Natural Radioactivity in Ground Water — a Review

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INTRODUCTION

Natural radioactivity and its effects on human health recently have become a major environmental concern because of the discovery of widespread occurrence of levels of radon in the air of homes at concentrations that exceed the U.S. Environmental Protection Agency's (EPA) recommended maximum levels, particularly in the eastern United States. Radon-222 in air, even in small concentrations, contributes to the high incidence of lung cancer among uranium miners in the Western United States (Archer and others, 1962). Recent estimates indicate that radon in indoor air may cause 5,000 to 20,000 lung-cancer fatalities annually in the United States (U.S. Environmental Protection Agency, 1986a).

A less publicized but also important health hazard is the presence in ground water of naturally occurring radioactive substances, which along with radon are known as radionuclides. In addition to radon, large concentrations of dissolved radium and uranium radionuclides have been detected in may ground-water supplies across the United States. All radionuclides dissolved in water are colorless, odorless, and tasteless and, thus, cannot be detected by our senses, unlike many water pollutants that may impart undesirable colors, odors, and tastes to water.

Although much is known about the theoretical geochemistry of radionuclides in ground water, it still is very difficult to forecast the amount of radionuclide activity in a particular ground-water supply because of the strong influence of local geologic, geochemical, and hydrogeologic conditions. Much of what is known about the distribution of radionuclides in water has been derived from analysis of water from public water-supply systems, which supply slightly more than 80 percent of the population in the United States (Solley and others, 1983). However, it is difficult to develop site-specific information about the occurrence and activities of radionuclides in specific aquifers because public water supplies commonly are a blend of water from numerous ground- and surface-water sources. Relatively little is known of the concentrations of radionuclides in private water supplies, which rely heavily on ground water and supply more than 20 percent of the total ground water used for human consumption.

In recent years, estimates have appeared in the scientific literature about the effects of radionuclides in ground water on human health. these estimates and the growing body of scientific knowledge of the distribution and levels of radionuclides in ground water, have stimulated a review of the adequacy of standards and regulations for radionuclides in drinking water. Fortunately, conventional methods for treating raw water for some other contaminants also are effective in removing radionuclides found in ground water. (See table 2.)

GEOCHEMISTRY OF RADIONUCLIDES

Radionuclides are found as trace elements in most rocks and soils and are formed principally by the radioactive decay of uranium-238 and thorium-232, which are the long-lived parent elements of the decay series that bear their names (fig. 18). The parent elements produce intermediate radioactive daughter elements with shorter half-lives (half-life is the time required for half of the initial amount of the radionuclide to decay). Decay occurs by the emission of an alpha particle (a nucleus of the helium atom) or a beta particle (an electron) and gamma rays from the nucleus of the radioactive element. The geochemical behavior of a daughter element in ground water may be quite different from that of the parent element. However, the parent may govern the occurrence and distribution of the daughter element.

The most common radionuclides in ground water are radon-222, radium-226, uranium-238, and uranium-234 of the uranium-238 decay series, and radium-228 of the thorium-232 decay series. Other radionuclides of these two decay series, and all isotopes of the uranium-235 decay series, generally are not present in significant amounts in ground water, because most are highly immobile and many have very short half-lives that preclude the buildup of large concentrations.

The occurrence and distribution of radionuclides in ground water is controlled primarily by the local geology and geochemistry. For daughter radionuclides to be present in large concentrations, the parent radionuclide must be present in the rock material composing the aquifer. Each radioactive decay product has its own unique chemical characteristics, solubility, mobility, and half-life, which can be very different from those of the parent. For this reason, parent and daughter radionuclides in ground water are not usually found together in similar amounts (Gilkeson and others, 1983, p. 22): nor do they decay at similar rates or produce the same level of radioactivity. Therefore, a high concentration of one radionuclide in ground water at a specific site does not necessarily imply that similar concentrations of other radionuclides in the same decay series are present. for example, the parent/daughter radionuclide pairs uranium-238 or uranium-234/radium-226 or radium-226/radon-222 usually are not present in high concentrations in the same ground water.

