topographic basins through permeable bedrock; interbasin flow in alluvial channels between basins with integrated surface drainage; deep regional flow systems in carbonate and volcanic rocks.
10. Columbia Plateaus	Regional shallow structural basin of basaltic lava flows; locally faulted and folded; mountain range on the west consisting of elongated chain of andesitic volcanic cones.	Basaltic lava flows range from highly permeable to dense nearly impermeable, creating regional aquifers with perched aquifers separated by confining beds. The ground water principally discharges to the major streams; locally discharges to a few closed basins.
11. Pacific Mountain System	Consists of several complex elements: Large uplifted and tilted blocks of granite with inliers of metasediments; folded and faulted sedimentary rocks; deep elongated troughs filled with fluvial sediments. with fluvial sediments.	Regional flow in deep intermontane sedimentary basins; igneous and metamorphic rocks of low permeability support shallow local flow systems related to topography and surface drainage.

report and provide a framework for understanding the variations in the distribution and activities of natural radionuclides in water. In fact, one hypothesis is that certain provinces or sub-provinces can be characterized as producing ground water with specific radionuclide problems, or conversely, without specific radionuclide problems. If this hypothesis can be verified, it has important applications to the development of regional guidelines for monitoring requirements in the revised regulations (La83).

OCCURRENCE OF Ra ISOTOPES IN PUBLIC DRINKING WATER

Introduction

Radium has two natural isotopes which are of concern in public water supplies. Radium-226 is generated through decay of ^{238}U and is an α emitter with a $t_{1/2} = 1,622$ yr. This is the isotope which is commonly referred to as Ra and has been measured in many water supplies. The other isotope, ^{228}Ra , is generated directly by ^{232}Th decay and is a shorter-lived, weak β

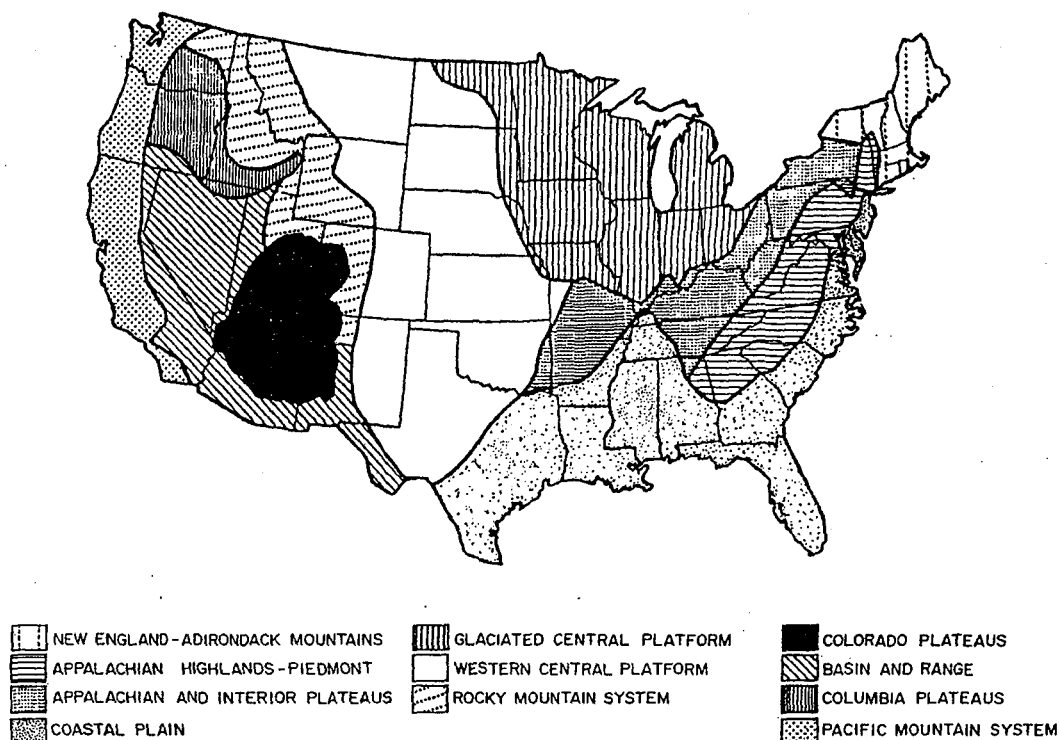


FIG. 1. Geological provinces of the United States, according to Beddinger (Be81).

emitter ($t_{1/2} = 5.7$ yr). There is a third isotope of Ra which is of possible concern, ^{224}Ra , with a $t_{1/2} = 3.64$ days. Its occurrence is not well known; only a few data are available from samples at the well head. The U.S. Environmental Protection Agency (EPA) established interim regulations in 1976 for maximum levels of radioactivity in drinking water as follows:

“Maximum contaminant levels (MCL) of combined Ra-226 and Ra-228—5 picocuries per liter (pCi/l); gross alpha-particle activity—15 pCi/l excluding radon and uranium” (Ep76a).

These MCLs were set under the authority of the Safe Drinking Water Act to protect health, taking treatment costs in consideration. In an effort to minimize the costs of analysis and monitoring, the EPA established a series of screening steps to test for compliance with the interim regulations. These criteria stated that

when the average gross α -particle activity of four quarterly samples or composites exceeds 5 pCi/l, the same or equivalent sample shall be analyzed for ^{226}Ra . If the activity of ^{226}Ra exceeds 3.0 pCi/l, the sample shall be analyzed for ^{228}Ra . Inherent in these regulations were the assumptions that ^{226}Ra was to be the dominant radioactive contaminant in drinking water and the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio was less than 1.0. The regulations required all systems supplying 25 or more people to be monitored every 4 yr.

Since the interim regulations were established, much more information on the occurrence of Ra isotopes is now available from state compliance data and from detailed studies on the correlation and interrelationships of ^{228}Ra and ^{226}Ra in ground water with specific geological provinces (Mi80; As81; Ki82; Mi82; Kris82). In light of these new data, the key issues to be considered for revision of the regulations are:

(1) prioritization of specific areas for monitoring for ^{228}Ra and ^{226}Ra ;

(2) reduction in the interval frequency or complete omission for specific areas for repeat monitoring; and

(3) decoupling ^{228}Ra analysis from ^{226}Ra , with criteria for when ^{228}Ra is to be measured.

The purpose of this paper is to concisely review the existing information on the geochemistry and occurrence of ^{228}Ra and ^{226}Ra , and to provide guidelines for regulatory revision.

Geochemistry of Ra isotopes

The distribution of ^{228}Ra and ^{226}Ra in water is a function of the Th and U content of the aquifer, the geochemical setting of the aquifer solids, and the $t_{1/2}$ of each isotope. There are specific geological and chemical processes that control the Th and U content in aquifers, which are discussed in detail by Olson and Overstreet (Ol64), Cherdynstev (Ch71) and Gableman (Ga77). In fact, Th and U have very similar behavior, with one important exception which is most responsible for their eventual separation. Thorium has one oxidation state and is immobile at low temperatures. Therefore, Th distribution is controlled by primary geochemical processes (such as magmatic crystallization) or secondary physical processes (such as sedimentary enrichment in placer deposits). Uranium has two oxidation states and the +6 state (uranyl) can form highly soluble complexes which can be transported long distances by oxidizing ground water before being removed by adsorption or reduction to the +4 state. The estimated average crustal Th/U activity ratio is 1.2–1.5 so that, in the absence of enrichment or depletion processes, ^{228}Ra activity should be higher than ^{226}Ra . However, the tendency for U enrichment under certain geochemical conditions results in regions of higher ^{226}Ra , thus the EPA's decision to emphasize ^{226}Ra in the interim regulations.

Radium enters ground water by dissolution of aquifer solids; by direct recoil across the liquid-solid boundary during its formation by radioactive decay of its parent in the solid (both isotopes have Th as the immediate parent); and by desorption. The mechanism of α recoil is an important factor in the higher solubility of progeny isotopes compared with their parents. Uranium-234/uranium-238 activity ratios in

ground water are generally greater than 1.0 and can be as high as 28 (Gi82). Radium-224/radium-228 activity ratios in South Carolina ground water range from 1.2–2.0 (Mo83) and in Connecticut from 0.8–1.7 (Kris82). However, when the progeny/parent pair consists of different elements, geochemical factors become important controls of their relative solubility. An extreme example is ^{222}Rn , the immediate progeny of ^{226}Ra ; $^{222}\text{Rn}/^{226}\text{Ra}$ activity ratios in water can be as high as 10^6 . Because of α recoil and the different solubilities of the Th and U series isotopes, extensive disequilibrium occurs in ground water.

Recent studies have suggested that Ra is rapidly absorbed from ground water. King *et al.* proposed that the distance of Ra transport in ground water was less than that of ^{222}Rn (with a $t_{1/2} = 3.8$ days) due to continual adsorption of Ra onto the aquifer solids (Ki82). Krishnaswami *et al.* calculated adsorption and desorption rate constants for Ra in Connecticut aquifers and proposed that Ra removal rates are rapid, as short as a few minutes (Kris82). Equilibrium between adsorption and desorption is also quickly established, but Krishnaswami *et al.* concluded that the partition coefficient strongly favors the solid phase, and almost all Ra introduced into the ground water studied resides on particle surfaces in the adsorbed state. However, the extent of sorption is controlled by the geochemical reactivity of the aquifer material. King *et al.* (Ki82) note that the average ^{228}Ra and ^{226}Ra activity in the crystalline aquifers of South Carolina was lower than for the Coastal Plain sediments, even though the Th and U content of the rock aquifers was higher. Furthermore, the ^{222}Rn activity in the crystalline aquifers was 10 times greater than the aquifers sampled in the Coastal Plain. King *et al.* concluded that the affinity of Ra for adsorption sites in the fresh rock surfaces which have higher cation-exchange capacities was greater than for the sand and gravel deposits composed of refractory minerals such as quartz. Thus, Ra in ground water does not accumulate with ground water transport in aquifers; it stays very close to the area in which it is produced.

The insolubility of Ra and Th can be inferred from studies of potential contamination of

ground water due to seepage from U tailings ponds in New Mexico reported by Kaufmann *et al.* (Ka76). At one such pond, they estimated that nearly 3×10^9 liters of seepage entered the shallow aquifer during a 20-yr period. The wastes in this pond contained approximately 200 pCi/l of ^{226}Ra and 166,000 pCi/l of ^{230}Th . Thus, nearly a curie of ^{226}Ra and 500 Ci of ^{230}Th were available to leach with the shallow ground water; yet, in 1975, monitoring wells located 1 km down-gradient from the pond showed no evidence of contamination.

Through an understanding of the physical and chemical processes which control Ra distribution, we can now begin to interpret the new data base from state compliance reports, and to develop predictive models for Ra occurrence on which new regulations should be structured. These proposed models would characterize certain geological settings or aquifer types as producing ground water with high or low Ra content. The EPA has begun to develop a predictive model for the occurrence of ^{228}Ra , with a pilot study completed for two geological provinces, the Atlantic and Gulf Coastal Plain sedimentary aquifers and the Piedmont rock aquifer of the eastern United States (Mi82). Information on areas of high Ra occurrence is necessary to provide guidance to states for additional monitoring. From a regulatory point of view, areas of low Ra activity are very important, in that they could have a different monitoring priority and schedule. A predictive model for ^{228}Ra would also be valuable because so few samples were measured under the present analytical scheme.

Occurrence of ^{226}Ra and ^{228}Ra in drinking water

All but six states (Illinois, Nebraska, Colorado, Utah, Montana and Oregon) have reported known MCL violations for Ra as required by the interim regulations. There are approximately 200 reported public water suppliers with ^{226}Ra activities in excess of 5 pCi/l after normal treatment (Co83c). The following sections discuss these results and other studies by water types, geological setting, and isotope.

Surface water

The Ra content of surface water is usually very low. Radium-226 generally ranges between 0.1 and 0.5 pCi/l and the $^{228}\text{Ra}/^{226}\text{Ra}$ activity

ratio is generally greater than unity (Mo69; El82). Also, standard water treatment methods are known to remove Ra (Ep76b). To the best of our knowledge, no surface-water violations for Ra have been reported by the states. Thus, surface-water systems should be separately evaluated; perhaps they could be released from monitoring requirements for Ra once the source stream was documented as having low natural radioactivity.

Ground water

Out of the nearly 60,000 public water supplies in the United States, about 80% use ground-water sources. More than 90% of the ground-water supplies serve less than 3,300 people and are classified as small or very small. In general, Ra in drinking water is a small-system problem. Figure 2 is a compilation of the areas and specific sites which have high Ra in ground water from both state compliance data and published studies.

The available state compliance data for Ra comes almost exclusively from samples which first showed a gross α -particle activity of ≥ 5 pCi/l. Iowa used a screen of 2 pCi/l for gross α -particle activity. In some areas, states would analyze additional samples in an area where high radioactivity was found during the initial sampling. Radium-228 data were provided for about one-half of the 200 ^{226}Ra values reported. Statewide summaries of ^{226}Ra and ^{228}Ra data have been published for Georgia (Cl83), South Carolina (Ki82), Iowa (Krie82) and Illinois (Ro77); Lucas reported results for more than 90% of the communities in Illinois, Iowa, Missouri, and Wisconsin (Lu82).

