ROOTING OUT RADIOACTIVE

The Afra springs draw from the Nubian Sandstone aquifer in southern Jordan. In similar groundwater in the Negev in Israel, researchers have found high levels of naturally occurring radiation.

All photos courtesy of Avner Vengosh

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When the Chernobyl nuclear power plant exploded in 1986, a radioactive plume spread more than 100 kilometers from the plant, affecting thousands of people. Over time, radioactive particles worked their way into the groundwater of northern Ukraine, eventually entering the region's major waterways. The accident demonstrated the fragility of any nuclear facility and raised the level of awareness over the health threats that radiation poses to people and the environment.

Although radioactive contamination still exists from weapons production and test sites in the United States, the former Soviet Union and elsewhere, the radioactivity exposure of most of the populations close to these sites is extremely low, and the spread of these contaminants in local groundwater is usually minimal. Yet the general population is still at risk from a different source: Naturally occurring radioactive particles exist in many groundwater systems worldwide.

CANCER CONNECTION

Unstable isotopes undergo radioactive decay, in which the "parent" element transforms into a "daughter" element and emits radiation in the form of alpha, beta and gamma particles. For example, the element radium disintegrates into daughter isotopes of radon and releases radiation. The collision of alpha particles with living cell tissue can damage the tissue and lead to cell mutation and cancer.

The International Commission on Radiological Protection estimates that for every 100,000 people exposed to a minimum level of radiation every year, five people will die of cancer from the exposure. Following this guideline, different countries define the "maximum contaminant level" of radioactive particles, or "radionuclides" in drinking water.

The concentration of radioactivity is the rate at which a radionuclide disintegrates. The international unit of radioactivity is the Becquerel, and in the United States, the unit is the picocurie.

In the United States, the Environmental Protection Agency (EPA) established a maximum contaminant level of 5 picocuries per liter for combined radium-226 and radon-228. In the European Union (EU), the corresponding values are 14 picocuries per liter and 5 picocuries per liter, respectively.

Epidemiological studies have found an association between bone cancer and elevated radium levels in drinking water. For example, a 2003 report released by the New Jersey Department of Health and Senior Services

'E GROUNDWATER

Scientists are discovering potentially alarming levels of naturally occurring radioactive particles in groundwater around the world.

found that males in southern New Jersey, where radium concentrations in some spots exceed the U.S. maximum contaminant level, had a threefold higher risk of developing bone cancer.

Environmental radon is considered to be the second leading cause of lung cancer in the United States, after smoking, with wide-ranging estimates of 3,300 to 38,600 deaths per year out of a total of more than 171,000 lung cancer deaths per year. Most of the exposure is from airborne radon entering homes in areas with high radon levels in soil, typically sitting atop rocks enriched in uranium (for example, granite). EPA recommends a threshold airborne radon value of 4 picocuries per liter, but new studies have argued that the risk for lung cancer is elevated even at that maximum level.

High radon concentrations in drinking water can also pose health hazards from inhalation of degassed radon in household showers. Recent experiments with shower water from high-radon well water in western North Carolina have shown extremely high levels of airborne radon (more than 50 picocuries per liter) in bathrooms during and after showers. The United States has no regulated maximum contaminant level of radon in drinking water, but EPA recommends an upper limit of 300 picocuries per liter.

GLOBAL REACH

Several surveys in the United States have attempted to evaluate the radioactivity level in drinking water across the country, with the U.S. Geological Survey (USGS) conducting the most recent study. USGS found groundwater with radium levels exceeding the maximum contaminant level in southern Minnesota, northern Illinois, Iowa, Missouri, and southern and eastern Wisconsin. In these states, the groundwater comes from wells that penetrate into the deep aquifers of Cambrian and Ordovician sandstone and dolomite, as well as into Cretaceous sandstone. In the southeastern United States, other research groups also found amounts of high radium in the Upper Coastal Plain adjacent to the contact point with the granitic rocks of the Piedmont province.

Most of these surveys found that wells serving small populations and also private wells have the highest radionuclide concentrations. Thus, it seems that although most of the U.S. population is not directly exposed to radioactive water, in some areas, local communities and private homeowners may consume groundwater with an extremely high level of radionuclides.

In the Middle East, researchers have also recently discovered groundwater with extremely high naturally occurring radioactivity. The highly radioactive water is in the deep Nubian Sandstone and carbonate aquifers in Israel's Negev desert.

In the Nubian Sandstone, the brackish (slightly salty), often low-oxygen, groundwater has high concentrations of radium-226, radium-228, and radium-224, indicating a large mobilization of radium from the aquifer rocks. The level of total radium in the Nubian Sandstone aquifer varies from 5 to 63 picocuries per liter, which means that none of the wells have concentrations below the U.S. maximum contaminant level value. In the carbonate aquifer, the radium concentration ranges from 1.7 to 93 picocuries per liter, excluding a large fraction (almost 80 percent) of the aquifer from use for drinking water.