The movement of many radionuclides is very dependent upon the radionuclide's solubility in water. Uranium, which is most soluble in bicarbonate-rich oxidizing (oxygen-rich) ground water with low total dissolved-solids content, is easily dissolved and transported by oxidizing ground water; thus, it can be transported to areas far from its original emplacement. Solubility of uranium tends to be enhanced by association with carbonate, phosphate, and fluoride ions, or with organic compounds, especially humic substances (Langmuir, 1978, p. 556; Turner-Peterson, 1980, p. 163). Uranium is less mobile in reducing ground water, and it tends to be adsorbed very strongly onto humic substances in the aquifer. Conversely, radium is most mobile in chloride-rich reducing ground water with high total dissolved-solids content (Tanner, 1964, p. 261).

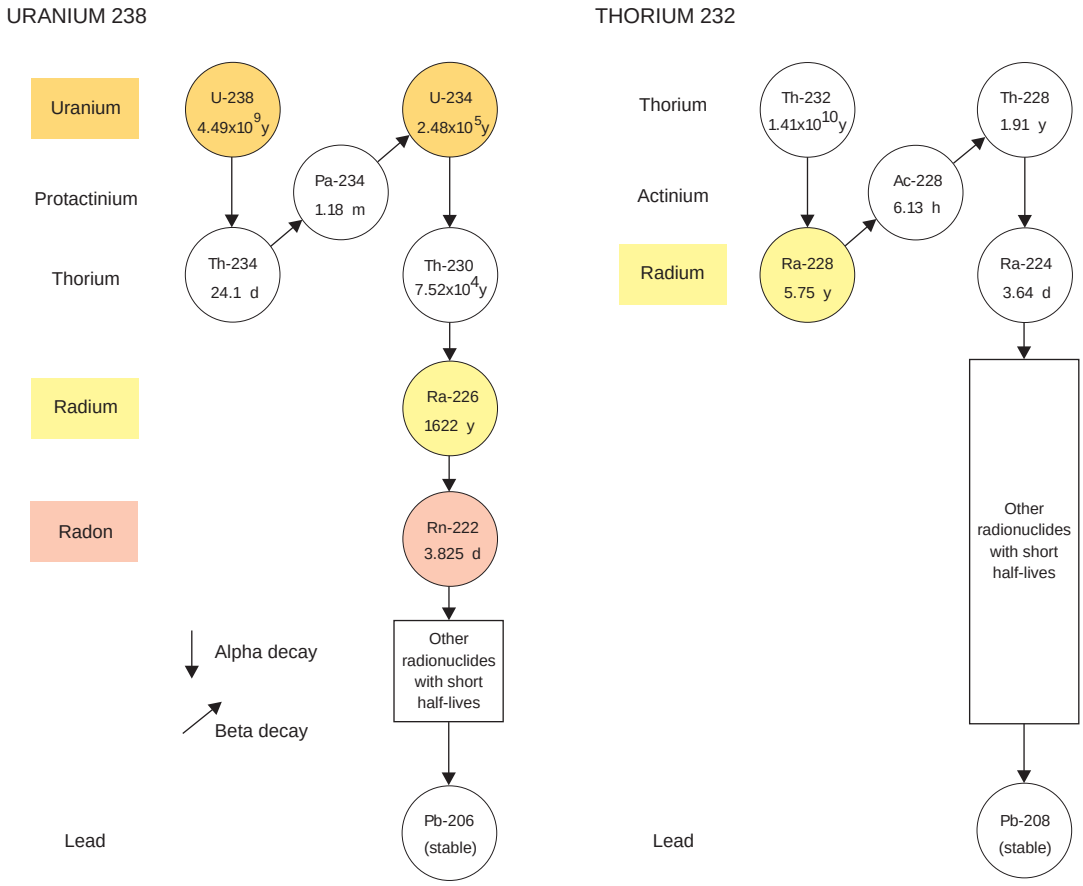

Figure 18. Uranium-238 and thorium-232 radioactive decay series. Colors indicate those radionuclides found most frequently in ground water. Times shown are half-lives – y=years, d=days, h=hours, m=minutes.

Field measurements or dissolved-oxygen concentration and oxidation-reduction potential (Eh) in parts of New Jersey have shown that these are important controls on radium-226 and uranium concentrations in ground water (Szabo and Zapecza, 1987). Where the ground water is reducing, elevated levels of radium-226 are associated with elevated gross alpha activities. Where the ground water is oxidizing, only small concentrations of radium-226 are associated with high levels of gross alpha activity. The high levels of gross alpha activity in oxidizing water are caused by dissolved uranium.

