Environment International, Vol. 8, pp. 59-66, 1982
Printed in the USA.

0160-4120/82/070059-08$03.00/0

VARIATIONS OF AIRBORNE AND WATERBORNE Rn-222 IN HOUSES IN MAINE

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Concentrations of airborne radon ranging from 0.05 to 135 pCi/L were found in houses in Maine. Track-etch cups were placed in five positions for 100 houses to determine integrated average radon concentrations over the period October 1980–May 1981. To investigate the association between elevated radon concentrations in well water and the indoor airborne radon concentrations, the radon in the water supplies of these houses was measured by liquid scintillation. Monitors of airborne radon, recording in intervals of 10 min for periods of 5–7 days, were used for dynamic studies in 18 houses, determining the component of airborne radon associated with major water uses, such as showers, laundry, and dishwashing, which liberate radon in bursts. House residents kept logs noting the time of major water uses. For some of the houses, ventilation rates ranging from 0.3 to 2 air changes per hour were determined by analysis of the dynamic data. The component of airborne radon associated with water sources was found to vary inversely with ventilation rate and directly with waterborne radon concentration, with 0.8 ± 0.2 pCi Rn/L air per nCi Rn/L water at a ventilation rate of 1.0 air change per hour. The data are pertinent to a study which has revealed significant correlations between county averages, from the National Cancer Institute, or age-adjusted cancer mortality rates in Maine and average values of radon concentrations in water for the counties.

Introduction

Measurements of radon in the air of houses have been of interest in recent years due to the expectation of increased radiation doses to lungs as house ventilation rates are reduced for energy conservation. Radon in the air in uranium and other mines has been found to cause lung cancer, and, as a result, radon levels are presently controlled to below prescribed limits by forced ventilation of the mines with fresh air. Radon levels in the air in houses are generally much lower than levels which caused lung cancer in the poorly ventilated mines, but people spend much more time in houses and other buildings, perhaps as much as 90% of their lives. If an assumed linear relation between radiation dose and cancer risk is correct, then the low doses experienced by large populations from normal radon levels in the air in houses may produce significant numbers of lung cancers, though the individual risk is usually low compared to the previously uncontrolled uranium mine situations.

The current standard for uranium miners requires that annual exposures from radon daughters be below three working level months per year, corresponding to a 0.25 working level exposure rate for 12 months at 170 working hours per month. A person will receive an exposure of three working level months if he spends 18 h every day for a year in a house with air having 0.078 working levels of radon daughters. A few of the radon levels and corresponding working levels measured in houses in the present study are of this order, indicating that the risk of lung cancer from radon in some houses may be significant whether or not the linear dose-response hypothesis is correct.

The risk from radon and its daughters in houses has been estimated in a recent U.S. Environmental Protection Agency study (Guimond *et al.*, 1979). Using standard models for the linear extrapolation of dose rates and health effects from uranium mine conditions to household conditions, the lifetime risk from lifetime residence in housing having 0.02 working levels was estimated to be 1000 to 3000 lung cancer deaths per 100,000 persons.

Measurements of radon levels in the air of dwellings are underway in a number of countries. Reports of surveys of radon levels have been published for houses in England (Cliff, 1978), Norway (Stranden *et al.*, 1979), Sweden (Swedjemark, 1980), Hungary (Toth, 1972), Austria (Steinhousler *et al.*, 1978), and Poland

(Pensko, 1969). Radon and radon daughter measurements have also been reported for houses in Ontario (Taniguchi and Vasudev, 1980), (Dilworth, *et al.*, 1979), Saskatchewan (Keith, 1978), 13 major Canadian cities (McGregor, 1980), New Jersey and New York (George and Breslin, 1979), New York (Fleischer *et al.*, 1981), California (Nazaroff, 1981), Illinois (Toohey, 1981), Florida (Guimond *et al.*, 1979), and Colordo (C. E. McGuire, Inc., 1979).

In some regions where radon levels in household air have been found to be high, due to enhancement of the radium in soil or building materials, usually from use of materials with high radium content brought to the surface in uranium or phosphate mining, criteria for radon levels in air above which remedial action is recommended have been established by governmental agencies. In Grand Junction, CO, where houses have been built on uranium mine tailings, the U.S. Surgeon General's guideline for remedial action is 0.05 working levels, with action suggested for levels between 0.01 and 0.05 working levels as well (U.S. Government, 1972). For houses built on inactive uranium processing sites, the U.S. Environmental Protection Agency recommendation calls for remedial action for houses with more than 3 pCi Rn/L air (USEPA, 1980). In Sweden, the criteria allow a maximum of 1.9 pCi/L for radon daughters in new dwellings, 5.4 pCi/L in rebuilt dwellings, and 10.8 pCi/L in existing dwellings (Åkerblom and Wilson, 1981). The high levels found in the air of some Swedish houses to which these criteria apply are due to natural radium content of soils and building materials, with no technological enhancement.

