Environmental Radon — Controls on the distribution and possible health effects

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Introduction

Radon 222 (Rn222) has been known as a respiratory carcinogen for many years as a result of epidemiological studies of uranium miners (Archer, 1975). High concentrations of Rn222 in air in homes in the United States have resulted in the establishment of limits for Rn and daugthers in air in certain high risk situations, such as in the vicinity of and on mine tailings. However, no general guidelines have been developed for acceptable levels of Rn and daughters.

In 1976 we initiated studies in Maine, USA to determine the distribution of naturally occurring Rn222 in groundwater, to explore processes wich controlled Rn in home air, and to preliminarily explore possible relationships between Rn222 in water, air, and health statistics. These investigations were initiated when it was discovered in routine studies of natural background radiation near an atomic power plant that high levels of Rn222 existed in groundwater. We focussed on domestic water supplies, including wells drilled into bedrock or glacial material as well as surface supplies (dug wells, springs, streams, and lakes). Public water supplies were also evaluated. The methodologies used and results of our studies are presented in Hess et al. (1978, 1983) and Brutsaert et al. (1981).

Rn222 is a naturally occurring radionuclide which is formed during the decay of uranium (U238) to lead (Pb206) (Figure 1). Because of the widespread distribution of U in rocks, soil, and sediment, the daughter products are also widely distributed. Although chemical weathering of rocks and soils mobilizes nuclides in Figure 1 for movement through the ecosystem, little of the radionuclides is removed from bedrock. The daughters U234, thorium (Th230), and radium (Ra226) may be mobilized by chemical weathering but are later sequestered by geochemical processes into sediments. Because of the long half life of U238 (4.5 billion years) virtually all the U238 initially in a rock at the time of formation is still present; the daughter products have grown into secular equilibrium in a relatively short time (10^6y).

Radon, being an inert gas, has special capabilities for migration in our environment. When a Ra226 atom decays close to a rock/water interface (Figure 1) the Rn222 atom which is produced may be displaced by the recoil into the water. If the water is moving, Rn222 can be transported. Because of the short half life

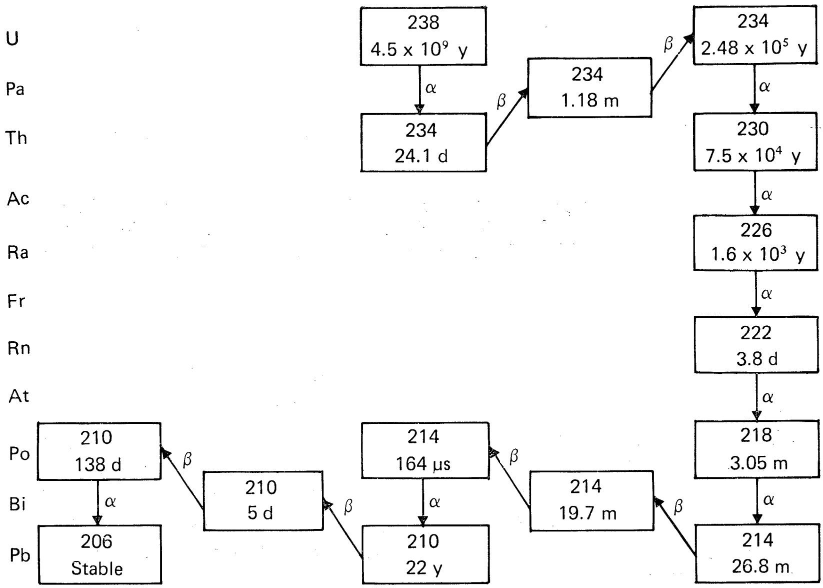

Figure 1: *Decay scheme for U238 to Pb206. Within each block are the atomic weight and the half life.*

(3.8d), Rn222 will decay nearly completely in a few weeks to produce polonium (Po218). Thus, unless water receiving Rn222 from the rocks is rapidely extracted from the ground, the Rn222 is lost by decay, providing that it is not replaced. If groundwater moves from a rock type with little Rn222 generation into one with elevated production, Rn222 wil grow into a state of secular equilibrium within several weeks. Furthermore, because of the slow rate of movement of groundwater, if Rn222 is present in a water sample, it is clear that the immediate parent, Ra226, was close by. If the decay of Ra226 occurs at a mineral surface in contact with soil air, the Rn222 may be liberated directly or indirectly into the atmosphere. Rn222 may also be added to the atmosphere by the degassing of groundwater to the atmosphere as it emerges into streams and lakes. As a consequence of this, public water supplies, which are commonly from surface waters and treated by aeration, have low Rn222 activity.