There have been several studies on the temporal variability of the activity of Ra isotopes in ground-water systems. Kriege and Hahne reported that the mean value for the average percent deviations of 141 samples during 18 yr in Iowa was 21% with a relative standard deviation of 15% (Krie82). Michel and Moore found a maximum variation of 19% during 2 yr in individual wells (Mi80). Therefore, in single-well systems, one sample should be representative of the average annual activity; also the present requirement for monitoring at 4-yr intervals would not be necessary unless changes to the system have been made. Systems with

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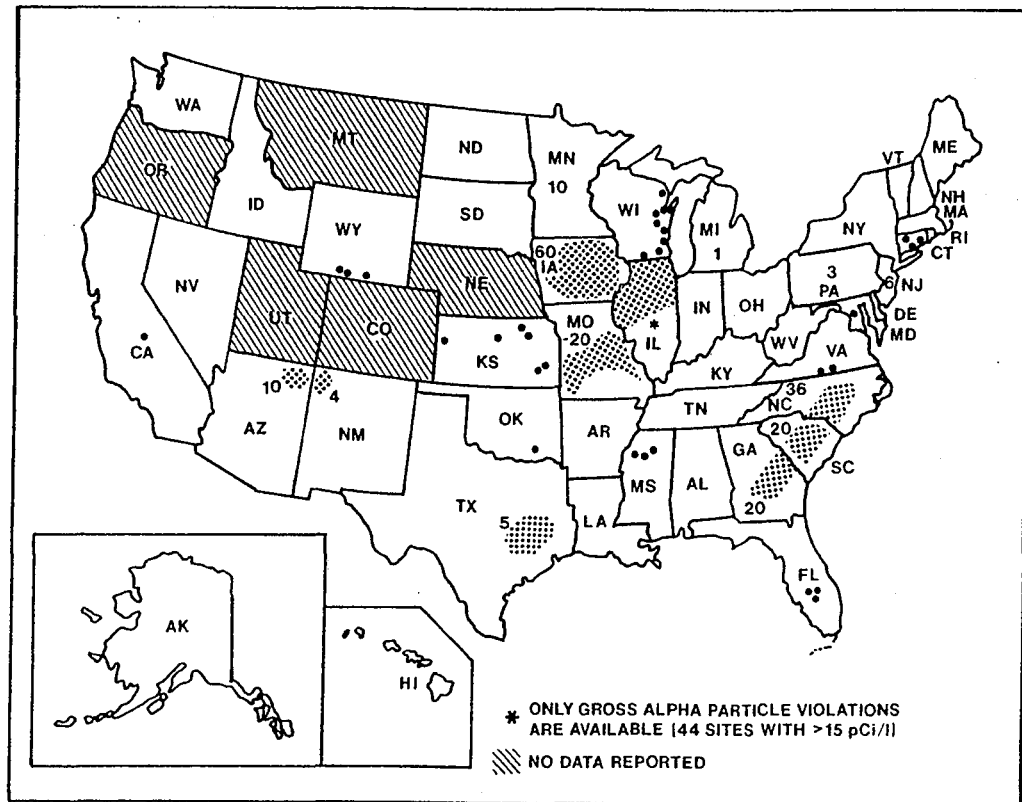


FIG. 2. The approximate locations and general areas of public water supplies which exceed 5 pCi/l of total Ra (^{228}Ra was reported or combined with ^{226}Ra for about one-half of the sites). Large dots represent individual violations. The dot pattern represents the general area of a group of violations, with the adjacent number indicating the number of violations in that group. When the locations were unknown, just the number of violations was indicated (modified after Cothorn and Lappenbusch (Co83)).

multiple wells have the potential problem of continuously variable Ra based on the relative contribution of each well when sampled.

From the data reported by the states, the mean total Ra activity for supplies exceeding the MCL was almost 10 pCi/l. Radium-226 activity was generally greater than ^{228}Ra activity, but these data were initially biased toward high ^{226}Ra . King *et al.* found that the average $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio was 1.2 for more than 180 samples throughout South Carolina (Ki82). Of these, 10 samples had ^{226}Ra less than 3 pCi/l, but total Ra greater than 5 pCi/l. King *et al.* estimated that perhaps 40–50% of the total Ra violations for the Piedmont and Coastal Plain provinces were missed using the prescribed

screening procedure which couples ^{228}Ra analysis to ^{226}Ra (Ki82). Kriege and Hahne reported additional sampling which identified eight violations for total Ra although the ^{226}Ra was less than 3 pCi/l (Krie82).

From the available data, there are two specific geological regions where more than 75% of the known Ra violations occur. They are: (1) the Piedmont and Coastal Plain provinces in New Jersey, North Carolina, South Carolina and Georgia; and (2) a north-central region, consisting of parts of Minnesota, Iowa, Illinois, Missouri and Wisconsin. The rest of the violations are generally scattered clusters, notably along the Arizona-New Mexico border, Texas, Mississippi, Florida and Massachusetts (Fig. 2). All of

these scattered violations had high ^{226}Ra activities, as would be expected from the screening methods used to detect them. Radium-228 activities in these systems were very low. We believe that the current analytical protocol has detected a large percentage of the systems with high ^{226}Ra . Cothorn and Lappenbusch have used the compliance data for ^{226}Ra to estimate that approximately 500 systems will be determined to exceed the MCL of 5 pCi/l (Co84).

Improvement on this estimate is difficult with the existing data base, which is comprised mostly of reported MCL violations for ^{226}Ra . Statistical analysis of these data is not possible because they were not randomly sampled. In this respect, states should be requested to submit all Ra results to facilitate further analysis. However, some calculations can be made to corroborate the previous estimates of MCL violations. Data from South Carolina showed that approximately 3.0% of the ground-water supplies exceeded the 5.0 pCi/l limit for total Ra (Ki82). (Note that the prescribed screening procedures detected only one-half of these violations). Applying that percentage to North Carolina and Georgia, both of which have similar hydrogeology, provides an estimate of 150 violations for all three states. In Iowa, approximately 10% of the 605 supplies sampled to date, using a lower screening criteria, exceed the MCL. Again, applying this percentage to all the ground-water systems of Iowa and half of Illinois, Missouri and Wisconsin yields 120 violations for Iowa, 75 violations for Illinois, 50 for Missouri, and 60 for Wisconsin. We can estimate violations for the states that have not reported as follows: 10 each for Utah, Colorado and Nebraska, and zero for Oregon and Montana. There are 71 violations reported in all other states. The total of these known and estimated violations is 556. Assuming 10–25% of the actual violations are missed during the prescribed screening procedure (actual data for Iowa, 8 out of 60 or 13%; for South Carolina, 8 out of 30 or 26%), the number of violations ranges between 600 and 700.

Estimates of population exposure nationwide can only be broadly made without additional information on populations served by the MCL violations, as well as on the results of all analyses. Lucas estimated that 91 communities in Illinois, Iowa, Missouri and Wisconsin with a population

of 599,000 consume water with ^{226}Ra greater than 5.0 pCi/l (Lu82).

Under the present screening methods, however, there was concern that ^{228}Ra violations were not being detected. Thus, the EPA recently funded a study to determine if a predictive model for the occurrence of ^{228}Ra could be developed. The Piedmont and Coastal Plain aquifers were selected as a pilot study area for development of a model because the radiochemistry of these provinces had been extensively studied, these were areas of known high ^{228}Ra activities, and nearly 300 values for ^{228}Ra were available.

The model comprises a multilevel classification of aquifer characteristics for each ^{228}Ra datum. The nature of the data precluded use of such analytical tools as regression analysis to establish a quantitative relationship between, for example, Th content and ^{228}Ra . As a result, the underlying model structure was evaluated to assess the existence of differences not only between the major aquifer types but also between lower-level classification within a major aquifer type. A detailed description of the model development, parameters, methodology and results can be found in a report by Michel and Pollman (Mi82). Only a summary is discussed below. Also, although the model was developed specifically for ^{228}Ra , values for ^{226}Ra were available and similar statistics were calculated. There is much to be learned from these differences in the results for these two isotopes.

Table 2 summarizes the means and ranges for those aquifer types which had significantly different ^{228}Ra distributions. Note the striking differences in the means, although the ranges are similar in some cases. Arkosic (immature, feldspar-rich) sand aquifers had mean values for both Ra isotopes up to an order of magnitude greater than quartzose sands. Limestones and metamorphic rock aquifers in the study area had very low activities of both Ra isotopes. Table 3 shows a ranking of all structural levels used in the pilot study, with symbols indicating groups of similarity of ^{228}Ra distribution (with ^{226}Ra means are also given without a ranking). Classes identified by the same letter code in the grouping column represent subsets of a group that are statistically indistinguishable from other classes of the same group. However, groupings

Table 2. Summary of ^{228}Ra and ^{226}Ra distribution in ground water by aquifer type for the Atlantic Coastal Plain and Piedmont provinces

Aquifer Type	Number of Values	Ra-228		Ra-226	
		Geometric Mean (pCi/l)	Range (pCi/l)	Geometric Mean (pCi/l)	Range (pCi/l)
Igneous Rocks (acidic)	42	1.39	0.0-22.6	1.80	0.0-15.9
Metamorphic Rocks	75	0.33	0.0- 3.9	0.37	0.0- 7.4
Sand	143	1.05	0.0-17.6	1.36	0.0-25.9
Arkose	92	2.16	0.0-13.5	2.19	0.0-23.0
Quartzose	50	0.27	0.0-17.6	0.55	0.0-25.9
Limestone	16	0.06	0.0- 0.2	0.12	0.0- 0.3

that overlap indicate that the particular individual groups are not unique. For example, group A as a class is not statistically different from groups B, C and D; groups E, F and G, however, represent groups of classes with significantly lower ^{228}Ra activities than group A. Although some groups were not significantly different, the ranking followed the anticipated trends within major types. For example, the ranking of igneous rocks, with syenite > granite > diorite, follows Th abundance in these rock types. Arkosic sand aquifers are ranked in order of Th content of the source rock, from high to low. This pilot study showed that specific aquifer types and geochemical conditions can be characterized as producing ground water with high or low ^{228}Ra activities. Its application can be demonstrated for the aquifers of the Piedmont and Atlantic and Gulf Coastal Plain provinces; high ^{228}Ra was likely to occur in aquifers composed of (1) acidic, igneous rocks and (2) arkosic sands with sources having high-to-medium Th content.

The results from the model were used to map specific areas (aquifer types) from New Jersey to Alabama that would be likely to produce ground water with high ^{228}Ra . In fact, in the Piedmont province, granitic rock aquifers younger than 350 million yr were shown to produce high-Ra ground water. Older rocks had undergone metamorphism which has tended to re-

crystallize Th and U into resistate minerals in which Ra is more tightly bound. The arkosic sand aquifers were restricted to the upper Coastal Plain from Virginia to Georgia. These aquifers are composed of sediments eroded from the nearby Piedmont rocks and are mineralogically immature. They contain higher amounts of Th- and U-bearing minerals than the middle and lower Coastal Plain sediments which were deposited farther from the source rocks. Thus, the trend in the Coastal Plain aquifers for ^{228}Ra , whose parent is not subject to secondary transport processes, is one of decreasing activities with distance from the Piedmont source. Only one out of 50 samples from the quartzose sands of the middle and lower Coastal Plains aquifers was greater than 3 pCi/l for ^{228}Ra . In contrast, ^{226}Ra in the middle Coastal Plain aquifer is highly variable, with values from 0-196 pCi/l, due to the ability of its parent to migrate in ground water and undergo secondary enrichment.

Knowledge of the conditions where very low radioactivity will occur is also very important. In the area studied, low ^{228}Ra occurred in aquifers of (1) metamorphic rocks, (2) quartzose sands and (3) limestone. Thus, the lower Coastal Plain, composed of extensive limestones and deep quartzose sand aquifers, is notable for its total lack of ^{228}Ra greater than 1.0 pCi/l in

Table 3. Ranking of all structural levels used showing groupings of similarity in Ra distribution. Radium-226 values are given for comparison but are not ranked

Grouping*	Mean Ra-228 (pCi/l)	No.	Structural Level	Mean Ra-226 (pCi/l)
A	3.03	2	Igneous, acidic, composite Th, syenite	0.75
A	2.49	46	Sand, unconsolidated, arkosic, high-Th source	2.03
B A	2.14	43	Sand, unconsolidated, arkosic, medium-Th source	2.73
B A C	1.59	35	Igneous, acidic, composite Th, granite	2.31
B D A C	0.85	3	Igneous, acidic, composite Th, diorite	0.99
B D E C	0.52	31	Sand, unconsolidated, quartzose, medium-Th source	1.72
F D E C	0.39	37	Metamorphic, high-grade, nonspecific Th	0.32
F D E	0.34	10	Metamorphic, low-grade, nonspecific Th	1.41
F D E	0.28	3	Sand, unconsolidated, arkosic, low-Th source	0.32
F D E	0.28	13	Metamorphic, medium-grade, nonspecific Th	0.24
F D E G	0.24	15	Metamorphic, high-grade, specific Th, monazite	0.24
F E G	0.12	2	Igneous, acidic, refractory Th, monazite	0.14
F G	0.09	19	Sand, unconsolidated, quartzose, low-Th source	0.09
F G	0.06	16	Chemical precipitates, limestone	0.12

*A>B>C>D>E ($p < 0.05$).

ground water. Radium-226 will be more variable because of the high solubility of U complexes in the carbonate system, but it is generally detected by the gross α -particle activity screen. Nevertheless, there have been only three ^{226}Ra violations reported for the entire lower Coastal Plain province from New York to Texas. These

violations were all from one region in Florida.

