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Radon, Geology, and Human Health in Maine

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ABSTRACT

The distribution of radon (Rn-222) in ground water in Maine is controlled primarily by bedrock geology. Stratigraphic units at various metamorphic grades below sillimanite grade yield ground water with overlapping ranges of values that do not vary systematically with grade. In these rocks, means fall between 1,000 and 5,000 pCi/l (37,000 and 185,000 Bq/m^3). Waters from sillimanite and higher metamorphic grade rocks have mean activities consistently in the 10,000 to 15,000 pCi/l (370,000 to 555,000 Bq/m^3) range. Granites yield water with the highest mean activity of 22,000 pCi/l (814,000 Bq/m^3).

The major pathway for Rn and its daughters to man is via air to the lungs. Major sources of airborne Rn include degassing of well water indoors and entry of soil gas into dwellings. The strength of these sources is closely linked to bedrock geology. Calculated average Rn activities for ground waters, by county in Maine, correlate significantly with the incidence of all cancers and cancers of the lung.

INTRODUCTION

In the late 1950's, a young lad received a rather primitive (by modern standards) Geiger counter for Christmas. Because of his curiosity, the water supply for the Dielectric Products Company in Raymond, Maine was found to have had an exceptionally high level of natural radioactivity. This observation resulted in a study of 500 water supplies in that region (Grune et al., 1960). Later studies by Smith et al. (1961), Lowder et al. (1964), Richardson (1964), and Hoxie (1966) found some of the highest Rn-222 values in the United States for water supplies. However, interest in the issue diminished.

In the late 1960's and early 1970's, Hess et al. (1973) investigated the levels of natural radioactivity in and around Wiscasset, Maine to characterize the radiation environment prior to the start-up of Maine's only nuclear power station. They rediscovered the high Rn values in Maine ground water, but in a different area. Realizing that high concentrations of Rn in ground water were both natural and widespread, we initiated a series of investigations to establish the extent of the problem, the controls on Rn distribution, pathways to man, and related health effects (Hess et al., 1978, 1979, 1980, 1981a,b, 1983; Brutsaert et al.,

1981). We focused on domestic water supplies, including wells drilled into bedrock or glacial material and surface supplies (dug wells, seeps, streams, and lakes). Public and water company sources, and schools were also evaluated. This paper summarizes many of the geologic, hydrologic, and health aspects of our research.

Methods of collection and analysis of water samples for Rn and other constituents are detailed in Brutsaert et al. (1981). Air samples for determination of daughters of Rn were collected and analyzed by methods detailed in Hess et al. (1981a). Water samples from 350 wells were chemically analyzed for content of major cations to assess relationships between chemical weathering and Rn activity. Methods are detailed in Hess et al. (1979). Preliminary epidemiological studies used methods which were outlined in Hess et al. (1983).

In 1979 we initiated research on the removal of Rn from water. This research is still ongoing (Hess et al., 1979; Lowry et al., 1987). Since about 1983, our research has focused on Rn in Maine homes, sources of Rn other than from ground water, and the epidemiology of Rn. The results of these efforts may be found in Hess et al. (1985), Wadach and Hess (1985), and Lanctot (1985).

RESULTS AND DISCUSSION

Hydrologic Controls

Rn-222 is one of the daughter products of the decay of U-238 (Fig. 1). The immediate parent of Rn is Ra-226 which has a half-life of 1600 years. Repeated measurement of the concentration of Rn in the same ground water samples, to evaluate the amount of Ra support for Rn, has shown that there is virtually no Ra in solution in Maine ground water that we have examined, i.e. the Rn is unsupported. Consequently, Rn must be derived from the decay of Ra which is either adsorbed or co-precipitated with solid constituents of the bedrock/soil complex. Considering the rate at which ground water moves through bedrock (typically less than 1 m/day with no pumping stress) and the short half-life of Rn-222 (3.8 days), the Ra responsible for the production of the Rn must be in close proximity to the well producing the water (Tanner, 1964). The radius for the source area of the Rn is almost certainly less than 100 m from the drill hole for most domestic wells.