FROM ROCKS TO WATER

Most radionuclides in groundwater result from interactions with rocks. The contents of these elements in groundwater and thus the levels of radioactivity depend on the combination of several factors, including the concentrations of these elements in the aquifer rocks, chemical reactions and the physical processes of decay along the water-rock interface.

The first factor that determines occurrence of radioactive groundwater is the geology. High abundances of parent elements of the decay chains are typically associated with high activity of one or several radionuclides in the associated groundwater. Sedimentary rocks, such as shale and phosphate rocks, are predominantly enriched in uranium and thus the decay daughter of the uranium decay chain, radium-226, characterizes the associated groundwater. Similarly, silica-rich igneous rocks, such as granite, are enriched in both uranium and thorium, and so associated groundwater contains their decay products, radium-226 and radium-228.

For example, groundwater in the sand aquifers along the upper part of the Coastal Plain in the southeastern United States has high radium levels above the maximum contaminant level of 5 picocuries per liter. This enrichment occurs from Virginia to Georgia, along the transition (known as the Fall Line) between fractured rock aquifers (granite and gneiss) in the Piedmont province to unconsolidated sand aquifers of the Coastal Plain province.

BREAKING DOWN THE WATER

The second factor that controls mobilization of radionuclides from the aquifer rocks is the chemical condition of the groundwater, which can result in significant leaching of these elements into groundwater. In particular, the levels of acidity (pH), salinity, temperature and oxidation of groundwater affect radionuclide mobilization.

Additionally, the geochemical properties of each radionuclide determine its availability in water. Radium, for example, can be kept out of groundwater by sorption onto clay minerals, precipitation with secondary minerals and radioactive decay. Many studies have shown that an exchange reaction with clay minerals is the predominant process that controls the radium activity in low-saline groundwater.

In southern New Jersey, the USGS team has shown that high radium concentrations in the Kirkwood-Cohansey aquifer system — 33 percent of 170 wells are above the maximum contaminant level — are associated with low pH and high nitrate concentrations. High radium levels in wells from agricultural areas are associated with recharge of agricultural return flows, which induces nitrification and reduces the pH. Such a combination enhances the extraction of radium from exchange sites on clay minerals, and consequently increases the radium levels in the associated groundwater.

The New Jersey case study provides an alarming message. Given that nitrate pollution is one of the most frequent phenomena of groundwater quality deterioration worldwide, additional side effects, such as the formation of acidic water and the rise of radionuclides in groundwater, may have further severe consequences.

Many other studies have shown that salinity also plays a major role in radium distribution. For example, groundwater from aquifers in parts of Missouri, Kansas and Oklahoma show almost a linear correlation between radium-226 and salinity. A strong correlation between radium activity and salinity was also evident in groundwater along the rift valley between the Sea of Galilee and the Dead Sea, as well as in groundwater from the Judea Group rocks in the Negev. The general rule is that in freshwater conditions, most of the radium remains in the aquifer rocks, while in saline conditions the radium escapes from the rocks and has high concentrations in groundwater.

Other studies have established that groundwater with no oxygen also is typically enriched in radium. For example, researchers from Columbia University in New York found that in the Delaware Basin of southeastern New Mexico, groundwater lacking oxygen contains low uranium and high radium concentrations. Likewise, an inverse correlation between dissolved oxygen and radium concentrations was identified in groundwater from the Judea Group carbonate aquifer. The high radium activity in oxygen-free water seemingly comes from the mobilization of manganese. With oxygen present, radium strongly bonds to manganese oxide and yet in oxygen-free conditions, manganese oxide is no longer stable, and both radium and manganese are released into the associated groundwater.

Finally, temperature is an important factor that also enhances radium leaching from rocks. In the southern Arava Valley in Israel, the radium activity increases with the temperature of groundwater, which also reflects the depth of wells in the Nubian Sandstone aquifer.

Many geothermal waters are enriched in radium, as documented by studies in Yellowstone in Wyoming, the Dead Sea Rift Valley, Central French Massif and thermal springs in Eastern Europe. Because radium leaching from rocks is enhanced by both temperature and salinity, mineralized thermal springs that are used as a source for bottled mineral water may have high concentrations of radium.

A survey of the radioactivity in geothermal and bottled mineral waters in Hungary, for example, shows that many of the thermal springs, including the world-famous baths of Budapest, have high radium-226 concentrations of up to 27 picocuries per liter. Radium-226 concentrations were also high in bottled mineral water commercially available in Hungary. A positive correlation between the level of mineralization (total dissolved salts) and radium-226 is evident. For example, highly mineralized bottled water with total dissolved salts of about 2 grams per liter would have a radium concentration of 78 picocuries per liter, which is approximately 15 times the U.S. maximum contaminant level.