DRINKING-WATER STANDARDS AND MONITORING RQUIREMENTS

Because of the health hazards of radium-226 and -228 in drinking water the EPA has established maximum contaminant levels (MCL) to regulate total radium concentration in public water supplies. According to the EPA'S National Interim Primary Drinking-Water Regulations (U.S. Environmental Protection Agency, 1986b) the maximum contaminant levels for radionuclides are:

Radium-226 and radium-228 combined . . . . . 5 pCi/L
Gross alpha-particle activity (including radium-226 but excluding uranium and radon . . . . . 15 pCi/L
Gross beta-particle activity . . . . . 4 millirems per year

Radioactivity in ground water commonly is measured in picocuries per liter (pCi/L)—1 pCi/L is equal to 0.037 disintegrations of the radionuclide per second per liter of fluid. In the fall of 1986, the EPA announced its intention to expand its regulations to control radionuclides—radium-226 and -228, natural uranium, radon, and gross alpha, gross beta, and gamma emitters—in public water supplies (U.S. Environmental Protection Agency, 1986c.)

The EPA requires that all public-water suppliers analyze composite water samples from their distribution systems for gross alpha-particle, gross beta-particle, and radium-226 activity every 4 years. Samples also are analyzed for radium-228 when the radium-226 activity exceeds 3 pCi/L. Gross alpha- and beta-particle activity are used to determine if further radiochemical analyses are necessary. Alpha emitters, such as radium-226, radon-222, and uranium add to the total gross alpha-particle activity. Radium-228 is beta-particle emitter.

Although standards have not yet been established by the EPA for radon-222 or total uranium in drinking water, health physicists propose a 10,000 pCi/L limit for radon-222 in water (Cross and others, 1985, p. 649). On the basis of several assumptions about the volume of air in a dwelling, the amount of water consumed daily in various domestic uses, the efficiency of removal of radon from water by aeration and heating, and the proportions of water used for showering, laundering, and cleaning, Gesell and Prichard (1975) estimated that water containing 10,000 pCi/L of radon per liter would contribute about 1 pCi/L per liter of air in a dwelling. The EPA has recommended a 4 pCi/l limit for radon in air. A 10 pCi/L MCL for uranium in water has been suggested by Cothern and others (1983, p. 377).

Under present regulations, uranium and radon activity is subtracted from the total gross alpha concentration of the sample. Therefore, drinking water with high concentrations of radon or uranium can be supplied legally if the water does not exceed standards for other radionuclides.

Another deficiency in the current screening technique arises from the analysis required for monitoring radium-226, an alpha emitter, and radium-228, a beta-emitter. Historically, it has been assumed that these radionuclides were present in water in a 1:1 ratio. However, in recent years investigators have shown that very little correlation exists between levels of radium-226 and radium-228 and that separate guidelines for each isotope are needed (Michel and Moore, 1980, p. 66; King and others, 1982, P. 1173; Menetrez and Watson, 1983, p.13; Kriege and Hahne, 1982, p. 558; Cecil and others, 1987, p. 444). Hess and others (1985, p. 563) report that present screening procedures (measuring for radium-228 only if radium-226 is greater than 3 pCi/l) can miss from 10 to 50 percent of violations for total radium.

DISTRIBUTION OF RADIONUCLIDES IN GROUND WATER

Figure 19 shows generalized areas of the conterminous United States where various radionuclides exceed the following concentrations:

Radon	10,000 pCi/L
(equivalent to about 1 pCi/L in air)	
Radium	5 pCi/L
Uranium	10 pCi/L

The generalized areas shown in figure 19 reflect the dominant radionuclide detected in ground water; when other radionuclides are known to be present in the same area, they are indicated by a number. Information in figure 19 is based on a study of published reports; no field investigations were performed by the authors. Most of the data are from work by Cothern and Lappenbusch (1984) and Hess and others (1985), who examined the results of compliance data from a nationwide monitoring of more than 50,000 public water supplies for radioactivity in drinking water, and from data provided in published reports on individual States as indicated in the following discussions.