Brutsaert et al. (1981) found a slight tendency for decreasing Rn activity with increasing well depth, especially for wells deeper than about 50 m. This trend is probably related to a longer residence time for the water in the borehole due to the larger volume in deeper boreholes. They also demonstrated that Rn activity decreased as a function of increase in well yield, all other variables being controlled. They interpreted this to indicate that low yield wells had a high ratio of fracture surface area to stored water. With increased transmissivity and water yield due to more open fractures, the ratio decreased so that less Rn was available per unit volume of water. The combined magnitude of these two effects is less than a factor of two. Consequently, the Rn activity in a ground water sample is characteristic of the strength of the proximal Ra source, somewhat modified by local hydrology and well depth. The determination of the distribution of Rn thus produces a map of the distribution of Ra.

Bedrock Geologic Controls

If U-238 and the daughter Ra-226 have not been widely separated by geochemical processes, the U content of rocks ultimately controls Rn activity. Certain rock types tend to have high U content, either by virtue of magmatic processes (granites, pegmatites, and migmatites) or sedimentary processes. For example, sulfidic and/or carbonaceous sedimentary rocks and their metamorphic equivalents are typically high in U. With this background, we attempted to characterize the various stratigraphic units and igneous rocks in Maine in terms of their ability to supply Rn to ground water.

Figure 2 depicts the distribution of the larger granite bodies within the state and the distribution of metamorphic isograds. Water samples collected across the state enabled us to characterize many granites, stratigraphic units, and metamorphic zones. The following conclusions can be drawn:

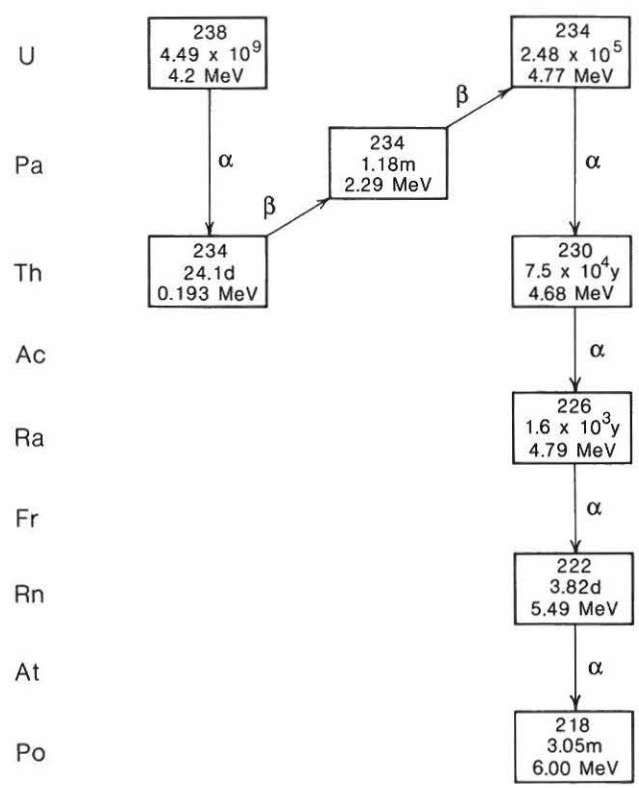

Figure 1. Decay scheme for U-238 to Po-218. Within each block are atomic weight, half life, and energy of the decay.