Despite these studies, however, no systematic monitoring of the level of radionuclides takes place for imported bottled mineral water. The scientific and medical communities should thus further evaluate the radiological risks for long-term consump-

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The Yuma desalting plant in Arizona is one of the largest desalination plants in the world, although it has not operated at full capacity in recent years. As groundwater in many areas faces saltwater contamination, it becomes more at risk for high levels of radioactivity.

tion of mineralized thermal-spring and bottled water.

MOVING RADIATION

The final factor affecting movement of radionuclides into groundwater is physical. Because of the high kinetic energies that are associated with radioactive decay, daughter isotopes typically move from the parent isotopes within the rock into the water, in a process known as recoil. The relationships between the radioactive decay rates of the radionuclides and the flow rate of the groundwater will determine the amount of radioactivity that is added to the water by this mechanism.

Stagnant (and old) groundwater, for example, is generally enriched in both short- and long-lived isotopes, given the long contact time between water and the aquifer rocks. And because of increased contact between rocks and water in the cracks, extremely high levels of radon in groundwater typically occur in association with fractured granite rocks.

The Department of Environmental and Natural Resources of North Carolina recently documented high radon content in shallow groundwater in the fractured granite and gneiss aquifers of state's western mountains, with concentrations of up to 45,000 picocuries per liter. In most wells, the radon content exceeds the EPA-recommended value of 300 picocuries per liter in drinking water. Similar high-radon groundwater in silicate-rich igneous terrains has been reported from many areas in the United States, particularly in Pennsylvania, Maine and New Hampshire.

Having a very slow flow rate of groundwater in an aquifer is the ideal setting for maximizing the recoil effect, as the rate of decay of all radionuclides, including radium-226 with its half-life of 1,600 years, is slower than the groundwater flow. Thus, in very low-gradient aquifer systems, such as the Nubian Sandstone aquifer in the Middle East, the recoil process may have an important effect on groundwater radioactivity.

CHANGES AHEAD

Although the problem is persistent, natural radionuclide contamination has received little attention from the public and the scientific community. And because radium seems to thrive in oxygen-depleted, high-saline environments, the problem could become even more widespread, due to continued degradation of groundwater quality from agricultural activity or salinization of water resources (for example, deicing activities in the northeastern United States), which may mobilize more radium from the aquifer rocks for release into groundwater.

Additionally, with a scarcity of water in many areas, alternative water resources, such as tapping groundwater from deep aquifers and using marginal water for agriculture, could become more popular. Groundwater from deep aquifers is typically oxygen-depleted and has a very slow flow rate, and marginal water typically has high salinity. These alternative water resources may therefore also have high radium concentrations.

Given the continuous degradation of the quality of groundwater in many aquifers worldwide, and the increasing demand for using alternative water resources, the radioactivity factor may be more important than we realize. The bioaccumulation of radium in plants and fish may pose health risks, as people use slightly salty groundwater for agriculture or for fish farming.

In the Middle East, the lack of adequate water and the contamination of many groundwater basins have resulted in exploitation of the fossil groundwater from the Nubian Sandstone aquifer (see *Geotimes*, May 2004), which is considered to be the last available water resource for Saudi Arabia and other Persian Gulf states, as well as Jordan, Israel, Egypt and, on a massive and increasing scale, Libya. High radium concentrations found in groundwater from the Nubian Sandstone aquifer in the Negev may imply that similar groundwater from other Nubian Sandstone basins also may be radioactive. Consequently, utilization of these water resources would require significant remediation, to avoid health implications associated with long-term consumption of high-radium groundwater.

The challenges with respect to radioactive groundwater are threefold. First, high radium levels may exist in well waters in rural and suburban areas in several "hot spots" in the United States. Thus, long-term monitoring for radon and radium-226 and radium-228 is essential for protecting the health of residents in these areas. This problem is further intensified by the dearth of federal and state regulations of private homeowners' wells, and because radionuclide measurements are not part of routine inorganic measurements. This situation may exist in many other groundwater basins worldwide.

The second challenge is related to water quality studies that rarely include radium isotopes in their toolbox. Such monitoring, again, is key, as communities continue to change water quality through acidification or salinization.

The final challenge is remediation. Although some chemical filtration processes, such as ion exchange or reverse-osmosis desalination, remove significant amounts of radium from the water, the residual water can still be radioactive, due to the selective accumulation of radium. Thus, large-scale desalination of radium-rich slightly salty groundwater is not a simple task. The procedure for radium removal from radioactive groundwater must take into consideration the disposal of radioactive waste material.

The global community must aggressively address these challenges, to ensure a safe water supply. The first step in any problem such as this is recognition and awareness.

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