RADON

Radon in ground water is most prevalent in the Northeastern United states, especially, in the New England area. High concentrations of radon have been detected in ground water associated with granitic and metamorphic rocks of Maine and New Hampshire (Brutsaert and others, 1981; Hall and others, 1985). Radon concentrations commonly exceed 10,000 pCi/L, and a number of water samples were reported to have concentrations that ranged from 100,000 to 300,000 pCi/L. These high concentrations are attributed to uranium minerals in granites and uranium minerals in pegmatites associated with metamorphic rocks (Brutsaert and others, 1981, p. 413). Hess and others (1985, table 7) show that higher concentrations of radon occur most often in water from wells that yield small quantities of water. Many public supply systems in Maine draw their water from large-yielding less radioactive glacial sand-and-gravel aquifers; whereas most self-supply wells draw their water from small-yielding, uranium-rich granites, pegmatites, and metamorphic rocks. This difference is significant because 40 to 50 per cent of the population of Maine and New Hampshire depends on water from private self-supply wells (Hall and others, 1985). High concentrations of radon also have been detected in ground water associated with granitic and metamorphic rocks in Rhode Island (Hess and others, 1985, p. 571), Connecticut (Thomas, 1987, p. 352), Massachusetts (James H. Persky, U.S. Geological Survey, oral commun., 1987), New Jersey (Nicholls and Cahill, 1987, p. 424), Pennsylvania (Wanty and Gundersen, 1987, p. 135), North Carolina (Sasser and Watson, 1978, p. 667), South Carolina (King and others, 1982, p. 1175), and Georgia (Michel and Jordana, 1987, p. 236).

RADIUM

Radium, in concentrations greater than 5 pCi/L, is present in ground water in the Southeastern and the North-Central States. In the Southeastern States concentrations of radium that exceed EPA drinking-water standards were reported locally in public supply systems in Georgia (Cline and others, 1983), North Carolina (Menetrez and Watson, 1983), South Carolina (Michel and Moore, 1980; King and others, 1982), and Virginia (U.S. Geological Survey, 1984, p. 430).

From Georgia to Virginia, most of the higher radium concentrations in ground water straddle the Fall Line, which separates fractured rock aquifers of the Piedmont province from the unconsolidated sand aquifers of the Coastal Plain province. The source of the radium-226 and radium-228 radionuclides are uranium- and thorium-bearing minerals respectively contained in granites of the Blue Ridge and Piedmont provinces, and sands of the Coastal Plain that were derived from these granites (Michel and Moore, 1980, p. 665). Higher concentrations of radium-228 generally are found in Coastal Plain aquifers near the Fall Line. This is because the parent radionuclide thorium-232 is abundant in the sands, and