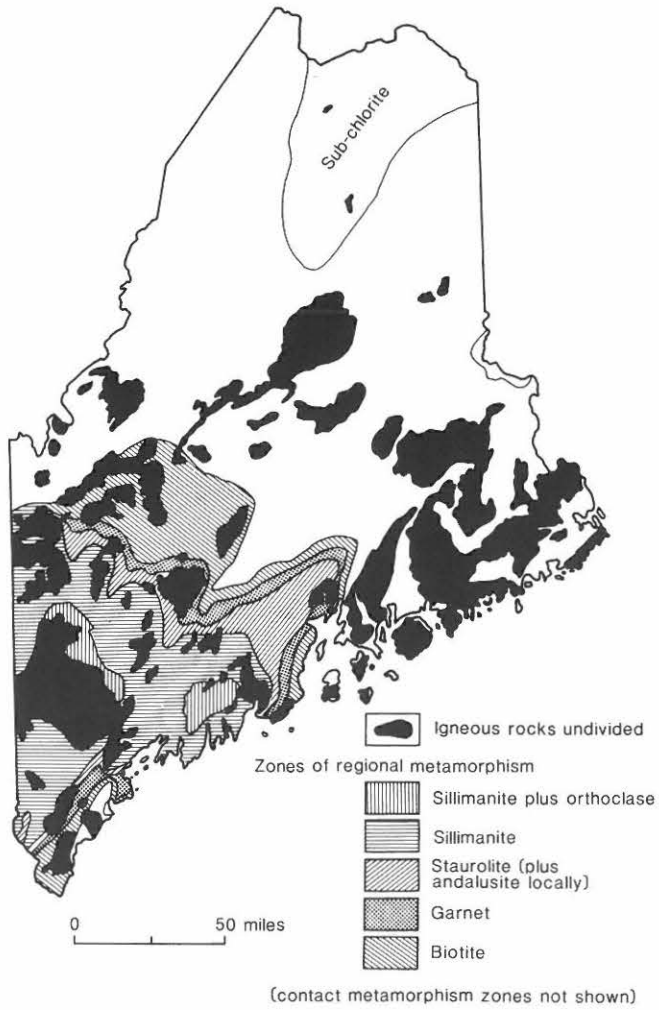

Figure 2. Geologic map of Maine showing the distribution of igneous rocks and metasedimentary rocks of various metamorphic grades (modified from Warner et al., 1967).

(1) Metasedimentary lithic types in the sub-sillimanite zones typically have partially overlapping ranges of Rn values and means between 1,000 and 5,000 pCi/l (37,000 and 185,000 Bq/m^3). Chlorite zone rocks (and lower?) have a mean value of about 1,100 pCi/l (40,700 Bq/m^3) (see Fig. 3).

(2) Rn activity was evaluated from the biotite isograd to the sillimanite isograd for the Vassalboro, Waterville, and Appleton Formations with 10 samples in each unit (Brutsaert et al., 1981). No relationship was present between activity and metamorphic grade over the range investigated. Variation between lithologies was far greater than within lithologies across metamorphic grades. However, rocks of chlorite grade yield distinctly lower values for Rn. Lanctot et al. (1985), in a more extensive study, found an increasing mean value of Rn in ground water with increasing metamorphic grade, especially between low-grade and medium-grade rocks. They divided samples into those originating from low-, medium-, and high-grade metamorphic zones and lumped data by zone, not by stratigraphic unit. The difference in trend reported by Lanctot et al. (1985) may be caused by non-equal areal distribution of various stratigraphic rock types with their characteristic U concentrations. Alternatively, our lack of trend could be caused by a small sample.

(3) Stratigraphic units at sillimanite grade and higher (Fig. 2) yield higher Rn-222 activity than their lower grade equivalents (Fig. 3). Pegmatites are common in sillimanite or higher grade rocks. Their presence implies that fluids have moved about, mobilizing U from preexisting sources and redistributing it in sillimanite and higher grade rocks. Additional U may also have been introduced from fluids associated with the cooling and crystallization of the various phases of granitic rocks associated with the Paleozoic and Mesozoic plutonism of Maine.

(4) Granites yield the highest Rn activity with values commonly exceeding 50,000 pCi/l (1,850,000 Bq/m^3) or 100X the suggested value for potable water (USEPA, 1976). Values are highly variable within a single granite body (Fig. 3; Table 1).

(5) The activity of Rn is not related to the major cation content (Ca, Mg, Na, or K) of solutions. Many wells showed evidence of road salt pollution (excess NaCl) or elevated Ca and/or Mg in excess of that expected from the local bedrock, presumably due to ground water pollution from soil amendments (limestone).