The state of Maine has significant fractions of its surface area covered with granites and other rocks bearing high radium concentrations. Granites in Maine have up to 25 ppm (μg/g) uranium, the ultimate progenitor of radon. Radon in water from 136 wells in granitic regions of Maine has been measured to average 22,000 pCi/L, with 13,600 pCi/L for 35 wells in sillimanite terrain, and 1100 pCi/L for 56 wells in chlorite grade terrains (Hess *et al.*, 1979). These conditions indicate a possibility for strong emanation of radon from the soil and rocks surrounding the foundations of houses, with consequent high inputs of radon through cracks or openings in foundations, and possibly high inputs of radon into houses with use of water from wells with high radon content. In addition, if construction materials are taken from local rock quarries, they may emanate radon directly into the household air from their radium content.

The concentration of radon in household air depends on the magnitude of each of these sources, the ventilation rate for mixing with outside air, and the radioactive decay rate for radon. The concentration of radon is determined by

$$dA/dT = R_{sg} + R_{bm} + R_w + R_o - (\lambda_v + \lambda_d)A,$$

where A is the pCi radon-222 per liter of house air, t is time, λ_d is the decay rate for radon 222, λ_v is the ventilation or air-change rate for the house, R_{sg} is the input of radon with soil gas, in pCi/L h, R_{bm} is the input for radon from building materials, R_w is the radon input from the water supply, and R_o is the input of radon with outside air. Any of the sources can be time dependent for a variety of reasons; for example, showering liberates a burst of radon during the short time it lasts, and changes in atmospheric pressure and humidity affect rates of soil gas penetration through foundations and outgassing of construction materials. Ventilation rates, λ_v, are normally greater than 0.1 air change per hour, while the decay rate for radon-222, with a 3.8-day half-life, is 0.0076/h, so λ_d is not significant in affecting indoor airborne radon concentrations. A solution for the equation, for the case where all sources are constant in time and are represented together as R, is

$$A = R/\lambda_v.$$

It can be seen that measurements of radon in air, A, will allow characterization of the sources of input, R, only if ventilation rates are determined along with the radon levels.

Methods

A set of 500 F-1 track etch cups (Fleischer *et al.*, 1980) was placed, 5 per house, in 100 houses in October 1980 and collected in May 1981 to determine integrated average radon levels during the cold season, when ventilation would be expected to be minimal for economy in heating. Of all the houses, 55 were located in geological regions in which high levels of radon in water are frequently found. For each house, cups were placed in a first-floor bedroom, first-floor bathroom, basement, living room, or kitchen, and shed or other well-ventilated place outside the living area.

At the same time the set of cups was placed, water samples were taken for determination of radon concentrations by the liquid scintillation method of Prichard and Gesell (1977).

The detectors (Wrenn *et al.*, 1975) used for the dynamic data on air radon concentrations collect daughters from the decay of radon which has diffused through a foam cover into a 1-L volume. An electrostatic field attracts positive ions onto a thin aluminized mylar film covering a ZnS(Ag) phosphor which is viewed by a photomultiplier tube. Alpha decays of radon daughters on the mylar film were counted for periods of 10–20 min; the sums were automatically printed on paper tape at the ends of the periods. The two detectors used were calibrated for radon sensitivity and humidity dependence by placing them in a sealed drum with a radium-226 standard solution. For one of

these detectors, calibration was checked at radon facilities at New York University, at the Department of Energy Environmental Measurements Laboratory in New York City, at the University of Texas School of Public Health in Houston, and at the Eastern Environmental Measurements Laboratory in Montgomery, AL.

The 18 houses studied with Wrenn detectors were located between Orono and Bar Harbor, ME. For each house, a detector was placed in the living room or kitchen and left operating for 5–7 days, during which time the inhabitants were asked to keep a log, noting the times of major water uses such as showers, baths, dishwashing, and laundry washing.

For some of the houses, radon daughter working level measurements were made by the Kuznets method (Kuznets, 1956), which involves counting alpha decays of radon daughters gathered on membrane filters with an air sampling pump. In some instances of high radon concentrations, relative radon levels were determined with series of small air grab samples, using 24-cm³ liquid scintillation vials coated on the inside with ZnS(Ag) powder over silicon grease. These grab samples, referred to as Z cells, were counted in a liquid scintillation machine, giving results useful for radon concentrations above 5 pCi/L air.