The second area of high radioactivity is the north-central region. Much of the ground water comes from deep aquifers, frequently having ^{226}Ra activities of 5-25 pCi/l; ^{228}Ra can be as high as 32 pCi/l (Lu82). There is no apparent correlation between ^{228}Ra and ^{226}Ra and no

specific trends in their distribution by aquifer, depth in the aquifer, or areal extent. Interpretation of the Ra distribution in this area is complicated by complex hydrogeology and multiply screened wells in different aquifers. There is evidence of significant U migration, both during geological time and the present, which provides a mechanism for high ^{226}Ra as well as resulting in a complex distribution and disequilibrium of U series isotopes (Gi82; Li02). Possible sources for high ^{228}Ra have not been identified, but the ^{228}Ra distribution may be able to be explained by analysis of the sources, depositional setting, and diagenesis of the sedimentary rock aquifers.

Limited work has been done on Ra occurrence in the other geological provinces. Radium-226 has been found to be high in areas of U mineralization, such as in Texas and the Colorado Plateau in Arizona and New Mexico (Fig. 2) and violations are expected in Utah and Colorado when these states report. Thorium enrichment zones, such as veins and placer deposits, are expected to produce only scattered, local ^{228}Ra problems, due to its limited transport in ground water. These areas would be extremely difficult to locate under the present regulations. However, aquifers with much lower but disseminated Th and U (such as granites, tuffaceous rocks, and immature sandstones) are more likely to have higher background radioactivity and wider occurrences of both ^{228}Ra and ^{226}Ra in ground water.

Occurrence of ^{224}Ra

Data on ^{224}Ra activities in ground water are scarce. However, it appears that the activity of ^{224}Ra is equal to or as much as twice the ^{228}Ra activity and therefore could be as high as 30–40 pCi/l. This ^{224}Ra activity is unsupported; activities of its parent, ^{228}Th , are usually less than 0.01 pCi/l. Thus it enters ground water by α recoil during decay of ^{228}Th adsorbed on the surface of aquifer solids. The radiotoxicity of ^{224}Ra and its progeny is small because of their extremely short half-lives.

RADIUM CONCLUSIONS

From the state compliance data and other studies, much more is now known about the occurrence of Ra isotopes in public drinking

water supplies, and this information should be incorporated into the revised regulations.

(1) Surface water has very low Ra activities; the monitoring interval after initial validation should be significantly lengthened, or perhaps omitted, for surface-water systems.

(2) Fewer samples may be needed to determine the average annual activities, particularly for single-well systems; the monitoring interval could be lengthened for unmodified systems.

(3) Monitoring requirements for ^{228}Ra should be decoupled from ^{226}Ra . Instead, separate guidelines for the occurrence of Ra isotopes are needed.

(4) Regional ^{226}Ra problems are fairly well known. The few additional occurrences of high ^{226}Ra activity will be difficult to find without analysis of every system. As important, however, are those areas which have low ^{226}Ra . All values should be compiled regionally or nationally, to document the ^{226}Ra distribution for each geological province, with the goal to classify areas with a high degree of certainty as producing low ^{226}Ra ground water. The revised regulations should include separate, less stringent and less costly monitoring requirements for such regions. This approach would shift monitoring efforts toward known or uncertain areas of high ^{226}Ra and provide more data on the actual distribution of high ^{226}Ra activities, which will allow for a better risk assessment.

(5) The occurrence of ^{228}Ra is not well known. It has been shown that, using the present screening procedures, 10–50% of the violations for total Ra are being missed. More extensive measurements of ^{228}Ra would be difficult because of the problems with the approved analytical method. An alternative approach would be to develop a conceptual, predictive model for ^{228}Ra occurrence, based on geochemical principles, to identify specific types of aquifers which are likely to have ^{228}Ra problems. Once verified, this model should be the basis for developing regional guidelines for monitoring in areas more likely to have high ^{228}Ra . This same approach can be used for refining and interpreting occurrence data for U and Rn as part of the regulatory process of developing standards for these isotopes.

(6) Finally, because aqueous radiochemistry

is a complex, technical field, the EPA should provide state water-supply personnel with background and explanatory guides in laymen's language, which will assist them in understanding radiological problems and in the implementation of the regulations.

OCCURRENCES OF U ISOTOPES IN PUBLIC DRINKING WATER

Introduction

Uranium has three natural isotopes with long half-lives ($t_{1/2}$) that permit transport into potable water supplies. These isotopes are ^{238}U (99.27% natural abundance), $t_{1/2} = 4.7 \times 10^9$ yr, ^{235}U (0.72% natural abundance), $t_{1/2} = 7.04 \times 10^8$ yr, and ^{234}U (0.006% natural abundance), $t_{1/2} = 2.54 \times 10^5$ yr. All of these isotopes emit α radiation and produce a long decay series of progeny. The group of U isotopes are found in the Earth's crust with an abundance of 4×10^{-4} % (Hu73) and are found in rocks and minerals such as granite, metamorphic rocks, lignites, monazite sand, and phosphate deposits as well as in U minerals such as uraninite, carnotite and pitchblend (Ca80). It is a trace element in coal, peat, and asphalt and is present in some phosphate fertilizers at a level of about 100 $\mu\text{g/g}$ or 67 pCi/g. Despite its widespread abundance it has not been shown to be an essential element for man (Hu73). There is no standard for U in water supplies as a radioactive element since, until recently, it has been considered by the U.S. Nuclear Regulatory Commission to be a toxic heavy metal with the standard for ingestion relating to its chemical toxicity (3×10^4 pCi/l); (10 *Code of Federal Regulations* 20, 601, Appendix B). However, some concentration measurements in potable water have been done in association with gross α measurements for the Ra drinking water standard. The U activity measured was to be subtracted from the gross α activity measurement to show compliance with the gross α standard (Dr81). A recent analysis of U in water supplies was conducted by Oak Ridge National Laboratory (Dr81) using 89,944 measurements of U surface, ground and domestic waters primarily obtained from the NURE program. The results of this study are reviewed in this report.

Geochemistry of U

Although there are geological processes which enrich U in certain rock formations, it occurs as a common trace element in most rock types. Because of the insolubility of U^{4+} , U must be oxidized in order to be transported in ground water. The greater solubility of U^{6+} is due in part to its tendency to form uranyl di- and tri-carbonate anions. Thus, U solubility is a function of not only the redox potential of water but also of the pH and the partial pressure of CO_2 in the system. In comparison to Ra, the stability of the uranyl carbonate complexes and their long half-life allow for U to be transported long distances under oxidizing conditions. Uranium is removed from solution by sorption or reducing barriers, a process which has been well described in the sandstone-type U deposits in the western United States (Ga77).

There have been many studies of the isotopic composition of U in natural waters which have shown that most contain more activity from ^{234}U than from ^{238}U . The $^{234}\text{U}/^{238}\text{U}$ activity ratio can be as high as 28, but usually ranges between 1 and 3 (Ch71; Gi82). The higher activity of ^{234}U in water is due to its selective mobilization by α recoil. The natural abundances of isotopes and the half-lives give 0.33, 0.015 and 0.33 pCi/ μg of natural U for ^{238}U , ^{235}U and ^{234}U , respectively, or 0.68 pCi/ μg total. Thus, isotopic enrichment can cause changes in the specific activity of the total sample of U. Total depletion of ^{234}U from the sample and replacement by an equal activity of ^{238}U will result in no net change of total activity; however, the total mass of U would almost double. The human dosimetry will also be changed since the α energies are not the same. Methods which depend on the mass of U will not predict the correct activity for samples with variable U isotope enrichment.

Occurrence of U in ground and surface water

Uranium concentration in water depends on factors such as the U concentration in the host aquifer rock, the presence of O and complexing agents, chemicals in the aquifer, chemical reactions with ions in solution and the nature of the contact between the U minerals and the water. These factors vary with regions of the

United ground genic f and su expect to state minous tions of are sho can be eraged l shown surprisi factors water ti identical average Table 4 in provi to 2.3 in groupin;



FIG.

United States due to rainfall, geology and ground-water flow patterns, and to anthropogenic factors such as use rate of ground water and surface water (Sc62). Thus, one would expect large variations of U content from state to state. The geological provinces of the coterminous United States derived from generalizations of rock types and hydrological flow systems are shown in Fig. 1 and Table 1. These zones can be compared to the U concentrations averaged by state from the (NURE) measurements shown in Figs. 3, 4 and 5. These averages are surprisingly similar, showing differences of only factors of 4 higher concentrations in ground water than in surface water, with many states identical for ground and surface water. The average values for each province is given in Table 4 with average values ranging from .02 in province 2, the Appalachian Mountains, up to 2.3 in province 8, the Colorado Plateau. By grouping low, medium and high averages, one

sees the four major zones of similar concentration: Zone 1, the Appalachian Mountains and New England; Zone 2, Appalachian and Interior Plateau, and Coastal Plain; Zone 3, the Glaciated Central Platform, Western Central Platform, Rocky Mountain System, and Colorado Plateau; Zone 4, the Basin and Range and the Columbia Plateau and Pacific Mountain System.

The provinces chosen by Beddinger (Be81) may be compared with those chosen in 1962 by Scott and Barker (Sc62). These provinces are shown in Fig. 1 and compared by region in Table 1. Concentrations of U are given in Table 4 for provincial schemes. The data of Scott and Barker comprise 561 samples collected in the coterminous United States from 1954-58 and are expressed in $\mu\text{g/l}$ (thus they represent ^{238}U only).

Table 5 shows population vs U concentration for drinking water sources with more than 10,000 people (Dr81). The levels are given up



FIG. 3. Population-averaged U concentration in picocuries per liter for surface water in the United States.

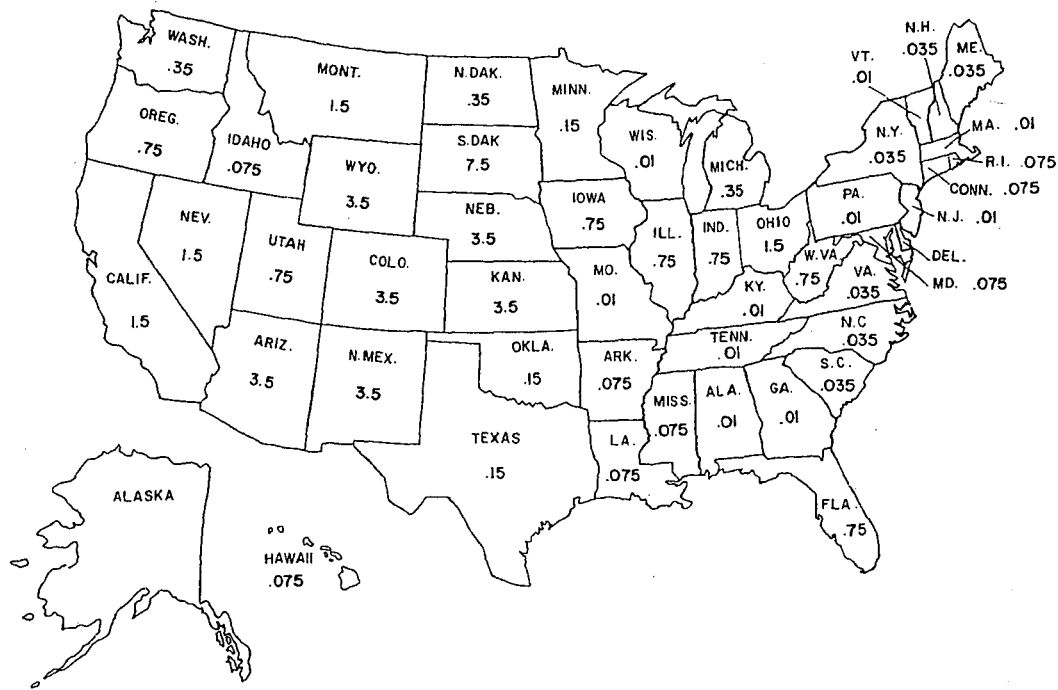


FIG. 4. Population-averaged U concentration in picocuries per liter for ground water in the United States.

to greater than 100,000 population. Due to limitations in the source information (Dr81), no information is available for cities of populations less than 10,000, showing a need for more information on small systems.

Relative source contribution of U

The dietary intake of U in United States food is variously reported from 0.87–0.94 pCi/day (Ha73) to 0.2–0.9 pCi/day (UN77) with an average of 0.4 pCi/day. The comparison with drinking water of average concentration of 2 pCi/l and two liters per day consumption gives 4 pCi/day of water-derived U which is 5–10 times greater than the food-derived U. Air contributions of U are much smaller than the food and water contributions.

Uranium conclusions

(1) The data shows that elevated levels of U found are found in surface water as well as in ground water.

(2) The highest average values of U concen-

tration are found in decreasing order in the following provinces (see Fig. 1): Colorado Plateau, Western Central Platform, Rocky Mountain System, Basin and Range and Pacific Mountain System. The state with the highest U concentration is South Dakota. Modeling these variations would be very helpful for regional standards.

(3) High U concentrations in the East are widely separated, and most values in the East are low.

(4) Isotopic estimates are needed for ^{238}U and ^{234}U since they are found in disequilibrium in water. Regional variations should be modeled.

(5) More analyses are needed for low population systems.