Surficial Material Controls

Soil thickness and soil type overlying the bedrock have little effect on the variation of Rn in ground water (Brutsaert et al., 1981) if wells penetrate into the bedrock. However, wells developed in surficial material overlying granites have relatively low Rn concentrations in the water. Large variations in Rn activity in wells within the same bedrock body may be attributed to heterogeneous distribution of Ra in the bedrock aquifer or to

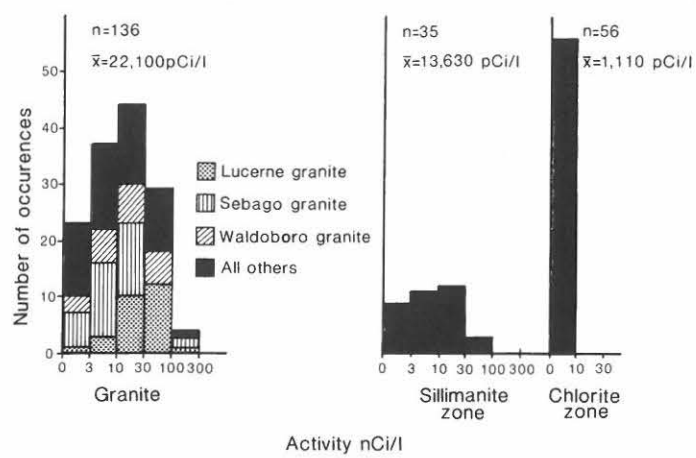

Figure 3. Numbers of occurrences of radon concentrations in water for three types of rock. Based on 1977-1978 data.

TABLE 1. SUMMARY OF RADON VALUES FOR GROUND WATER FROM VARIOUS GRANITES IN MAINE

	# Wells	$\overline{\text{Radon}}$ (pCi/l x 10^3)	S.D.	# Wells of Known Depth	$\overline{\text{Depth}}$ (ft)	S.D.	# Wells of Known Yield	$\overline{\text{Yield}}$ (gpm)	S.D.
Sebago granite	34	17.7	31.5	23	205	152	13	12.0	9.9
Lucerne granite	27	35.8	27.4	24	199	134	9	15.5	18.6
Waldoboro granite	22	19.6	18.3	18	146	96	6	13.7	10.8
Aurora granite	12	13.8	10.4	10	141	80	5	24.3	33.2
Mt. Waldo granite	10	31.0	25.0	8	152	78	2	7.5	3.5
Mt. Desert (W) granite	9	15.5	8.8	6	146	78	2	53.5	65.8
Mt. Desert (E) granite	6	3.0	2.2	9	98	45	3	6.2	3.4
Red Beach granite	6	21.8	16.7	4	95	35	3	14.3	15.5
Meddybemps granite	6	39.7	58.7	6	150	72	5	14.0	11.1
Biddeford granite	4	1.5	0.7	3	92	52	2	32.5	38.9
	$\Sigma = 136$								

	N	$\overline{\text{Radon}}$ (pCi/l x 10^3)		N	$\overline{\text{Radon}}$ (pCi/l x 10^3)
Well Depth:			Overburden:		
0-100 ft	41	14.9	0-10 ft	25	27.3
101-200	44	24.3	11-50	25	19.6
201-400	20	23.8	51-	19	13.6
401-	6	36.3	Unknown	67	23.0
Unknown	25	23.8			
	$\Sigma = 136$			$\Sigma = 136$	
Overburden:			Yield:		
Till	73	24.5	0-4 gpm	12	38.3
Sand/Gravel	36	9.8	5-10	18	21.7
Clay	7	17.6	11-25	12	15.0
Unknown	20	35.4	26-50	4	8.7
			51+	4	2.0
	$\Sigma = 136$		Unknown	86	22.1
				$\Sigma = 136$	

variable recharge rates which affect the residence time of water in the aquifer. Brutsaert et al. (1981) did find a weak inverse relationship between Rn activity and Na concentration; the relationship was significant only in the Appleton Formation and suggested some effect of rate of recharge on Rn activity.