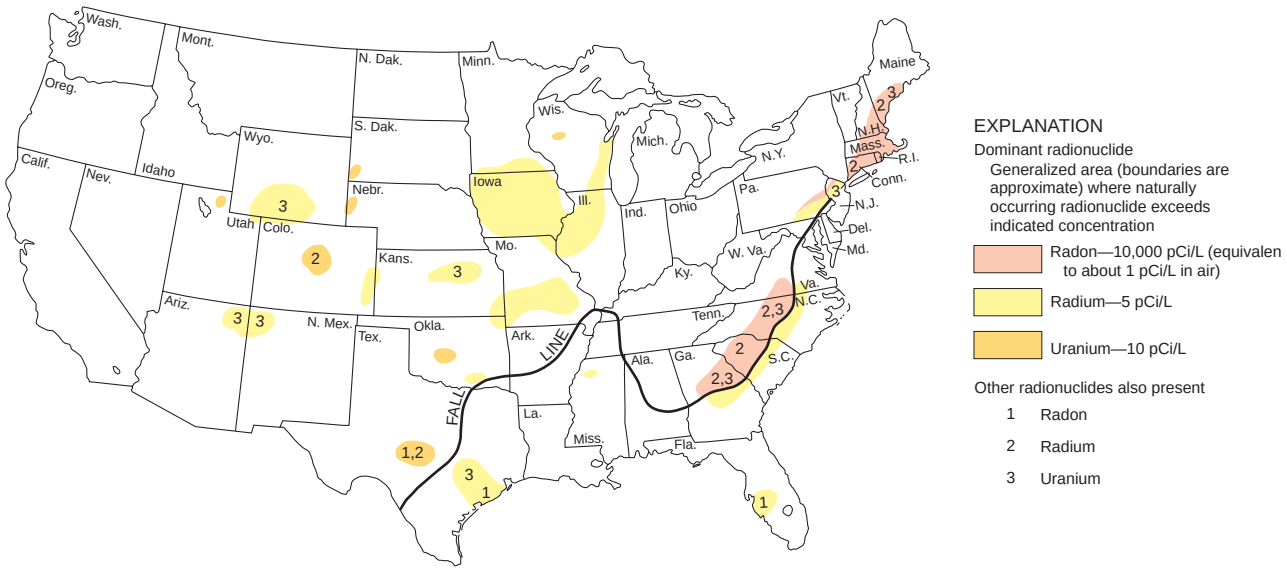

Figure 19. Generalized areas of the conterminous United States known to have high concentrations of naturally occurring radionuclides in fresh ground water. [Data are in picocuries per liter (pCi/L). (Source: 1985; Brutsaert and others; 1981; U.S. Geological Survey files, and other sources indicated in the text.)

because thorium-232 is virtually immobile in ground water and has not migrated elsewhere. Therefore, radium-228 levels in ground water in the southeastern Coastal Plain decrease with distance from the Piedmont source (Hess and others, 1985, p. 561). Radium-226 is more widespread in ground water of the southeastern Coastal Plain because of the mobility of its parent uranium in ground water and the presence of uranium-rich phosphate deposits.

Radium concentrations that exceed EPA drinking-water standards occur less frequently in the fractured rock aquifers of the Piedmont province because nonsedimentary rocks adsorb radium onto mineral-grain surfaces much more effectively than do the unconsolidated sand aquifers of the Coastal Plain. This adsorbed radium, however, is a possible source for radon levels present in ground water of the Piedmont in this area (King and others, 1982, p. 1180).

Radium-226 and radium-228 in concentrations that exceed EPA drinking-water standards also are found widely throughout the North-Central States, particularly southern Minnesota and southern and eastern Wisconsin (Hahn, 1984), northern Illinois (Gilkeson and others, 1983, 1984), Iowa (Kriege and Hahne, 1982), and Missouri (Hess and others, 1985). Much of the radioactive water comes from wells that tap deep aquifers of Cambrian and Ordovician sandstones and dolomites and Cretaceous sandstones. Significant sources of dissolved radium-226 in ground water are the parent uranium-238, uranium-234, and thorium-230 radionuclides that have been chemically precipitated or adsorbed on the surfaces of silica in sandstone aquifers. Dissolved radium-228 in ground water is due primarily to the occurrence of thorium-232 enriched accessory minerals in the sandstones (Gilkeson and Cowart, 1987).

Other areas where radium concentrations in ground water exceed EPA drinking-water standards are more widely scattered, which could in part be because of insufficient sampling. These areas are near uranium rich zones in Colorado, Wyoming, the Four Corners Region of Arizona and New Mexico, southeastern Texas, Kansas, Oklahoma, and northern Mississippi (Hess and others, 1985, p. 559; Cowart, 1981; Scott and Barker, 1961). High concentrations of radium-226 in reducing ground water have been detected in association with uranium-rich black mudstones in the Newark Basin of New Jersey (Zapecza and Szabo, 1987, p. 47; Szabo and Zapecza, 1987, p. 283) and near uranium-rich phosphate beds in central Florida (Miller and Sutcliffe, 1985, p. 1). High concentrations of radium-226 and radium-228 occur in acidic, iron-rich ground water in a quartzite aquifer in eastern Pennsylvania (Cecil and others, 1987, p. 437).

URANIUM

Uranium is widely dispersed in ground water because of the great mobility, the long half-life, and the relative abundance of this element. The highest concentrations in ground water are found in uranium-ore provinces, granites, and sediments derived from these granites in Colorado, Wyoming, New Mexico, Arizona, Utah, Oklahoma, South Dakota, Nebraska, and Kansas (Scott and Barker, 1962; Hess and others, 1985). In the Easterm United States, concentrations of uranium that exceed 10 pCi/L have been found in southeastern Maine (Wathen, 1987, p. 34), in the Piedmont of New Jersey (Szabo and Zapecza, 1987, p. 283), and in the Piedmont of Georgia and North Carolina (Cline and others, 1983; Menetrez and Watson, 1983).

HEALTH EFFECTS OF RADIONUCLIDES

In the past several years, there has been a renewed concern about the health effects of exposure to radon. Radon in water is a twofold health problem—it can enter the body by direct water consumption, or through inhalation when radon is liberated from water used for cleaning, showering, and various other purposes.