OCCURRENCE OF Rn ISOTOPES IN PUBLIC DRINKING WATER

Introduction

There are two isotopes of Rn, with half-lives long enough to be considered as drinking water radionuclides. The first is ^{222}Rn which is the

progeny half-life of is the pro "thoron," time delay water of allows ma is not ob: henceforth and can ingested a well as by lungs. Wt washing, b from the v into α -em: eny are cl aerosol pa smoke, or and may t lung, bring

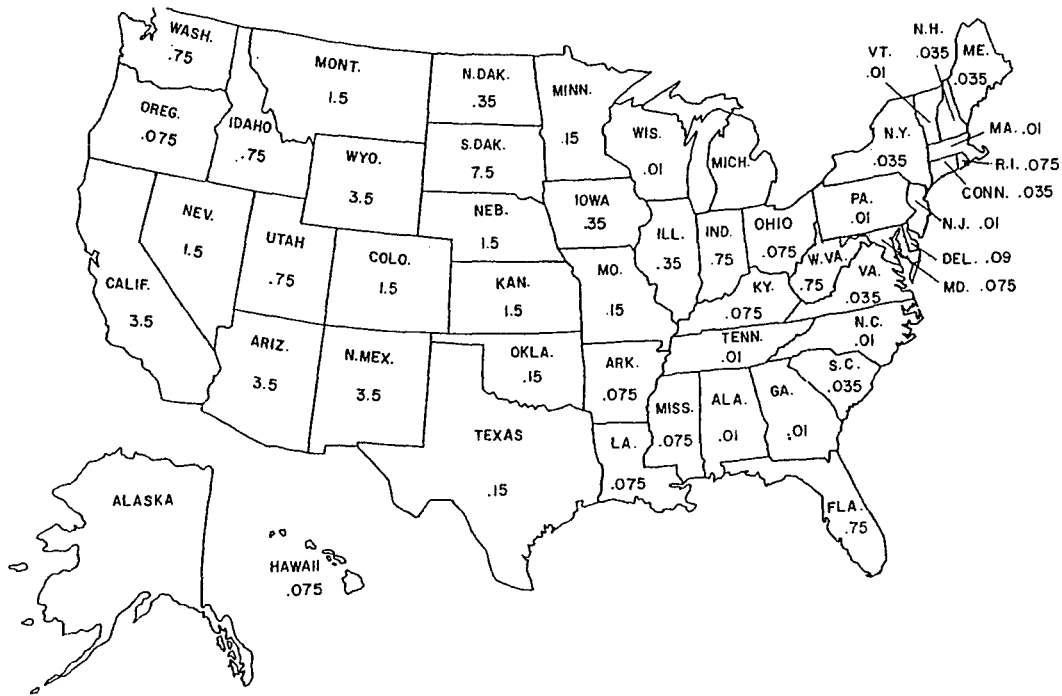


FIG. 5. Population-averaged U concentration in picocuries per liter for domestic water in the United States.

progeny of ^{226}Ra , called "radon," and has a half-life of 3.84 days. The second, ^{220}Rn , which is the progeny of ^{224}Ra , was historically called "thoron," and has a half-life of 56 sec. The time delay from production to consumption of water of a few hours to a few days for water allows many decay half-lives for ^{220}Rn , and it is not observed in water supplies. Radon-222, henceforth simply Rn, is transported by water, and can lead to public exposures by being ingested and exposing the digestive system as well as by becoming airborne and exposing the lungs. When water is used for cleaning, dish-washing, bathing, or clothes washing, Rn escapes from the water into building air where it decays into α -emitting progeny. The resulting Rn progeny are charged and will frequently attach to aerosol particles in the air. The dust, cigarette smoke, or aerosol particles will then be inhaled and may become attached to the interior of the lung, bringing the α particle-emitting Rn progeny

into close association with the cell lining of the respiratory system (Ar75).

There is no federal standard for Rn in water, although studies on ingestion doses (Hu65; As79) and inhalation doses have been done (Pr81; He83a; He79). There have been standards for Rn in mine air, and for Rn from soil gas in buildings placed on mine tailings in the United States and Canada (Us79; At77). Sweden has standards for Rn from soil gas in areas of alum shale and granites (Ak81). Some of these standards are given in Table 6. In the past, Rn and its progeny have been excluded from the drinking water standards, and considered only to be an interference in the Ra measurements.

Information about levels of Rn have been obtained in state studies and by a federal study done by the U.S. Environmental Protection Agency Environmental Radiation Facility in Montgomery, AL, and by the University of Texas at Houston.

Table 4. Uranium concentration in the provinces

Beddinger Provinces	Beddinger's uranium concentrations (pCi/l)		Scott's uranium concentrations (pCi/l)	
	Arithmetic Mean	Standard Deviation	Arithmetic Mean	
1. New England-- Adirondack Mts.	0.46	0.03] II	.34
2. Appalachian Highlands Piedmont	0.020	0.013		
3. Appalachian Interior Plateaus	0.137	0.27	III	.34
4. Coastal Plain	0.108	0.19	I	.14
5. Glaciated Central Platform Aquifers	1.04	2.17	V	.34
			VI	.71
6. Western Central Platform	2.1	2.2	VI	1.5
7. Rocky Mountain System	1.99	1.27	VII-VIII	1.15
8. Colorado Plateaus	2.31	1.4	IX	.54
9. Basin and Range	2.15	1.53	VIII	1.15
10. Columbia Plateaus	0.52	0.389] X	0.14
11. Pacific Mountain System	1.41	1.81		

Radon-Radium geochemistry

Radon is a water soluble inert gas and its occurrence is controlled by physical variables such as pressure, temperature, emissivity of Rn from rocks, as well as by time, and by the geochemistry of its parent ^{226}Ra . High activity of Rn is associated with granitic rocks (St81), U minerals (Ta78), such as uraninite, carnotite and with tailings from phosphate fertilizer processing (Us79) and U mines.

Transport of Rn in water from rocks

As discussed in the geochemistry section, the occurrence of Rn in water is controlled by chemical concentration of Ra in the host soil

on rock and by emissivity of Rn into the water. The physical condition of the rock matrix appears to play a greater role in Rn production than does the concentration of parent Ra. Several investigators have examined the mechanisms influencing the release of Rn from rock grains and the transport of Rn through an aquifer (An72; Ra83; Ta64; Ta80). Experimental and theoretical considerations indicate that diffusion along microcrystalline imperfections dominates the release of Rn into the surrounding interstitial waters. The movement of Rn in water is governed by water transport rather than diffusion in most cases, i.e. cases in which the percolation velocity is greater than 10^{-5} cm/sec.

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Table 5. Population vs concentration of U distribution for drinking water sources that serve more than 10,000 people. The numbers in parentheses are the number of cities sampled followed by the total in the category

POPULATION	0.05 -0.5	0.51 -1.0	1.1 -2	2.1 -3	3.1 -4	4.1 -5	5.1 -6	6.1 -7	7.1 -8	8.1 -9	9.1 -10	10./0 -11	greater than 11 pCi/l
10,001 - 50,000 (532/2407)	292	75	46	14	17	4	75	4		2		3	
50,001 - 75000 (63/224)	36	4	4	3	3		10		1	1			(12 pCi/l)
75,001 - 100,000 (31/101)	15	5	3	1	4		3						
greater than 100,000 (174/276)	117	15	10	7	8	3	7	4	2				(30.2 pCi/l)

*assumed secular equilibrium of U-234 and U-238.

The variation of Rn concentration with rock type for well water in Maine is illustrated in Table 7. This variation of a factor of 20 illustrates variation in rock types in Maine. Public utility water measured at the wells, which is also presented in the Maine list, is a factor of 5 lower than the state average. This is due to the higher capacity of the gravel aquifers used for utility water supplies. These gravel aquifers

allow more water to mix with the same amount of Rn, leading to lower concentrations of Rn.

Occurrence of Rn in public water supplies

Concentrations of Rn in various water sources conform to the log normal distribution. Table 8 shows the results of a blind sampling of public water supplies in the central United States. A reanalysis of other published data (Table 9)

Table 6. Standards for Rn in air and proposed action to be taken

<u>United States (Us79)</u>	<u>In phosphate-mining regions in Florida</u>
	4 pCi/l: take remedial action
	2 pCi/l: reduce to as low as is reasonably achievable
<u>Canada (At77)</u>	<u>In uranium-mining regions</u>
	30 pCi/l: take prompt remedial action
	4 pCi/l: take remedial action
	2 pCi/l: investigate
<u>Sweden (Ak81)</u>	11 pCi/l: for existing buildings
	4 pCi/l: for houses undergoing remodeling
	2 pCi/l: for newly constructed houses

RADIOACTIVITY IN PUBLIC WATER SUPPLIES

Table 7. Average Rn values in private and public water supplies

	Arithmetic Mean pCi/l	Maximum pCi/l	Number of Samples
Maine (He79,83b)	10,000	1,000,000	2,000
In granite zones	22,000	300,000	136*
In sillimanite grade zone	13,600	100,000	35*
In chlorite	1,100	2,500	56*
In public utilities	2,000	11,700	64

*Rock grade determined by geologist for each private well.

Table 8. The distribution of Rn in municipal water supplies in the central United States

STATE	Well Waters			Distribution Systems		
	N.	G.M. (pCi/l)	G.S.D.	N.	G.M. (pCi/l)	G.S.D.
Arkansas	6	135	2.66	20	47	1.95
Indiana	10	151	2.13	23	70	2.20
Iowa	33	175	2.13	31	111	2.45
Louisiana	61	151	1.84	22	93	2.54
Minnesota	28	252	2.08	28	183	2.65
Nebraska	47	262	1.85	21	178	3.68
New Mexico	27	287	2.39	20	220	2.16
Oklahoma	7	117	1.74	6	134	1.17
COMPOSITE	209	197	2.10	174	115	2.75
----- Texas				278	131	2.70

G.M. = Geometrical Mean

G.S.D. = Geometrical Standard Deviation

N. = Number of water supplies sampled

Table 9. Radon-in-water results by state and source. Results are geometric means in units in pCi/l. Parentheses values are numbers of samples

STATE	PRIVATE WELL	PUBLIC WATER SUPPLY*	PUBLIC GROUND-WATER SUPPLY	PUBLIC SURFACE WATER SUPPLY
				**
AL	120 (22)	8 (31)	70 (182)	ND (8)
AR	230 (2)	1400 (1)	12 (22)	ND (1)
AZ	--	--	250 (124)	ND (6)
CA	43 (6)	790 (2)	470 (15)	ND (2)
CO	--	--	230 (76)	--
DE	--	--	30 (72)	--
FL	6000 (34)	320 (2)	30 (327)	--
GA	2100 (2)	44 (32)	67 (225)	43 (2)
IA	--	--	220 (85)	ND (2)
ID	--	--	99 (155)	--
IL	--	--	95 (314)	--
IN	--	--	35 (185)	--
KS	--	--	120 (47)	74 (2)
KY	1500 (10)	ND (18)	32 (104)	ND (5)
MA	1000 (8)	7 (2)	500 (212)	38 (2)
ME	7000 (24)	990 (71)	--	--
MN	1400 (1)	600 (1)	130 (233)	--
MO	ND (2)	--	24 (138)	ND (2)
MS	--	260 (2)	23 (104)	--
MT	4300 (8)	--	230 (71)	ND (6)
NC	15 (29)	27 (2)	79 (404)	ND (4)
ND	--	440 (2)	35 (133)	--
NH	1400 (18)	9 (12)	940 (52)	ND (6)
NJ	--	--	300 (38)	--
NM	59 (14)	45 (8)	55 (171)	ND (18)
NV	--	--	190 (57)	--
NY	1500 (4)	34 (20)	52 (292)	ND (1)
OH	--	--	79 (165)	--
OK	--	--	93 (83)	--
OR	450 (18)	--	120 (69)	ND (4)
PA	910 (16)	--	380 (105)	--
RI	6500 (69)	5200 (6)	2400 (575)	ND (10)
SC	1100 (28)	--	130 (384)	ND (14)
SD	4200 (2)	59 (2)	210 (155)	--
TN	ND (2)	ND (2)	12 (98)	--
UT	--	--	150 (195)	--
VA	560 (42)	--	350 (284)	ND (4)
VT	210 (23)	840 (4)	660 (71)	13 (16)
WI	730 (40)	28 (4)	150 (278)	ND (12)
WY	--	--	330 (32)	ND (2)
US	920 (434)	68 (224)	130 (6298)	1 (131)

*May include both ground water and surface

**ND - Not detected above background levels

shows a similar trend. Some sources appear to be samples from a single log normal distribution, others from two, or perhaps three distributions, as indicated by the sharp breaks or bends in the

plots. For this reason geometrical averages are used for the sample shown below.