On the other hand, soil thickness and type must play an important role in the control of the distribution of Rn gas in the soil atmosphere. The latter has been demonstrated to generally be more important in determining the total Rn activity in home atmospheres (Wadach and Hess, 1985; Lanctot et al., 1985).

The degree to which soils reflect the composition of the bedrock varies widely. Along the coastal zone, below the Holocene marine limit (approximately 80 meters above sea level near the coast), marine silts and clays overlie bedrock or other glacial deposits. The marine limit rises to about 130 m inland (Thompson and Borns, 1985). These marine deposits are largely rock flour and are not chemically the same as the bedrock, except locally and fortuitously. Inland, tills more commonly mimic the bedrock chemically. Ice contact deposits including eskers, deltas, kames, and outwash sand and gravel deposits typically reflect bedrock types within a few tens of kilometers of a particular location, but may not accurately reflect the underlying bedrock.

Mobility of Rn derived from the soil is largely determined by the diffusion coefficient for Rn and the permeability of soil. Permeability is a function of soil texture and structure. Both the diffusion coefficient and permeability are strongly controlled by soil moisture content. For example, silty tills, usually wet because of low hydraulic conductivities and perched water table conditions, would normally be quite impervious to soil gas and allow little movement of Rn except with mass movement of the water. Eskers, usually dry with low water tables, are quite pervious to air and may exhibit substantial movement of soil gas related to the displacement associated with the rapid passage of infiltration fronts or fluctuating water tables.

Rn in Homes

Rn can be introduced into homes by three processes:

(1) Degassing of Rn-rich domestic well water. When Rn-rich water is agitated with air as in a shower, Rn degasses into

the air. Typical partitioning of Rn from water to air is about 4.5×10^{-5} to 13×10^{-5} (Hess et al., 1987).

(2) Soil gas entry through basement foundations or soil basement floors. The Rn enters by direct transfer of soil air into the house through pores and cracks in the basement wall, sumps, and water drains, and to a much lesser extent, through diffusion. The latter method is relatively ineffective because of the short half-life of the Rn. In turn, the mass movement of soil gas depends on the permeability of the soil and pressure difference between the soil gas reservoir and the house, which is typically at reduced atmospheric pressure due to stack effect, wind effects, or air pumping due to furnaces and wood stoves.

(3) Production and release of Rn from Ra-rich building materials. Radium-rich commercially available building materials, as have been documented in, e.g., Polish homes (Pensko et al., 1980), are seldom a major source of Rn in the United States (Ingersoll, 1983). However, high Rn concentrations have been observed in cellar atmospheres where granitic material has been used as fill under and around foundations (Wadach and Hess, 1985). This is apparently related to permeability of the wall to soil gas.

The relative effects of soil gas and domestic water supplies in determining the Rn release into houses can be seen by examination of data from two houses in Maine (Fig. 4). House A, with airborne Rn activities close to 100 pCi/l (3,700 Bq/m^3) has an attached greenhouse with a porous sand floor. Rn-bearing soil gas entered House A largely through the greenhouse floor as well as emanating directly from the floor material itself. The water supply for the house had about 28,000 pCi/l (1,036,000 Bq/m^3). Water-use, marked by letters l and d for laundry and dishwashing, shows only small influence on the Rn activity curve. House B shows little evidence of Rn entry through the soil gas and a much larger proportion of Rn is attributed to the degassing of Rn from the water supply. The maximum Rn activity in air is 10 pCi/l (370 Bq/m^3), whereas the minimum is at 0.5 pCi/l (close to the value for outdoor air). Virtually all the Rn in the house atmosphere can be attributed to water usage and degassing.