Certain rock types typically have higher concentrations of U, either because of magmatic processes (as is the case for granites and related rocks) or sedimentary processes (e.g. the alunskifer in the Oslo graben). Carbonate rich (CaCo3) sediments (and their metamorphic equivalents) typically have higher than normal U because of coprecipitation of U in the CaCO3. Some marine sediments may also be enriched in Th as a result of interaction of clays with sea water. Carbonaceous terrestrial sedimentary rocks commonly

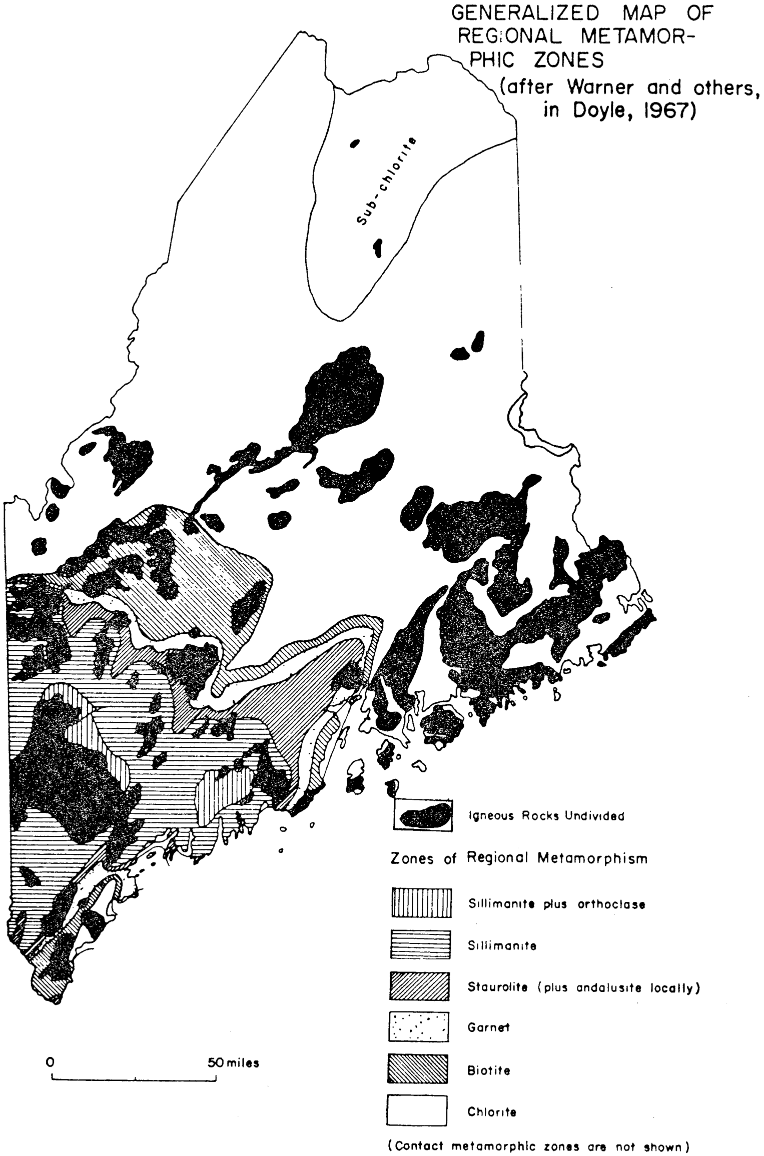

Figure 2: *Geologic map of the state of Maine, USA showing the distribution of igneous rocks (primarily granite) and metasedimentary rocks of various grade.*

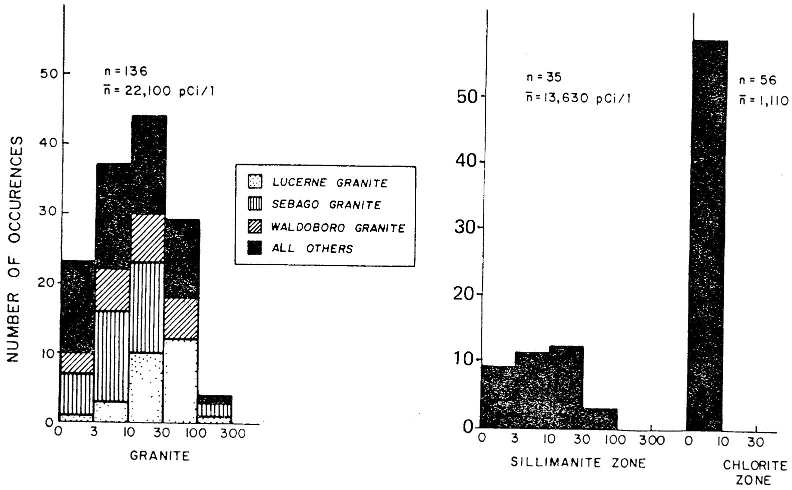

Figure 3: *Number of occurrences of radon concentrations in water for three types of rocks. Based on 1977—1978 data. Values are 10^3 pCi/l.*