The occurrence of Rn in public ground-water supplies in the United States is shown in Fig. 6

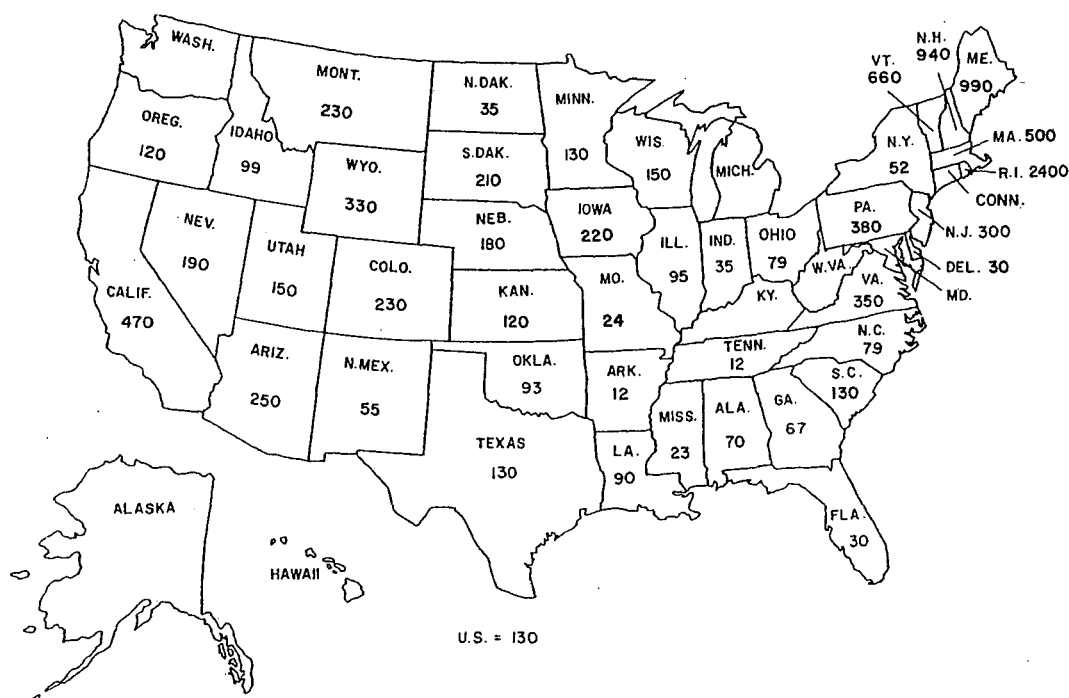


FIG. 6. Geometric average Rn concentration in picocuries per liter for public ground-water supplies in the United States.

and Fig. 8 (taken at the tap) and Table 9 (mainly utility samples). Radon activities are thousands of times higher than U or Ra, probably due to absorption of the Ra and U by the host rock (K_i82). The results of these geometric means show highest values in mountain states especially in the Appalachians with the highest states Rhode Island, Maine, New Hampshire, Vermont, Massachusetts, Pennsylvania and Virginia. California is the highest western state. The high Rn values associate with the granitic areas in the Appalachian Highlands Piedmont Provinces (Fig. 1). Midwestern and coastal plane values are lower and mountain states in the west are higher. These results of geometrical averages show the private supplies are higher by a factor of 3–20 times the public ground-water samples. This factor results from the use of low capacity wells for private supplies while public supplies use high capacity sand or gravel wells. The higher states in the private well list are Rhode Island, Florida, Maine, South Dakota,

Montana and Georgia. Larger numbers of samples would be desirable to strengthen these conclusions. Public surface supplies have Rn concentrations less than 100 pCi/l. Table 10 shows a breakdown of the Rn concentrations in water by state and by population of the town using the well. Highest Rn concentrations are found in the less than 100 category (see Maine and United States for examples). The United States geometric population average is 187 pCi/l.

Major sources of indoor Rn

Radon produced from Ra in the surficial soil and rock (A_k81) is released into houses from water, soil gas, fuel gas, and construction materials and outdoor air. Both water and soil gas can be transported into buildings through cracks, drain holes, as well as water and fuel gas supply pipes (S_c82). The Rn in the ground water is released as it is mixed with air in such indoor uses as cleaning, bathing, dish and clothes wash-

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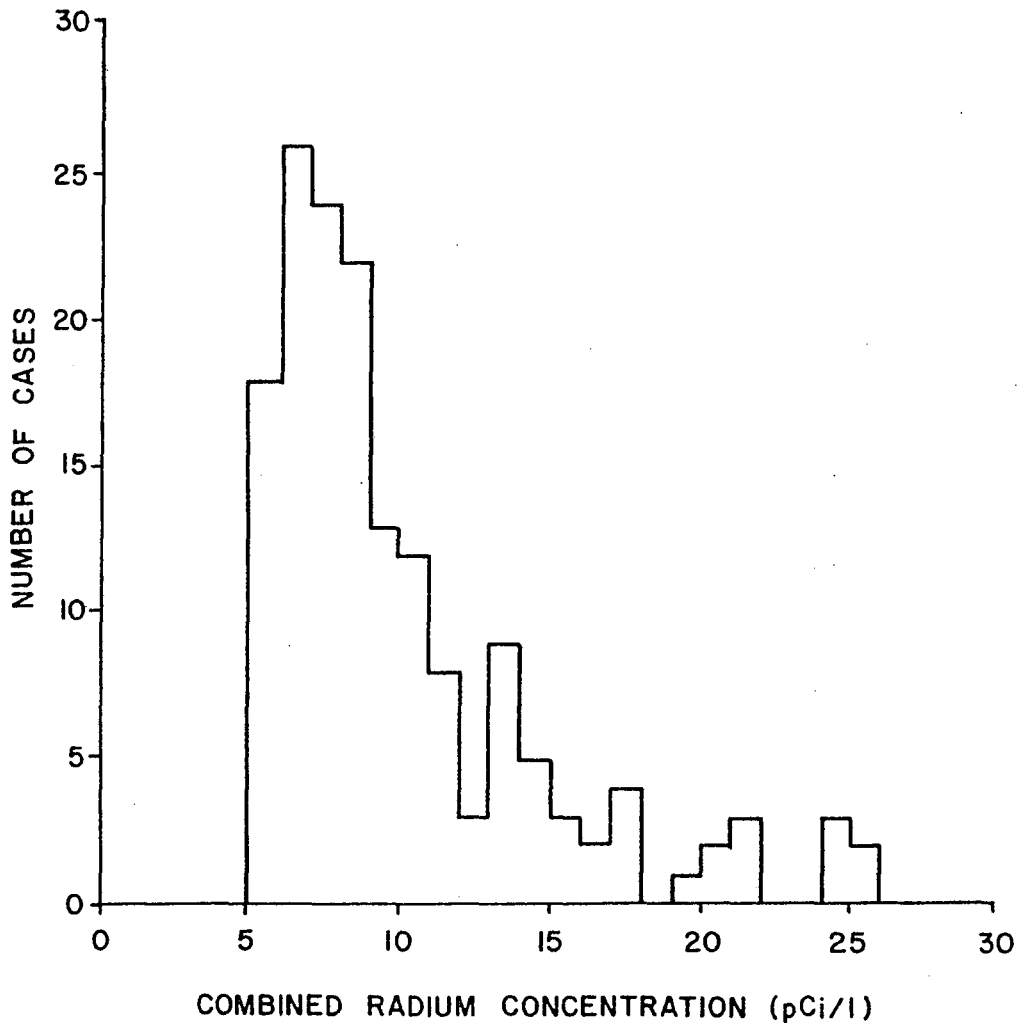


FIG. 7. Number of utility water supplies above 5 pCi/l with a measured Ra concentration.

ing and toilet flushing (He83a; Pa79). Soil gas will mix into building air and then diffuse throughout the house. Radon from fuel gas enters building air from unvented heaters or stoves. Thus, the Rn concentration in air will depend on the sum of all Rn sources (Ge78), the volume of the building, and the ventilation rates of the building (F180; Ne81). The average value for Rn in house air due to all these sources has been estimated at 0.3–2.2 pCi/l in normal regions, 1.1–1.67 in anomalous regions of the United States (Br81).

The soil. Radon diffuses from the soil through cracks in foundations, unventilated crawl spaces, basement drains, and other pathways into the living space. Direct outgassing from the soil is the dominant source of indoor Rn in most cases contributing 0.03–1.5 pCi/l in normal regions, and 0.3–15 pCi/l in anomalous regions. If most Rn enters structures through the basement or foundation, Rn concentrations would tend to decline markedly with story above ground, as shown in Table 11. The limitations of building materials and ground waters as

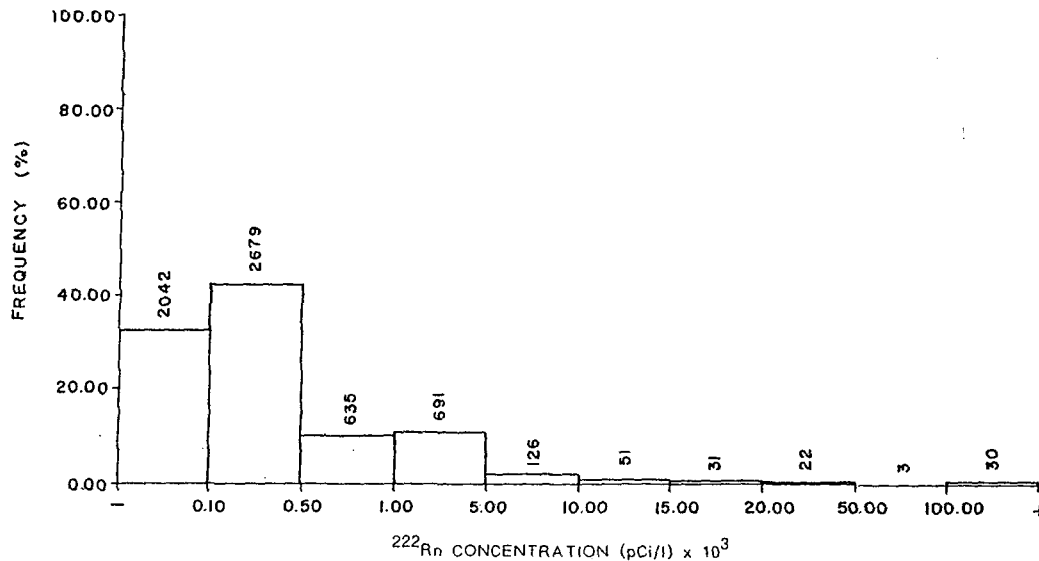


FIG. 8. Radon concentrations in picocuries per liter for all public ground water supplies in the United States.

sources of Rn (see below) coupled with the tendency toward single distributions in given areas combine to suggest that the soil is generally the largest source.

Ground water. Ground waters containing Rn can add substantially to the amount of Rn in the air of a dwelling. Much of the dissolved Rn can escape when water is used for various domestic purposes inside a dwelling. The amount of Rn in indoor air due to the use of water depends a great deal on architectural and life-style related variables. The most sensitive dwellings will be small, relatively tight structures in which large amounts of water are routinely used in household appliances. A model for the average increment to the indoor atmosphere can be expressed as:

$$C_a = \frac{C_w}{24RV} \sum e_i w_i,$$

where C_a and C_w are the concentrations (pCi/l) of Rn in the air and water, respectively, R is the air change rate (hr^{-1}), V is the dwelling volume, w_i represents the average amount of water (l) used daily in the i th domestic appli-

cation, and e_i denotes the transfer efficiency, or the fraction of Rn released to the air for the i th application.

A number of investigators have made semi-empirical determinations of C_a/C_w ; most are on the general order of 10^{-4} pCi/l in the air per pCi/l in the water. Table 12 shows a number of such estimates and the underlying assumptions used in their derivation. Using Table 12 the range of values for Rn in air from water in the United States ranges from 0.2 pCi/l average for Rhode Island to as low as 1.2×10^{-3} pCi/l average for Tennessee.

The relative importance of water as a source of indoor Rn will of course depend upon the amount of Rn in the water and the magnitude of other sources. Upper limit calculations on one recent data set (Table 12; Pr83a) show water as the source of up to 35% of the net (indoor minus outdoor) Rn observed in a set of 81 bedrooms in single-family houses in the State of Maine. The highest Rn levels were seen in the basement, which suggests that the soil is a major source of indoor Rn. The long-term average levels noted in the bathroom were also higher than those noted in the other living areas, however. Both the integrated indoor air Rn

Table 10. Radon-in-water results by state and population. All results are geometric means in units of pCi/l. Parentheses values are in numbers of samples

STATE	Population Ranges					
	< 100	100-1000	1000-5000	5000-10,000	> 10,000	Unknown
AL	--	83 (10)*	59 (76)	82 (46)	68 (44)	170 (6)
AR	--	32 (6)	6 (3)	--	12 (2)	8 (13)
AZ	--	240 (2)	200 (68)	350 (22)	340 (30)	160 (2)
CA	--	--	--	--	--	470 (15)
CO	--	130 (6)	220 (54)	400 (8)	300 (8)	--
DE	--	100 (4)	39 (44)	11 (12)	23 (12)	--
FL	--	320 (2)	290 (4)	49 (78)	24(243)	--
GA	57 (4)	42 (12)	130 (56)	190 (27)	52 (45)	39 (81)
IA	--	1120 (13)	230 (36)	150 (24)	66 (10)	75 (2)
ID	6 (3)	210 (14)	130 (83)	36 (25)	100 (30)	--
IL	--	71 (1)	81 (30)	80 (98)	110(185)	--
IN	--	--	45 (71)	17 (58)	50 (56)	--
KS	260 (2)	230 (12)	43 (11)	320 (4)	58 (8)	210(10)
KY	--	20 (10)	42 (76)	36 (8)	5 (10)	--
MA	3300 (2)	--	380 (47)	540 (67)	510 (88)	580(8)
ME	670 (3)	1600 (23)	690 (33)	2700 (7)	450 (4)	400(1)
MN	--	180 (22)	150 (76)	68 (43)	140 (90)	330(2)
MO	ND (2)	58 (54)	26 (54)	1100 (4)	78 (6)	ND(18)
MS	--	--	15 (45)	51 (26)	23 (33)	--
MT	740 (4)	280 (6)	270 (49)	160 (8)	18 (4)	--
NC	11,000 (6)	250(111)	45 (229)	16 (32)	21 (16)	8100(10)
ND	--	13 (10)	39 (112)	97 (5)	10 (6)	--
NH	1700 (2)	1200 (4)	960 (24)	1000 (14)	550 (8)	--
NJ	--	--	--	360 (6)	1200 (12)	120 (20)
NM	1300 (2)	620 (7)	48 (89)	42 (16)	33 (49)	450 (8)
NV	--	220 (2)	240 (36)	72 (9)	530 (2)	180 (8)
NY	--	--	56 (59)	31 (85)	71(113)	56 (35)
OH	--	300 (2)	160 (4)	56 (76)	100 (83)	--
OK	--	260 (1)	65 (33)	190 (21)	79 (26)	96 (2)
OR	--	210 (2)	110 (48)	23 (6)	320 (6)	180 (7)
PA	1900 (2)	--	260 (34)	450 (32)	440 (37)	910 (16)
RI	1700(91)	3900(325)	980 (71)	1300 (30)	1200 (58)	--
SC	1400 (32)	870 (30)	92 (229)	74 (60)	60 (31)	410 (2)
SD	310 (16)	300 (41)	160 (85)	210 (8)	200 (5)	--
TN	--	160 (4)	19 (48)	6 (25)	5 (21)	--
UT	--	260 (2)	140 (104)	200 (39)	170 (48)	150 (2)
VA	85 (6)	880 (56)	140 (151)	320 (12)	720 (14)	2300 (45)
VT	120 (1)	--	540 (24)	--	1000 (2)	750 (44)
WI	--	--	150 (169)	190 (61)	130 (48)	--
WY	--	880 (6)	230 (19)	530 (6)	54 (1)	--
US	990(175)	620(777)	98(2446)	92(1098)	88(1464)	140(338)

*Number of data points used to calculate geometric mean.