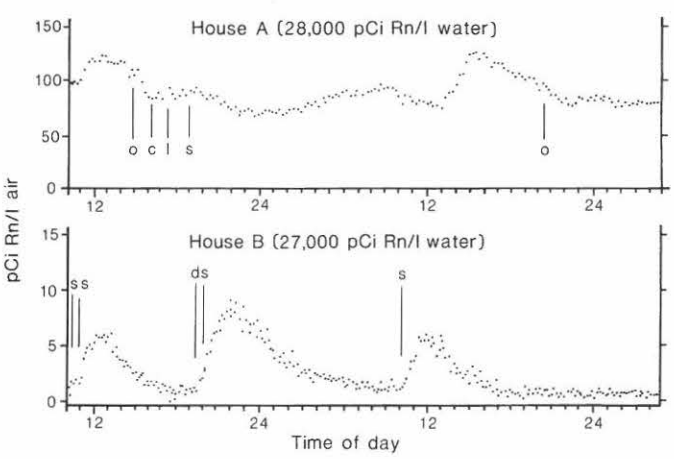

Figure 4. Time variations of radon in two houses in Maine, measured with an electrostatic diffusion alpha detector. Events of interest are labeled: (o) opened window to outside; (c) closed window; (l) laundry machine used; (s) shower; (d) dishwasher used.

Current measurements of airborne Rn in Maine homes (Hess et al., 1985) suggest that elevated indoor Rn concentrations in many homes may be related to high permeability in soils that are developed on coarse-grained glacial deposits, such as eskers, outwash, and deltas. Many of these deposits coincide with rural roads and hence a large portion of the rural population. These coarse-grained materials are typically in thick deposits with an unsaturated zone that may commonly exceed 5 m, providing for very mobile soil air if the water table should rise due to infiltration. Soil air will then migrate to points of lower pressure, e.g. basements of houses sited in these deposits. Impermeable surficial material such as clay-rich till or marine silt deposits may effectively cap more permeable strata, enhancing the migration of soil gas to low pressure regions when the water table rises. Even a snow pack may operate in such a fashion.

Rn and Human Health

We calculated the effects of human exposure to Rn from water in the following manner. We derived average Rn activity in water for counties by making a number of assumptions. The county division was taken as the regional geographic unit because it is the basis for health statistics (Mason et al., 1975).

Average expected Rn activity for a county was assessed by weighting the area of a county underlain by each rock type yielding a certain activity. These county average values are shown on Figure 5. There are several difficulties in applying these data to the population at large and assessing related health effects. In doing so, one must assume that (1) the population is evenly distributed throughout a county, (2) the population which is served by public water supplies and/or removed from the influence of Rn degassed from water and soils is evenly distributed by county, (3) the residence time for individuals is sufficient for effects to be caused by exposure to Rn and daughters, and (4) Rn concentrations in homes are related to water quality or soil characteristics, both of which may be strongly linked to bedrock geology.

The sixteen county averages were then compared in a group with age-adjusted cancer rates (Mason et al., 1975). The rates were derived from cancer mortality from 1950-1969. This group of people was mainly 50-70 years old and represented a population born near 1900. This approach did not correct for those individuals whose water source is a public water supply, the concentration of the population in towns or cities, or population mobility. Whereas much of the Rn dose comes from soil gas, the effect of public versus private water supply is reduced in significance. Similarly, Maine's population is known for its comparative immobility, reducing possible effects of in-migration and out-migration of individuals. All of these corrections would act so as to increase the relationship between Rn dose and cancer,