Table 2. Summary of conventional water-treatment methods that remove the most abundant naturally occurring radionuclides from ground water

[Source: Completed from information in Reid and others, 1985; Brinck and others, 1978; Hahn, 1984; Lowry and Lowry, 1987; and Menetrez and Watson, 1983.=greater than]

Water-treatment method				Effects of treatment		
	Suitable user				Potential problems	
Type	Public supply	Self supply	Efficiency in removal of radionuclide, in percent	Additional benefits	Radioactive-waste byproduct requiring proper disposal	Other
				RADON		
Granular activated carbon.	Yes	Yes	>90	No potential for radon to be released to indoor air. Little operational expertise required. Inexpensive. Long life: adsorbed radon decays away before radon adsorption capacity is exhausted.	Sludge—Can become radioactive and require licensed disposal at licensed radioactive-waste depository.	Cannister may have high gamma emissions; should be kept away from children. Radon removal capacity can be strongly diminished by organic compounds or iron in the water
Aeration	Yes	Yes	>90	No solid/liquid waste disposal problems. Little operational expertise required.	Radon in air.	Cannot be housed indoors, as can produce high levels of radon in indoor air; Moving parts need frequent repair.
				RADIUM		
Ion-exchange water softener.	Yes	Yes	81-97	Remove hardness. Little operational expertise required. Reliable. Removes radium until hardness-removal capacity is exhausted. Inexpensive and widely available.	Brine.	Adds sodium. Softened water is corrosive.
Radium selective complexor.	Yes	Yes	>90	Does not soften water. Little operational expertise required.	Sludge—Can become radioactive and require licensed disposal at licensed radioactive-waste depository.	Radium removal capacity can be diminished by high concentrations of other ions in waters, especially iron. Expensive.
Iron and manganese removal.	Yes	No	15-55	Removes iron and manganese. Does not soften water.	Liquid.	Low-removal efficiency, cannot be used on water containing more than 10 pCi/L radium. Unreliable. Operational expertise required. Suitable only for treating large volumes of water.
Barium co-precipitation.	Yes	No	High in laboratory studies.	Does not soften water.	Sludge.	Barium is a regulated pollutant and levels in water must be monitored. Operational expertise required. Suitable only for treating large volumes of water. Not tested extensively.
Manganese coated filters.	Yes	No	90	Does not soften water. Little operational expertise required.	Sludge.	May work only for a short period. Removal capacity can be affected by high amounts of iron in the water. Expensive. New technique and not readily available.
				RADIUM and URANIUM		
Reverse osmosis or electrodialysis.	Yes	Yes	>90	Decreases total dissolved solids.	Liquid.	Operational expertise required. Expensive to operate. Water can become corrosive.
Lime softening.	Yes	No	80-90	Removes hardness. Can operate continuously.	Liquid and sludge.	Adds sodium. Softened water is corrosive. Operational expertise required. Suitable only for testing large volumes of water. Unreliable in some circumstances.
				URANIUM		
Coagulation.	Yes	No	80	Removes other ions.	Liquid and sludge.	Operational expertise required. Sensitive to water pH. Suitable only for treating large volumes of water.
Anion exchange.	Yes	Yes	>90	-----	Brine.	Operational expertise required; difficult to recharge exchange resin. New, not widely available. Expensive.
Activated alumina columns.	Yes	No	>90	-----	Liquid.	Operational expertise required. New, not widely availiable.

The health risk from radon arises when it decays and its charge alpha-emitting progeny attach to dust, cigarette smoke, and other aerosol particles. These particles can be inhaled and attached to the lung interior, bringing alpha-emitting particles in constant and intimate contact with the cell lining of the respiratory system (Hess and others, 1985, p. 567). A recent estimate suggests that from 2,000 to 40,000 lung cancer fatalities per 70 years can be attributed to radon in public drinking water supplies in the United States (Cothern, 1987).