**ND - not detected above background levels.

concentrations and the concentrations of Rn in water were distributed log-normally, and a ratio of 4 pCi/l (bedroom air) per 10^5 pCi/l (water)

was found by regression. The application of this factor to the geometric mean of the water distribution led to a predicted increment of 0.23

Table 11. Indoor Rn concentrations (pCi/l) in "background" U.S. dwellings

Site	Number of Dwellings	Geom. Mean	Geom. S.D.	Eql. Ratio	Comments	Reference
New York						
New Jersey						
Basements	18	1.7	2.0	0.48	Ordinary Houses	Ge80
1st Floor	18	0.83	2.0	0.49		
2nd Floor	9	0.77	1.8	0.45		
Central Maine					Ordinary Houses	Pr83a
Basements	77	2.46	2.4			
1st Floor	82	1.40	2.4			
Bedroom (2nd)	81	1.12	2.4			
Bathroom	81	1.62	2.4			
Outside	67	0.46	2.4			
Houston, Texas					Houses, Apts.	Pr83a
Bedroom	103	0.39	2.5			
Bathroom	103	0.58	2.5			
Outdoors	81	0.22	2.6			
Eastern Pennsylvania						Sa81
Living Area	36					
Summer		1.22	4.6			
Winter		4.40	3.7			
Basement	36					
Summer		3.40	6.4			
Winter		5.90	4.0			

pCi/l in the air vs a net bedroom concentration of 0.66 pCi/l. This is an upper limit estimate, based on the assumption that the magnitudes of other sources of Rn are independent of the amount of Rn in the water. In fact, the concentration of Rn in the basements was found to be correlated with the concentrations of Rn in water at the $p = 0.01$ level.

Construction materials. A number of recent publications have addressed the problem of elevated indoor Rn concentrations arising from the use of building materials containing ^{226}Ra (Br81). A portion of the Rn arising from construction materials is able to diffuse into the living area, where the ultimate concentration increment will depend on the volume of the dwelling and the ventilation rate ranging from

.003 pCi/l to 0.3 pCi/l in the United States (Br81).

Perhaps the best publicized case involving construction materials occurred in Grand Junction, CO, where U-mill tailings were once frequently used as fill materials around foundations. More than 5,000 buildings were associated with tailings material to some extent, and 3,000 of those buildings were built on top of a layer of tailings. When Rn levels in some of the dwellings were found to be markedly elevated a general survey was conducted, and eventually a remedial action program was implemented for those structures exceeding national standards. (See Table 6).

A similar situation arose in the phosphate-mining area of Florida. Dwellings were built on mining lands reclaimed at least in part with

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Table 12. Factor relating Rn in indoor air to Rn in water (pCi/l in air per 10 pCi/l in water)

Factor	Reference	Conditions
14	(Ka80)	Calculated value for typical Finnish single and double-family structures, based on experimentally determined radon releases.
	(Ge80)	Calculations based on 4 occupants, experimentally determined Rn releases.
		Volume Air Change Rate
50		15000 l 0.25 hr ⁻¹
10		34000 l 0.50 hr ⁻¹
5		34000 l 1.00 hr ⁻¹
4	(Pr83a)	Observation obtained by regression from 3 months' integrated data from 80 houses in Maine, causality not strictly implied.
< 1	(Mc80)	Air and water grab samples in 13 structures in Halifax, Nova Scotia.

phosphate-rock residues. Radon concentrations well over 10 pCi/l have been noted in buildings containing materials such as gypsum wall board derived from phosphate residues or cinderblocks made of fly ash or blast furnace slag (US79).

Fuel gas. Radioactivity in natural gas was noted as early as 1904 (Sa18), but it was not until the 1970s that the potential contribution to indoor Rn concentrations was seriously investigated. Interest in the health implications of naturally occurring Rn in gas deposits was stimulated by investigations of the practicality of stimulating natural gas yields by detonating a nuclear warhead in the appropriate rock formation (Bu66). Estimates of dose increments caused by the combustion of natural gas in the home were made by Barton (Ba73), Johnson (Jo73) and Gesell (Ge74). Similar calculations were made for liquified petroleum gas (LPG), which, because of boiling point considerations,

contains a higher concentration of Rn than the natural gas from which it is made.

The increment to the indoor environment depends on the amount of gas of LPG burned in unvented ranges or heaters, the size and infiltration characteristics of the dwelling, and of course, the concentration of Rn in the fuel. The contribution from this source is usually quite small (0.15 pCi/l) due to the low use of unvented heaters (Ge77).

Occurrence of Rn in indoor air

There are a number of situations in which indoor Rn levels are especially elevated. These situations occur when the structure contains a stronger than usual source of Rn, or when the structure has especially low ventilation and infiltration rates or both. Rising heating and air conditioning costs in the last several years have

encouraged people to reduce air infiltration rates.

A number of trends can be discerned in the recent literature. Within a given locale, indoor Rn concentrations tend to be distributed log-normally, and sample means vary markedly from area to area (Ge83). It is becoming increasingly apparent that local geological factors play a major, if not dominant role in determining the distribution of indoor Rn concentrations in a given area.

Concentrations of Rn in the indoor environment. One of the most extensive studies of Rn in dwellings on record is a recent survey of 12,000 Swedish homes (Hi81). All the dwellings involved were ones in which elevated Rn levels were expected. The results as shown in Table 13 and 14 are reported in working levels, and the presumed associated Rn concentrations, based on an equilibrium ratio of 0.5, are added in parentheses.

The measurements summarized above are not meant to be representative and are probably considerably higher than the true area average. Nevertheless, the number of dwellings involved and the high values observed combine to demonstrate the extent of the Rn problem in certain areas. Nearly half of the 12,000 dwellings were found to exceed current U.S. occupational standard for U mines (adjusted for constant occupancy vs a 40-hr week).

Another extensive survey involved nearly 10,000 randomly selected houses in 14 Canadian cities (Mc80). Single grab samples were obtained from basements or the ground floor during the summer months. In general, data from a given city were better fit by the log-normal distribution than by the normal distribution. The geometric means ranged from 0.14 pCi/l in Vancouver to 0.88 pCi/l in St. Lawrence, Newfoundland. The geometric standard deviations ranged from 2.78-6.77.

Table 11 summarizes a number of surveys conducted in the United States in areas not known or suspected to involve anomalies due to mill tailings or unusual mineralization. The data presented are either the average of a number of grab samples taken within a single dwelling, or were developed by long-term measuring devices. The equilibrium fraction (f) of Rn progeny is given where available.

Radon conclusions

(1) Radon concentrations in water are highest in ground water especially in granite areas. Radon concentrations in surface water are very low.

(2) Higher concentration occurs in small systems. Domestic supplies have higher concentrations of Rn in water than public wells. Utility systems are lower by a factor of 10 than private wells.

Table 13. Radon levels measured in air in houses

	Average pCi/l	Maximum pCi/l	Number of Houses
Maine (He82b)	3.0	63.0	85
New York			
Albany Area (F180)	3.1	26.0	21
New York City Area (Br79)	1.0	5.9	21
California			
San Francisco (Ne81)	0.4	1.1	28
Pennsylvania			
Eastern (Sa81)	10		36
Maryland (Mo81)	3.7	27.0	56

Table 14. Radon progeny in Swedish dwellings (Hi81)

Radon Progeny (Working Levels)	Radon (pCi/l, f=0.5)	Number of Dwellings	Percent of Dwellings
0.000 - 0.054	0 - 11	6326	52
0.054 - 0.108	11 - 22	4050	34
0.108 - 0.270	22 - 54	1545	13
> 0.270	> 54	162	1

(3) The highest average Rn concentrations in water are found in the provinces in decreasing order: New England, Appalachian Highlands-Piedmont, Pacific Mountain System, and Rocky Mountain System.

(4) Ventilation affects Rn concentration in air with an approximate value of $1. \times 10^{-4}$ for the ratio of Rn concentration in air to Rn concentration in water for a house with one air change per hour. Soil gas Rn contributes a sizeable portion of the total Rn in air.

(5) Additional measurements of Rn in systems serving less than a thousand users are required in order to better quantify exposures to the group that potentially represents the highest population dose. Because the number of such systems is quite large (~37,000), these measurements should be obtained from a representative sampling program guided by geological models.

CALCULATION OF WATER UTILITY RISKS FROM RADIONUCLIDES IN WATER BASED ON NATIONAL OCCURRENCE DATA

Each nuclide present in drinking water will present a risk to the utility users of the water which is related linearly to the occurrence, concentration, population exposed, and the individual risk rate (Ma83). The case of Ra in drinking water data allows a direct calculation. The average concentration of Ra in drinking water from utilities is 1.6 pCi/l (Ho83), and the population consuming this water is 70 million people which is the half of the U.S. population which uses ground water provided by utilities. Since the individual risk rate is hypothesized to

be by linear dose response 8.4×10^{-6} excess cancers/lifetime person pCi/l (using (Ma83), we can calculate

$$1.6 \text{ pCi/l} \times 8.4$$

$$\times 10^{-6} \frac{\text{excess cancers}}{\text{lifetime person}} \frac{1}{1 \text{ pCi/l}} \times 70$$

$$\times 10^6 \text{ people} \sim \frac{941 \text{ people}}{\text{lifetime in U.S.}}$$

Even the elimination of bone sarcomas at low concentration will leave the sinus carcinomas which are half of this number (Ma83). An additional 30×10^6 people are exposed at less than 0.5 pCi/l surface water provided by utilities. The distribution of occurrence of Ra concentrations permits estimates of the number of fatal cancers averted when the standard is placed at a particular concentration. Since the standard is at 5 pCi/l, it seems reasonable to estimate the fraction of cancers averted by the standard. The average for the supplies of greater than 5 pCi/l is 8 pCi/l. This concentration is multiplied by an estimate of the population which uses those supplies obtained from rounding the average ground water utility population from the U.S. Environmental Protection Agency summary. See Fig. 7 for the number of utilities vs Rn concentration.

$$8 \text{ pCi/l} \times \frac{1000 \text{ persons}}{\text{supply}} \times 500$$

$$\times \frac{8.4 \times 10^{-6} \text{ excess cancers}}{1 \text{ pCi/l person lifetime}}$$

$$\sim 33 \frac{\text{excess cancers}}{\text{lifetime}}$$

Number of Houses
85
21
21
28
36
56

Population risk can also be done for U in water by using a similar calculation. For U the population-weighted average radioactivity concentration is 0.8 pCi/l for the whole U.S. population (Dr81). This permits this result for the risk using individual risk factors from Ma83, Wr83 and Co83.

$$[0.8 \text{ pCi/l}] \times \frac{6 \times 10^{-7} \text{ cancers}}{\text{pCi/l person}} \times 220 \\ \times 10^6 \text{ people exposed} = \frac{105 \text{ persons}}{\text{lifetime in U.S.}}$$

Since there is no drinking water standard for U, at this time, we must use the fraction of people at each occurrence level to estimate the number of cases avoided by a standard for water utilities

Using values from Drury *et al.*, a water utility users' risk estimate table can be compiled (Dr81) (See Table 15.)

The population risk for Rn in utility water supplies can be calculated using the Rn concentration in utility water, the exchange rate from water into air, the cancer rate per working level month per million population and the exposed population in millions. From the work done by the Committee on the Health Effects of Radon in drinking water (Cr83), we can obtain the individual risk factor which includes the exchange rate for Rn from water into air and the cancer rate per working level month per million

population exposed to air for a lifetime of 20 yr. This factor is 3×10^{-7} lung cancers per pCi/l Rn in water. This factor is increased to 4×10^{-7} cancers when stomach and whole body cancers are included (Cr83). The Rn in water concentration data for the whole United States are geologically controlled and are generally a mixture of low values around 100–200 pCi/l and high values of 10,000–1,000,000 pCi/l. This extreme range of values leads to arithmetic averages which are strongly influenced by the highest few points. The geometric mean of these data will average the numbers with less weight for the high values. This geometric mean will be lower than the arithmetic mean. We have decided to calculate the risk with both of these means.