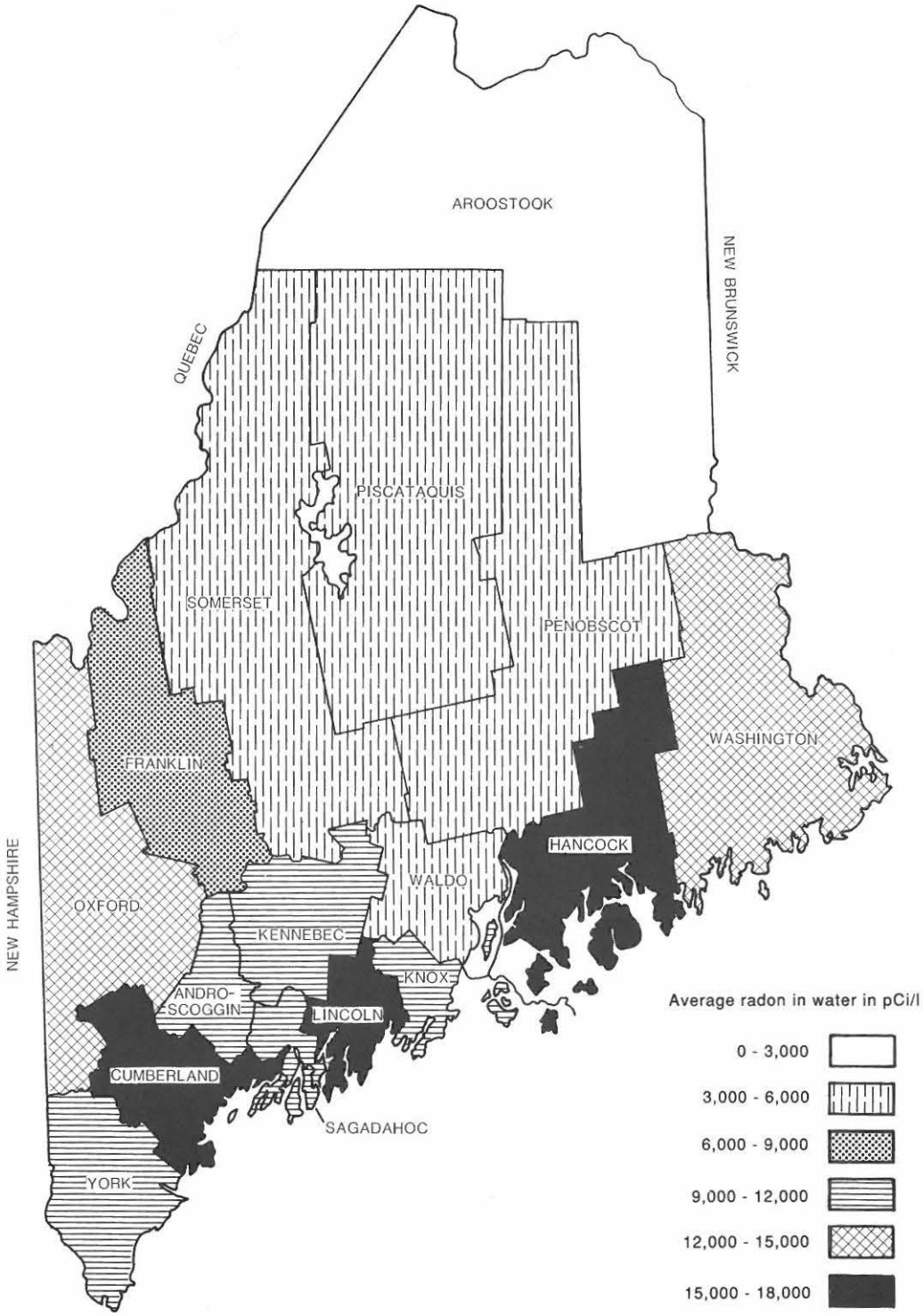

Figure 5. Map showing average values of radon concentrations in water for Maine counties.

but decrease the statistical significance of it (see Fig. 6). Significant correlations were observed for lung cancer and Rn and for all cancers and Rn. Figure 6 shows the comparison between Rn and lung cancer (trachea, bronchus, and lung combined) for the Maine population. The results are significant at the 0.05 level of confidence. The level of confidence for female lung cancer is 0.01. This is surprising because this group of women smoked much less than the contemporary population does.

The phenomenon of high Rn activity in ground water is not restricted to Maine. As shown on Figure 7, granites of ages similar to those in Maine are found throughout New England. Similarly, regions of sillimanite- and second-sillimanite zone rocks are common throughout much of New Hampshire, Massachusetts, and Connecticut (Thompson and Norton, 1968). Granitic gneisses and granites of Precambrian age are also common throughout western and southern New England. Thus one should anticipate that similar geology-Rn-health effects relationships may exist for New England in general (see e.g. Hall et al., 1987). Our reconnaissance of water quality in the rest of the states in New England indicates high Rn wherever the geology is suggestive of it.