have enriched zones of high U. Because of these natural variations in the abundance of U and daughters, we attempted to characterize the various lithologic units in terms of the activity of Rn222 that one might expect to find in groundwater pumped from them.

Results

Figure 2 is a generalized geologic map of the state of Maine, USA showing the distribution of igenous rock bodies (primarily granite). Sedimentary rocks and their metamorphic equivalents have been deformed into a series of northeast trending folds. These rocks have in turn been metamorphosed with the intensity increasing to the southwest (Figure 2). We investigated the influence of major geologic variables believed to control the distribution of Rn:

1. Major rock type — igneous versus all others
2. Sedimentary rock types — shales, limestones, sandstones, etc. and
3. Various metamorphic equivalents of the sediments.

Based on approximately 2000 water samples from wells, we concluded that:

1. Sub-sillimanite zone metasedimentary lithic units typically have characteristic ranges of Rn222 values which are partly overlapping and typically have means between 1000 and 5000 pCi/l (NOTE: 1Bq/l = 27pCi/l = 1 dps)). Chlorite zone rocks and unmetamorphosed rocks had a mean value of about 1100 pCi/l (Figure 3).

2. Radon in groundwater from the same stratigraphic unit was not a function of metamorphic grade up to sillimanite grade.

3. At sillimanite grade and higher (Figure 3), stratigraphic units yielded water with higher Rn222 activity than their lower grade equivalents. Intrusive pegmatitites