Consumption of water containing radium and, to a lesser degree, uranium can cause a significant accumulation of these radionuclides in human bone tissue. A significant dose may accumulate, producing bone and head-sinus cancers (Mays and others, 1985, p. 635; Wrenn and others, 1985, p. 601). Hess and others (1985, p. 579) estimate almost 1,000 fatal cancers may occur per lifetime (70.7 years) in the United States, on the basis of the average level of radium (1.6 pCi/L) in public ground-water supplies in the United States. Mays and others (1985, p. 635) estimate the cumulative lifetime risk to 1 million people, each consuming 5 pCi/L of radium per day, to be 9 bone and 12 head cancers fro radium-226. Radium-228, which is considered to be twice as hazardous as radium-226, was estimated to produce 22 bone cancers per million people. According to their estimates, lifetime ingestion of 5 pCi/L per day of uranium could induce 1.5 additional bone cancers per million people. Hess and others (1985, p. 580) estimate 105 fatal cancers may occur per lifetime in the United States on the basis of average concentration of uranium (0.8 pCi/L in public water supplies of the Unites States. Uranium in ground water poses additional health risks because it also is chemically toxic. Uranium has been detected in significant concentrations in soft body tissues, particularly the kidneys (Wrenn and others, 1985, p. 601).

REMOVAL OF RADIONUCLIDES FROM GROUND WATER

Conventional water-treatment methods can remove as much as 95 percent of the radionuclides present in ground water (table 2). Each radionuclide has its own specific treatment method(s) that will remove it with the greatest efficiency. If several different radionuclides are present in the water, no single treatment method will remove all of them, and multiple treatment techniques may have to be applied. Therefore, it is essential to identify the radionuclide(s) present before selecting the specific water-treatment method(s). Water purveyors will need to consider how the radionuclide removal methods can be best integrated into their existing water-treatment processes.

Each radionuclide treatment method, besides providing benefits, does pose potential problems. One of the most serious is that material removed from the water constitutes a radioactive-waste product that requires proper disposal. Disposal of the material must be carefully coordinated with appropriate environmental regulatory agencies. The self-supply well-owner is limited further by operating expenses and the lack of operational expertise. All the potential problems and benefits of a radionuclide water-treatment method must be carefully weighed before making a decision about which method to use.

Conventional water-treatment methods that remove the most abundant naturally occurring radionuclides (uranium, radium, and radon) from ground water are listed in table 2. Benefits and problems of each method are also given. Additional information including detailed methods of operation, comparison of operational expenses, and details about potential problems are reported by Brinck and others (1978), Hahn (1984), Reid and others (1985), Menetrez and Watson (1983), and Lowry and Lowry (1987), and in reports referenced therein. A summary of radioactive waste-disposal alternatives that can be managed by municipalities is given by Reid and others (1985, p. 685-686).

CONCLUSIONS

Health effects attributed to the consumption of drinking water containing radionuclides are based primarily on the analysis of water from public-supply distribution systems and not individual wells, and the risk factors generally are averaged nationwide. Water from private self-supply wells is not subject to EPA regulations and, therefore, relatively little data on the quality of the water are available. Of the more than 14 billion gallons per day of ground water withdrawn for human consumption by public-supply and self-supply wells, more than 20 percent is withdrawn by private self-supply wells (U.S. Geological Survey, 1985). In areas where radionuclides are present in ground water, self-supplied well water is suspected to contain higher levels than public supplies for the following reasons. In public-supply systems the residence time of ground water usually is relatively long because the water may be held in storage facilities and in the system itself for a period of time before being delivered to the consumer. This allows radionuclides with very short half-lives, such as radon-222 with a half life of 3.82 days, to decay to lower levels. In addition, public-water suppliers commonly mix water from several wells and/or surface-water sources. Generally, surface water has very low radionuclide activities (Hess and others, 1985, p. 563, 578). The mixing of waters with low levels of radionuclides dilutes the water and produces a finished water supply with lower levels of radionuclides when it is delivered to the consumer through the distribution system. In contrast, water supplies from domestic self-supply wells commonly are stored in small holding tanks for short periods and are not mixed with water from other sources.

Inasmuch as most of the data collected to determine radioactivity in ground water has come from the distribution lines of public water-supply systems, much additional work is needed to define levels of radioactivity in specific aquifers. Data collection and analysis are needed in rural areas where self-supply groundwater withdrawals are the primary source of water for human consumption. Areas near ore bodies, zones of uranium or thorium enrichment, and areas with high concentrations of radon in indoor air can be expected to have high concentrations of radionuclides in ground water. More detailed studies are needed to improve definition of the geochemistry of naturally occurring radionuclides, to identify constituents that might serve as indicators of their presence, to define mechanisms of their transport in ground water, and to determine their effects on human health.

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