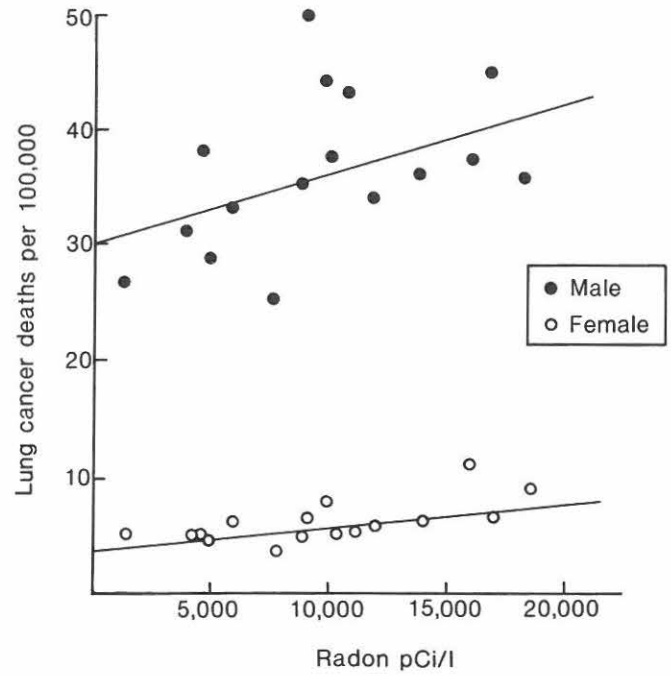

Figure 6. Age-adjusted mortality rates for lung cancer versus average radon concentration in water, by county.

ACKNOWLEDGMENTS

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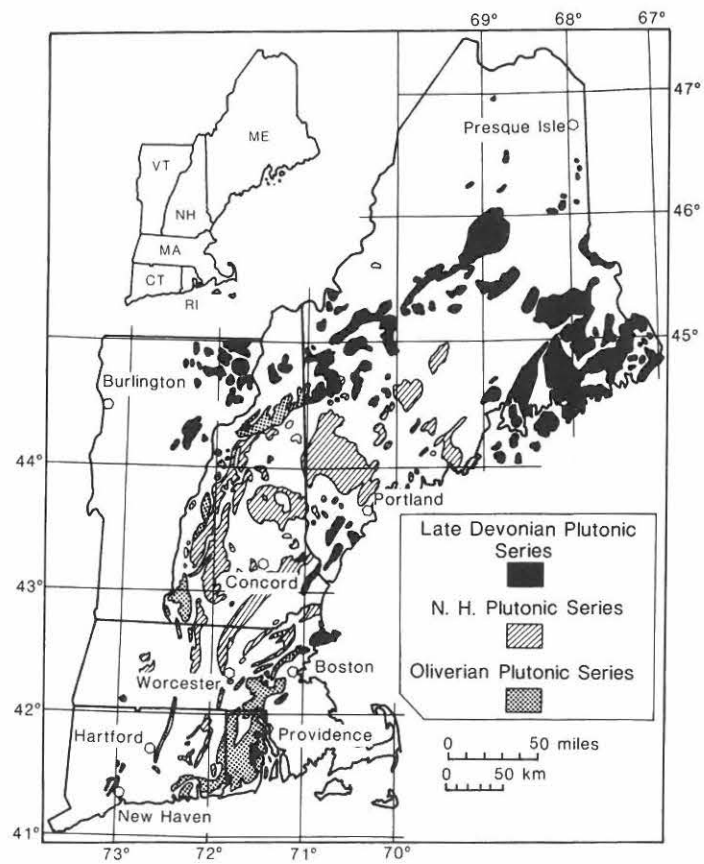

Figure 7. Geologic map of New England showing the distribution of Paleozoic to Cenozoic granitic rocks (from Page, 1968).

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