Using data from the geometrical and arithmetic means of Rn concentrations for utilities of different sizes, and the geometrical and arithmetic risk factor, we can calculate the population at risk for the utilities serving less than 100 population, 100–1000 population, 1000–5000 population, 5000–10,000 population, 10,000–100,000 population, and 100,000 and above population. Lifetime risk is shown for both geometric averages and arithmetic averages in the right column of Table 16.

OTHER NATURAL RADIOISOTOPES OF POSSIBLE CONCERN

The natural radioisotopes already discussed above (U, ^{228}Ra , ^{226}Ra and ^{222}Rn) are of greatest concern because of their long half-lives and the

Table 15. Cases prevented for U concentrations in public water

pCi/l	% Supplies	Number Supplies	People Exposed*	Average U Concentration	Cases
> 1 pCi	23.6	10,808	10.8×10^6	7.5	48
> 5 pCi/l	7.88	3,609	3.6×10^6	17.5	38
> 10 pCi/l	3.46	1,584	1.5×10^6	30.3	27
> 20 pCi/l	1.33	609	0.6×10^6	54.8	20

*1000/supply

Table 16. Assessment of water risks for Rn in public ground water

Population	Number of Utilities Sampled	Mean Concentration Of Radon in Water (pCi/l)	U.S. Population Using This Water (millions)	Lifetime Risk
		<u>GEOM - ARITH</u>		<u>GEOM - ARITH</u>
< 100	88	990 - 6500	1.03	407 - 2678
100 - 1000	377	620 - 4100	7.4	1835 - 12136
1000 - 5000	1223	98 - 390	14.0	548 - 2184
5000 - 10,000	549	92 - 350	8.4	309 - 1176
10,000 - 100,000	704	92 - 290	28.3	1018 - 3283
> 100,000	32	52 - 150	<u>14.3</u> 59.13	297 - 858 4414 - 22315

Population-weighted Average 187.0 pCi/l - 944.0 pCi/l.

FACTOR USED: $\frac{0.4 \times 10^{-6} \text{ deaths}}{\text{pCi/l water}}$

health risks associated with the activities that can be present in public drinking water. However, there are two classes of other natural radioisotopes that may be of possible concern: (1) relatively long-lived isotopes whose activity is derived from the aquifer, termed unsupported (^{232}Th , ^{230}Th , ^{210}Pb and ^{210}Po) and (2) very short-lived isotopes which "grow in" once the ground water is pumped from the aquifer and thus are supported (primarily ^{222}Rn progeny). However, very few data are available on the activities of these isotopes in drinking water, primarily due to their low solubility and/or the difficulty of measuring isotopes of short half-lives.

Table 17 shows the typical ranges of activities of the longer-lived radionuclides in ground water. Activities in surface waters will be extremely low (except perhaps in hot springs) due to rapid sorption onto suspended and bottom sediments. The highest known activity of ^{232}Th and ^{230}Th in U.S. drinking water is from a well in California that also contained large amounts of

dissolved organic matter which probably complexed with Th. Most other values are below 0.1 pCi/l. Thorium-230 would be expected to be slightly higher than ^{232}Th due to generation by ^{234}U in solution and by α recoil. Likewise, detectable activities of ^{210}Pb and ^{210}Po would be expected because of the relatively large amount of ^{222}Rn present in many ground waters. The insolubility of these isotopes and their short-lived precursors in the aquifer is demonstrated by the fact that more than 99.9% of the activity generated by ^{222}Rn decay in ground water is removed within the 2 hr necessary for equilibrium to be established between ^{222}Rn and ^{210}Pb . The only known anonymously high ^{210}Po value in drinking water is the surprisingly large activity in Louisiana for which the source has not been determined. In general, these longer-lived isotopes are not expected to occur in activities greater than 1.0 pCi/l.

The second class of radioisotopes of possible concern are ^{222}Rn progeny which reach equilibrium with ^{222}Rn within 2 hr. In untreated

Table 17. Concentrations of Th, Pb and Po isotopes in ground water (pCi/l)

Description	Th-232	Th-230	Pb-210	Po-210	Reference
<u>New Mexico</u>					
Grants Mineral Belt (54 wells)					
Paquate-Jackpile Area	<0.01	<0.02		0.39	Ka 76
Grants-Bluewater Area	<0.01	0-0.04		0-0.66	Ka 76
United Nuclear Area	0-0.03	0-0.099		0.3-2.3	Ka 76
Ambrosia Lake	<0.03	0-0.08		0-3.8	Ka 76
Gullup Area	<0.02	0-0.09		0-0.6	Ka 76
Rapides Parish					
Louisiana (1 well)				290-607	Mu 82
California well	1.3	1.1		ND	This paper
Arizona well	ND*	ND		0.9	This paper
Connecticut					
glacial drift			0.02	<0.001	Kris 82
glacial drift			0.02	0.004	Kris 82
crystalline rock			0.03	0.06	Kris 82
sandstone			0.12	0.020	Kris 82
sandstone			0.07	0.005	Kris 82
sandstone			0.03	0.004	Kris 82
Leesville, S.C.					
sand aquifer		0.01			This paper
ND = not detected					

ground-water systems, removal by adsorption would not be as rapid as in the aquifer due to the small surface area of the distribution system. Exposure from consumption of these supported, extremely short-lived progeny should be evaluated. Radon-220 and its progeny do not pose a similar problem primarily because the 54.5-sec half-life of ^{220}Rn is too short to allow diffusion from the aquifer materials and the initial activities are much lower.

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PREDICTING THE OCCURRENCE OF ^{228}Ra IN GROUND WATER*

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Abstract—A conceptual model is presented to predict the relative probability of the occurrence of elevated ^{228}Ra in public drinking water supplies (those which serve more than 25 people or 15 connections) using ground water sources. The model is based on an aquifer classification scheme, which is developed from an understanding of the geochemical and radiological behavior of ^{228}Ra and its parent, ^{232}Th . Using this model, all aquifer types are classified as low, medium, or high probability of having elevated ^{228}Ra . Summaries of the available data are discussed to show the actual concentrations found in each type of aquifer. As part of the initial application of this approach to develop a nationwide occurrence profile for ^{228}Ra , all counties in the United States were classified and a map presented to show the distribution of the three classes. Nationwide, 71% of the counties were ranked as low, 18% were ranked as medium, and 11% were ranked as high.

INTRODUCTION

IN 1976, the U.S. Environmental Protection Agency (EPA) established the national interim primary regulations for radioactivity in drinking water. These regulations set maximum contaminants levels (MCLs) for gross α -particle activity of 555 becquerels per cubic meter (Bq m^{-3}) or 15 picocuries per liter (pCi L^{-1}) and total radium activity (^{226}Ra plus ^{228}Ra) of 185 Bq m^{-3} (5 pCi L^{-1}). The ^{226}Ra activity measurement was required only when the gross α -particle activity exceeded 185 Bq m^{-3} (5 pCi L^{-1}) and ^{228}Ra measurement was required only when the ^{226}Ra activity exceeded 111 Bq m^{-3} (3 pCi L^{-1}). This screening protocol and the combination of Ra isotopes was proposed because it was believed that ^{226}Ra would be the dominant isotope in

public drinking water supplies and would avoid the successively more expensive ^{228}Ra analyses. Very little was known about the geographic distribution of ^{228}Ra activity in drinking water, even after the initial monitoring program was completed. Use of the monitoring results to better predict the occurrence of ^{228}Ra was difficult because the monitoring protocol required by the regulation produced a very biased database.

The monitoring program did identify some regions which had high natural radioactivity and these have been studied in detail by several groups (Ro77; Mi80; Ma81; Ki82; Kri82; Cl83; Gi84; Lu85). The results of these studies and an EPA-sponsored workshop on radioactivity in drinking water were summarized in Hess *et al.* (He85). One recommendation of the workshop group was that monitoring requirements for ^{228}Ra should be decoupled from ^{226}Ra and separate guidelines for ^{228}Ra were needed. The coupling of these isotopes was not justified because of the differences in the geochemical behavior of their parent iso-

* The thoughts and ideas expressed in this paper are those of the authors and do not necessarily reflect the policies of the U.S. Environmental Protection Agency.

topes. In fact, the monitoring scheme required by the regulations could even be biased away from areas of elevated ^{228}Ra where ^{226}Ra had been selectively removed because of the mobility of the parent U.

In the development of the Revised National Drinking Water Regulations, the EPA needs some way to estimate the occurrence of ^{228}Ra before formulation of criteria for the regulations. Without better occurrence data, it would not be possible to calculate population risks and treatment costs, or develop monitoring requirements. However, the existing data on ^{228}Ra were inadequate to even attempt any estimates based on extrapolation of available measurements. That analytical methods for measuring ^{228}Ra in water samples are difficult, time-consuming and costly are facts which limit the effectiveness of a large-scale monitoring program to actually measure levels of ^{228}Ra in drinking water. Also, the distribution of ^{228}Ra has been shown to be log-normal (Ki82), with most values below the present detection limit of 37 Bq m^{-3} (1 pCi L^{-1}).

The EPA is currently sampling about 1,200 randomly-selected ground water sources of drinking water in the United States. Public drinking water supplies are those that serve more than 25 people or 15 connections. There are about 48,000 such ground water supplies in the United States. Therefore, the effectiveness of this random program would be limited in predicting different exposure levels because of the small number of samples that are expected to occur above the detection limit. Without a new approach to estimate the occurrence of ^{228}Ra , human health and economic effects of revision of the drinking water regulation would be difficult to evaluate.

In this paper, an approach is presented which uses aquifer type and water-quality characteristics to classify specific areas according to the relative probability of having high ^{228}Ra in ground water. This concept is based on the fact that ^{228}Ra is not randomly distributed and thus cannot be predicted using statistical analysis of national survey data alone. There will be too few "hits" of the high values to be of use in identifying, to any degree of certainty, the true distribution of ^{228}Ra or detecting systems with high ^{228}Ra using random sampling techniques. Instead, the United States can be divided into smaller units which

are classified on a relative scale of low, medium, and high probability of including elevated ^{228}Ra concentrations. The classifications are produced from a conceptual model of the most important factors affecting ^{228}Ra distribution in ground water.

OCCURRENCE OF ^{228}Ra BY AQUIFER TYPE AND WATER QUALITY

The correlation of high ^{228}Ra concentrations in ground water with certain types of aquifers has been observed by many researchers, beginning with Krause (Kra59) who reported high ^{226}Ra and ^{228}Ra in the ground water from deep sandstone formations in midwestern states. Even then, Krause concluded that the $^{226}\text{Ra}/^{228}\text{Ra}$ activity ratio was not constant, and in order to calculate the total radiation dose, the ^{228}Ra content of the water would have to be determined separately. After the interim standards were set, detailed studies of ^{226}Ra and ^{228}Ra were conducted in Iowa (Kri83), South Carolina (Mi80; Ki82), and Georgia (Cl83). Also, the monitoring results provided a new source for ^{228}Ra values.

In 1982-83, the EPA sponsored two studies to determine the feasibility of using aquifer type and water-quality characteristics to predict the occurrence of ^{228}Ra in ground water (Mi82; Mi83). In these studies, an aquifer classification scheme was developed and tested on three major hydrogeological provinces, namely the Atlantic Coastal Plain, the Piedmont, and part of the glaciated Central Platform. The Atlantic Coastal Plain and Piedmont are two adjacent provinces which extend from New York to Alabama and east of the Appalachian Mountains to the coast. The parts of the glaciated Central Platform studied included the states of Wisconsin, Iowa, Illinois, and Missouri. These regions were selected for development of the predictive model because they had the largest existing database of ^{228}Ra measurements, which indicated that there were areas of both very high and very low ^{228}Ra concentrations in these regions.

The aquifer classification model was developed from an understanding of the geochemical and radiological properties of ^{228}Ra and its parent ^{232}Th within an overall framework of the geological processes which affect the composition of aquifers. Discussions of the model are given elsewhere (Mi82; Mi83; He85). The results of these

studies showed that there were distinct differences in the relative distribution of ^{228}Ra by aquifer type and water quality. Table 1 shows the summary statistics for the specific aquifers studied in detail during earlier studies. These results were used to develop the following aquifer classification model for predicting the ^{228}Ra content of ground water.

Aquifers which always had low activities of ^{228}Ra included the following:

(1) *Carbonate aquifers* (limestone and dolomite) had a log mean ^{228}Ra of 11 Bq m^{-3} (0.3 pCi L^{-1}) for 120 samples. These rock types have very low ^{232}Th content because they precipitate from water, and Th has very low solubility in water.

(2) *Metamorphic rock aquifers* also had a log mean of 11 Bq m^{-3} (0.3 pCi L^{-1}) for 75 samples. Metamorphism of all intensities and rock types tend to recrystallize any loosely-bound Th into resistant minerals which minimizes the ability of daughter ^{228}Ra to go into solution. Also, ground

water flow in these rock types is generally through fractures instead of through pore spaces, which decreases the rock surface area in contact with ground water.