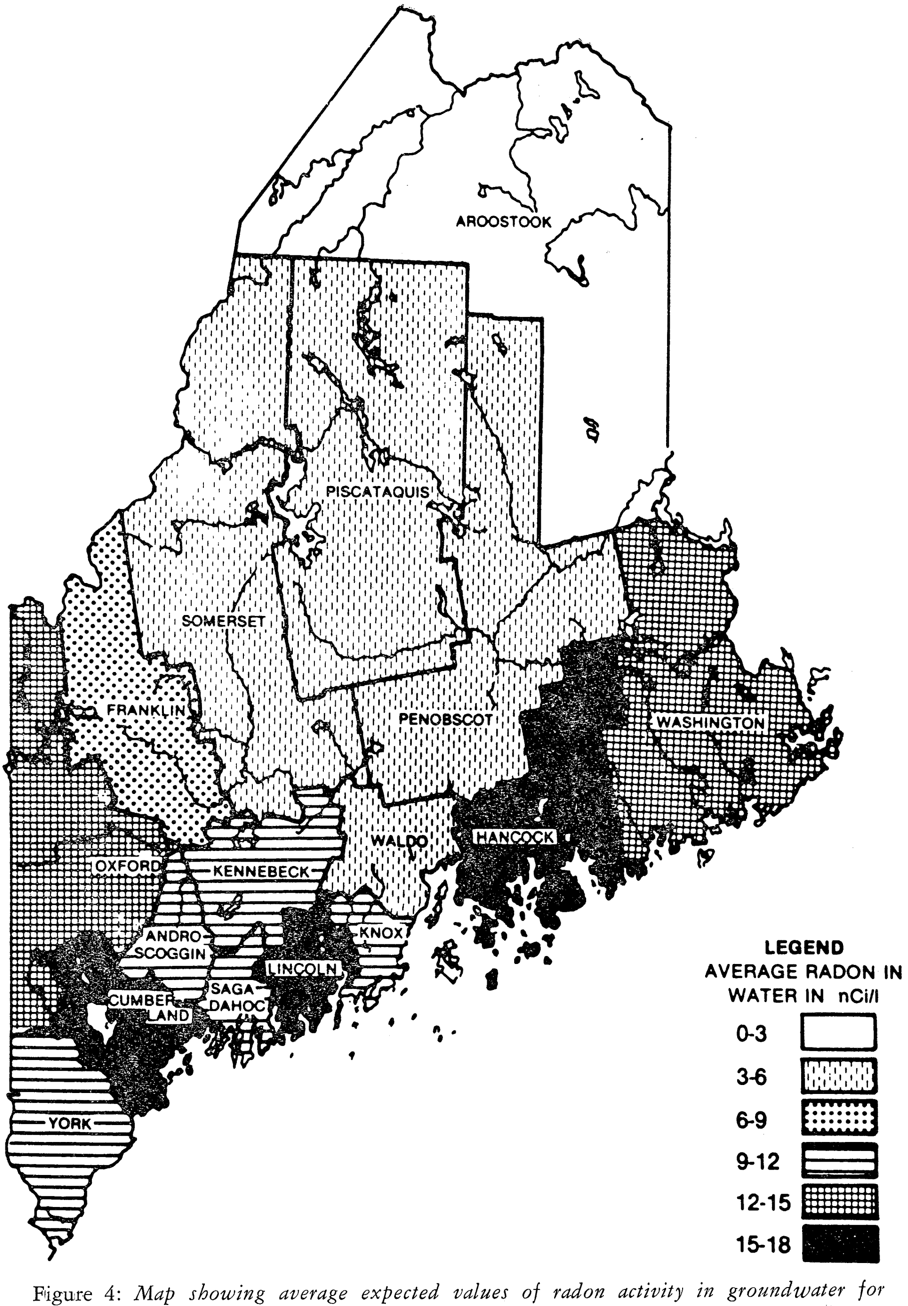

Figure 4: *Map showing average expected values of radon activity in groundwater for for Maine, USA. counties.*

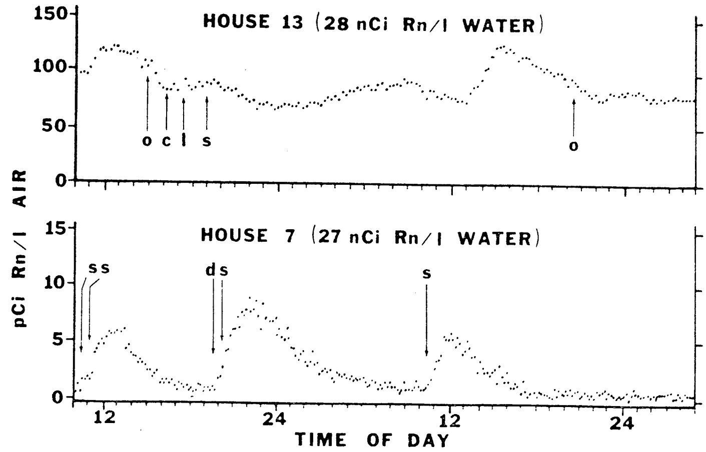

Figure 5: *Time variations of radon in two houses in Maine, measured with an electrostatic diffusion alpha detector. Events of interest are labeled: (o) opened window to outside; (c) closed window; (l) laundry machine used; (s) shower; (d) dishwater used.*

and anatectic lenses are common within sillimanite and higher grade rocks. This suggests the precence of aqueous fluids which may have been responsible for the emplacement of U in these rocks during metamorphism.

4. Waters from granites had the highest Rn222 values. They commonly exceed 50.000 pCi/l, or 100 times the recommended value for potable water (USEPA, 1976). Values were highly variable within a single granite body. The source of the variation has not been determined but may be local variation in Ra226 content of the host rock, variable surface area of rock which is exposed to groundwater, and variable amount of short term recharge from surface waters.

Utilizing the characteristic Rn222 activities in groundwater for each mapped rock type we determined, on an areal basis, the expected average Rn222 value for groundwater for each county in Maine (Figure 4). This approach was utilized because we wished to compare Rn222 values with the health statistics, gathered on a county basis. This was the smallest political subdivision with a sufficiently large population so that we could compare health and radon data in a statistically significant manner.

Pathways to Man

Rn222 may be ingested directly by drinking water but radiation dosage from this pathway is believed to be inconsequential because of self absorption by the fluid and food in the stomach, etc. Alternatively and more importantly, Rn222 may enter the body through the lungs, via air. Rn222 is introduced into the air in the home in two important ways from natural sources:

1. Degassing from water used in the home, particularly bathing. The relative importance of this process varies according

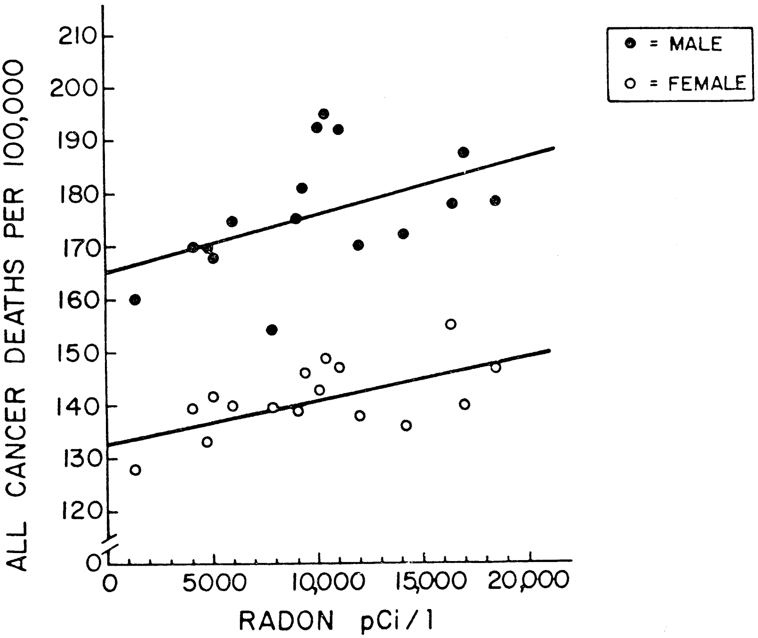

Figure 6: *Age-adjusted mortality rates for all types of cancer together versus average radon activity in groundwater, by couty. Lines represent the regressions:*
$y = 0.00109x + 165.24$ $(r = 0.47; p \leq .10)$
$y = 0.00081x + 133.20$ $(r = 0.61; p \leq .05)$

to the strength of the other sources. In a series of water degassing experiments in houses with variable Rn222 in water and variable air changes per hour, we determined that air-borne Rn222 was 0.8pCi/l per 10,000 pCi/l in water. Figure 5, lower panel, shows the airborne Rn222 as a function of time for House 7 where the groundwater had 27,000pCi/l. A clear and strong relationship between water usage and airborne Rn222 existed, with background being less than 1 pCi/l and short term values approaching 10, coincident with high use of water.

2. Degassing of Rn222 from soil beneath houses into the home environment is a very important process. In sharp contrast to House 7 is House 13, (Figure 5, upper panel) which had a water supply with 28,000pCi/l, virtually the same as House 7, but with background airborne Rn222 of 75pCi/l. In this house, water use has nearly no influence on airborne Rn222. Rather, background values and frequency of air changes are dominant determining factors. House 13 had a cellar with a dirt floor and there was a greenhouse attached to the house; the floor of the greenhouse was fine grained crushed granite! In this situation, water use is irrelevant to the activity of Rn222 in air. However, soil chemistry in much of Maine is related to the underlying bedrock. Consequently, where we have found high

Figure 7: *Areas in Norway underlain by granite and granitic gneisses (courtesy of L. Lien, NIVA).*

Rn222 in groundwater, soils are high emanator of Rn222 also. Therefore, high airborne Rn222 occurs where the bedrock yields water with high Rn222 *and* where degassing of soil into the home space is possible.

Radon and Health

Figure 6 shows on a county basis the relationship between cancer deaths and the average expected concentration of Rn222 in groundwater. The relationship is significant for «all» cancers, and for several types of cancers, including lung cancer.

A causal relationship between the two variables is difficult to establish. Many other factors have been linked with cancer, some of which may covary with Rn222. Personal habits such as smoking, time-in-residence, occupation variation in different parts of the state, the number of people on public versus private water supplies, and more precise determination of where people actually live in geologically heterogeneous counties must be investigated. The very strong variation of airborne Rn222 induced by the presence or absence of soil Rn in homes also must be controlled for in any epidemiological investigation. Elevated cancer rates in some areas, if causally linked to Rn222, may be caused principally from soil-derived Rn, and high concentration in groundwater in an area are just indicative of the potential problem.

Application to Norway

The geology of Norway is exceedingly complex but for the purposes of establishing the general location of the potential Rn222 problem, one needs onlys to locate those areas underlain by granite and granitic gneisses. Most of these areas are shown in black on Figure 7; they underlie many of the highly populated parts of Norway. Additionally, there are selected startigraphic units, such as the alunskifer, or zones where there has been high grade metamorphism of pelitic rocks, which may be of concern. The proportion of people in Norway who derive their drinking water from bored wells is probably less than a tenth of the population. However, the number of people living on areas with potentially high soil Rn is much greater. It would seem prudent to initiate a program of both water and air sampling, stratified by geology and soils.

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