(3) *Quartzose sand and sandstone aquifers* generally produced ground water with low ^{228}Ra , particularly if the total dissolved solids (TDS) were below 1,000 milligrams per liter (mg L^{-1}). The high quartz content of the aquifer material indicates that the sediments were intensely weathered or that they were eroded from a source which initially had low Th content. In either case, the only Th contained in the aquifer material is likely to be in heavy minerals which do not readily give up ^{228}Ra into solution. There are two special subclasses of this aquifer type. Sand and gravel aquifers of glacial origin are generally low in ^{228}Ra [log mean of 22 Bq m^{-3} (0.6 pCi L^{-1}) for 17 samples] regardless of their composition because of the large size of the clasts and the low ratio of surface area to water. Even when these aquifers are poorly sorted (i.e., contain various amounts of clay, sand, and gravel), the clay tends

Table 1. Summary statistics for ^{228}Ra content of ground water from specific aquifer types

Aquifer Type	N	\bar{x}		95% Upper Confidence Limits	
		Bq m^{-3}	pCi L^{-1}	Bq m^{-3}	pCi L^{-1}
Igneous Rocks (Granites)	42	52	1.4	590	16.0
Chemical Precipitates					
- Coastal Plain Aquifers	16	4	0.1	7	0.2
- Midwest Aquifers	104	22	0.6	107	2.9
Metamorphic Rocks					
- Piedmont Region	75	11	0.3	92	2.5
Sand Aquifers					
- Coastal Plain Fall Line Arkoses	89	81	2.2	630	17.0
- Coastal Plain - Quartzose	53	11	0.3	89	2.4
- Midwest Glacial/Alluvial	135	26	0.7	160	4.2
Sandstone Aquifers					
- Midwest Cambrian - Ordovician Aquifer	320	78	2.1	440	12.0

to rapidly adsorb radium. Sand and gravel aquifers of alluvial origins (such as those found in river valleys) also were found to have low ^{228}Ra , with a log mean of 26 Bq m^{-3} (0.7 pCi L^{-1}) for 118 samples. These aquifer types usually contain quartz-rich sediments and produce large quantities of water.

(4) *Basic, igneous rock aquifers* (such as basalt) are classified as having very low ^{228}Ra although there are few data from this type. Basalts have low Th content but, more importantly, this rock type rapidly sorbs ^{228}Ra from solution. Basalts are so efficient at sorption of metals from water that they are considered as one of the candidate rock types for long-term disposal of high-level radioactive wastes.

There are three main aquifer types which are predicted to have a high probability of producing ground water with elevated ^{228}Ra .

(1) *Granite rock aquifers* which had not undergone any metamorphism had a log-mean ^{228}Ra concentration of 52 Bq m^{-3} (1.4 pCi L^{-1}) and a range up to 850 Bq m^{-3} (23 pCi L^{-1}) for 42 samples. Granites are composed of large amounts of coarse-grained feldspar, a mineral which contains thorium, not always in resistant, crystalline forms but often loosely bound at sites between individual grains. Radium-228 produced from this type of Th distribution is more likely to go into and stay in solution. Igneous rocks of similar composition but fine-grained, such as light-colored volcanic rocks, would have low ^{228}Ra because of rapid resorption of ^{228}Ra from solution.

(2) *Arkosic sand and sandstone aquifers* had medium to high ^{228}Ra , with a log mean of 90 Bq m^{-3} (2.4 pCi L^{-1}) for 89 samples. Arkoses, by definition, contain a significant amount of feldspathic minerals, which can have high Th. If the arkosic sediments were derived from granitic rocks, they can have very high Th. If the sediments have undergone any weathering which has broken down the feldspar, the ^{228}Ra content of the ground water can be very high. There is a gradient of the ^{228}Ra content of these aquifers depending upon the amount and composition of feldspathic minerals and rock fragments in the sediments of the aquifer.

(3) *Quartzose sandstone aquifers with high TDS* were found to have a wide range of ^{228}Ra ,

including very high concentrations up to 1180 Bq m^{-3} (32 pCi L^{-1}). All data for this type of aquifer came from groundwater supplies in Iowa, Illinois, and Missouri, which had a mean ^{228}Ra concentration of 74 Bq m^{-3} (2 pCi L^{-1}) and usually had high ^{226}Ra as well. It was concluded that, at TDS of greater than $1,000 \text{ mg L}^{-1}$ in clean, quartz sandstones, competition for the few available sorption sites on the aquifer solids were the controlling factor keeping ^{228}Ra in solution. This aquifer type may be one of the most overlooked sources of elevated ^{228}Ra , especially in areas where U has been removed by ground water leaching. With U removed, the gross α -particle activity screen would be low and neither ^{226}Ra nor ^{228}Ra would be measured.

The purpose of this paper is to demonstrate a concept of an aquifer classification scheme to map areas nationwide on a relative scale of low, medium and high probability of having elevated ^{228}Ra concentrations. It should be noted that areas ranked high are not predicted to have only elevated ^{228}Ra in the ground water. All areas will have a large percentage of the community water supplies with very low ^{228}Ra in the ground water. Instead, these areas ranked high are more likely to have elevated ^{228}Ra occur more frequently. The mapping unit selected was by county, the smallest unit for which data on population and number of systems could be compiled. The EPA is currently conducting a field-monitoring program in which 1,200 community drinking water supplies are being sampled for various parameters, including ^{228}Ra . The results from this survey will provide the data to develop an occurrence profile for each class.

METHODS OF STUDY

To delineate aquifer types nationwide using the proposed classification scheme for 3,076 counties, many various sources of information were used. State and federal agencies responsible for geological, mineral, and water resources were contacted for pertinent literature and geological and hydrological maps. One of the most difficult tasks was identification of which geological units were used as aquifers.

After all the available data were reviewed, the main aquifers (not just rock types) for each state and county were identified, described, and clas-

sified as having a low, medium, or high probability of having elevated ^{228}Ra concentrations. For counties with multiple aquifer types, either areally or with depth, professional judgment was used in the final classification. For multiple aquifers with depth, the county was classified according to the aquifer of the highest classification. In some cases of areally different aquifers within a county, where information was available on population distribution from detailed maps, the dominant aquifer type most probably used for public water supplies determined the county classification.

Aquifers classified as low probability included those composed of limestone, metamorphic rocks (including high-grade metamorphosed granites), glacial and alluvial sand and gravel, volcanic rocks, and quartzose sand and sandstone of low-to-moderate TDS. Aquifers classified as medium probability included those composed of semi-arkosic sand and sandstone, low-grade metamorphic granites, quartzose sand and sandstone of moderate TDS, alluvial sand and gravels specifically described as containing feldspathic sedi-

ments, and a relatively small set of aquifers which could not be definitely classified as low or high. Aquifers classified as high probability included those composed of granite, arkosic sand and sandstone, alluvial deposits located immediately adjacent to granitic rock sources, and quartzose sandstones with high TDS, particularly in those areas where high ^{228}Ra has been reported.

RESULTS AND DISCUSSION

Figure 1 shows the areal distribution of the three different classes for the counties in the continental United States. All other states and territories were ranked as having low probabilities. Nationwide, 71% of the counties were ranked as low, 18% were ranked as medium, and 11% were ranked as high. It should be noted that in reviewing these data, that counties varied in size (by area) by over three orders of magnitude and in population by at least a factor of 3,000. Counties are by no means uniform units for direct comparison of population at risk or number of systems impacted. The EPA is currently compiling

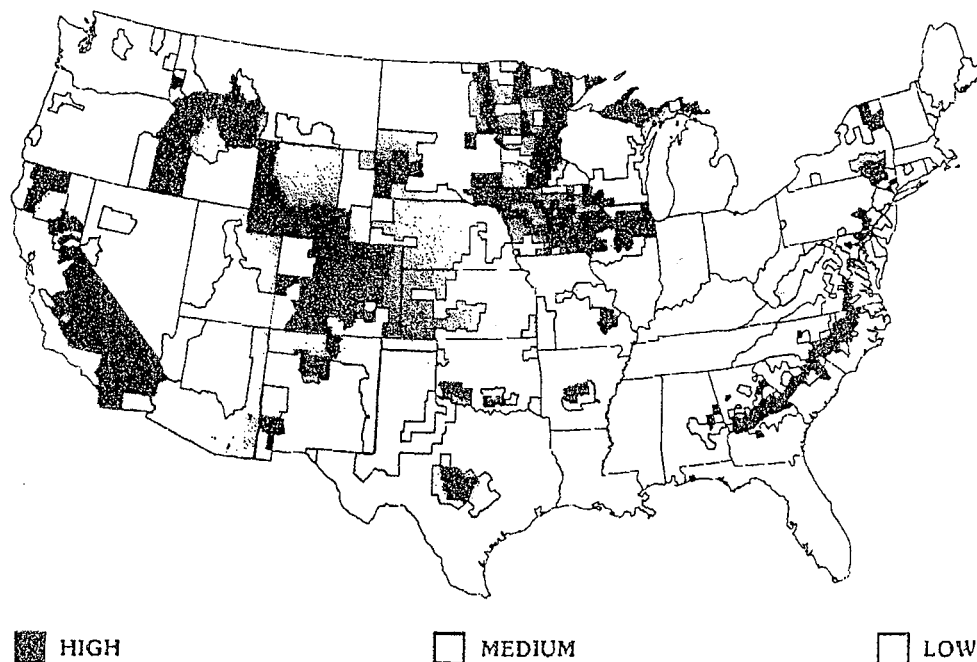


FIG. 1. Distribution of areas of relative probability of having elevated ^{228}Ra in community ground water supplies.

this information by county so that the actual population and number of systems can be determined at the county level. It should be pointed out that the population exposed in any one county may vary from nearly all to only a small portion of the total.

Even with a brief review of Fig. 1, some interesting observations can be made. There are large areas where ^{228}Ra is expected to be very low, with most values below 37 Bq m^{-3} (1.0 pCi L^{-1}) and probably below 18 Bq m^{-3} (0.5 pCi L^{-1}). These areas include the quartzose sand and limestone aquifers of the lower Coastal Plain along the Atlantic and Gulf coasts. The U mineralization belt in Texas is ranked low because much of the U is leached from volcanic rocks which do not release Th. The very old carbonate and quartz-rich sandstone aquifers underlying the general area between the Appalachians and the High Plains were ranked low except for localized areas, such as the Oachita Mountains in Arkansas and the St. Francois Mountains in Missouri. Much of the area ranked low in the Northeast is composed of metamorphic rock terrains of low ground water production, overlain by glacial sand and gravel aquifers, from which most of the ground water is obtained. In the West and Northwest, low-ranked areas were predominantly basalt, alluvial sand and gravel, and clean sandstones.

Areas which were ranked as high included a band from Pennsylvania to Georgia which represented mostly the Fall Line sand aquifers which are located at the contact between the rock terrain of the Piedmont province and the sediments of the Coastal Plain. These aquifers have been shown to have high ^{228}Ra and ^{226}Ra (Mi80). This zone also included the granitic rock aquifers in the adjacent Piedmont which are used by small suppliers.

Large areas of northern Illinois, Iowa, Minnesota, and Wisconsin were ranked as high, because of granitic terrain or the presence of sandstone aquifers with high TDS. Counties ranked low or medium in this area were those where ground water was obtained mostly from alluvial or glacial sand and gravel aquifers or from carbonate aquifers. It should be noted that this area is mapped in great detail because it was one of the detailed study sites used in developing the aquifer type model.

Parts of Colorado and Montana were ranked high where the granites of the Rocky Mountains might influence the ^{228}Ra content of the aquifers. However, the ground water use in this area should be small, and therefore, the relatively large area ranked as high may not represent many users. In Idaho, the high areas are those underlain by the extensive Idaho granite. The granite functions as a poor aquifer, but it could have an impact on radiological water quality in that the granite is a significant source of the sediments that fill the alluvial valleys which are important aquifers.

Much of California is underlain by granitic igneous rocks which were ranked high, although community ground water use in these areas is probably small. The complicated hydrogeology of California, which is composed of 394 ground water basins with no widespread aquifer systems, made classification of the region difficult and the results very weak. The rest of the areas ranked as high risk were scattered occurrences where granitic intrusions or sediments derived from them constituted local aquifers.

There were several large areas designated as having medium probability, the largest of which was the High Plains area, underlain by a shallow sand aquifer known regionally as the Ogallala. This aquifer supplies much of the ground water for irrigation of farms from Texas to South Dakota. The sediments of this aquifer are thought to have been eroded from the granite of the Rocky Mountains and deposited by streams draining east. The sediments can contain significant amounts of feldspathic minerals locally and the entire aquifer is ranked as medium. There are very few ^{228}Ra measurements from this aquifer to indicate what the actual concentrations are.

The remaining large area of medium probability (parts of Idaho, Montana, Utah, and Arizona) are counties where granitic rocks were thought to have contributed some but not significantly to the sediments of the alluvial valley and sandstone aquifers.

No attempt was made to quantify the relative rankings of low, medium, and high. With the existing ^{228}Ra data, which are nonrandom and of variable quality, and for which the aquifer type is frequently unknown, it is not possible to calculate occurrence statistics with any degree of certainty. It is hoped that the current EPA ground

water survey will provide some indication of the actual distribution and confirmation of the relative rankings. If so, this approach can be refined if necessary and used in better determining the population risk, the costs of compliance, and monitoring guidelines. The map in Fig. 1 could be used to identify specific areas requiring different monitoring strategies.

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