Results

Track etch cups

The results for radon concentration averages measured with track etch cups in the five different positions in the houses are shown in Fig. 1 as numbers of houses versus radon concentrations in 1 pCi/L increments. The number of cups collected, from which data was obtained, ranges from 68 to 85 for the five positions. The data for radon levels outdoors or in very drafty sheds has an arithmetic mean of 0.8 pCi/L. The averages for the other positions are 3.0 pCi/L in bedrooms, 3.4 pCi/L in bathrooms, 3.1 pCi/L in kitchens and livingrooms, and 6.4 pCi/L in basements. Deletion of the data for two houses with extraordinarily high overall radon levels reduces the average for each position by about 10%.

The average basement radon level is nearly twice as high as the averages in the other rooms. This indicates that radon entered the houses in the basements, possibly with soil gas diffusing through foundations or entering via drain pipes, or by emanation from basement water wells or basement building materials. Reduction of radon concentrations on the first floors may indicate that they have weaker radon sources or higher ventilation rates than the basements. A few track etch cups placed in second-floor bedrooms indicated that second-floor radon concentrations were lower than first-floor concentrations by roughly a factor of two.

To test the hypothesis that radon in water supplies is a source of airborne radon, radon in bathroom air, as measured in the track etch cup studies, is plotted versus radon in the corresponding water supplies in Fig. 2. The regression line shown for the data is significant, with $r = 0.307$, $n = 85$ measurements, slope = 0.107 pCi/L air per nCi/L water, and intercept at 1.6 pCi/L, indicating that water may be the source for some, but not all of the radon in the air. The results for radon in air in other positions in the houses show similar correla-

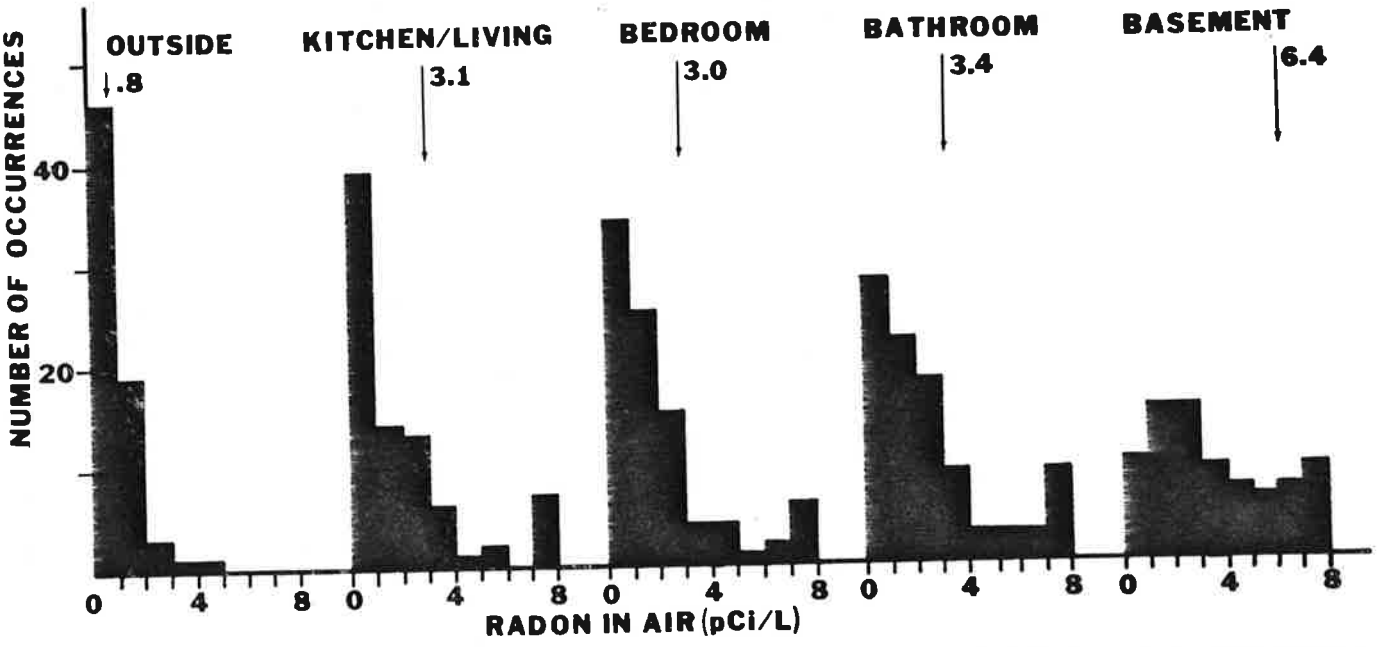

Fig. 1. Histogram of number of houses vs radon concentration in air measured with track etch cups in kitchens or livingrooms, bedrooms, bathrooms, basements, and outside. Occurrences of greater than 8 pCi/L are included in the eighth increment for each place.

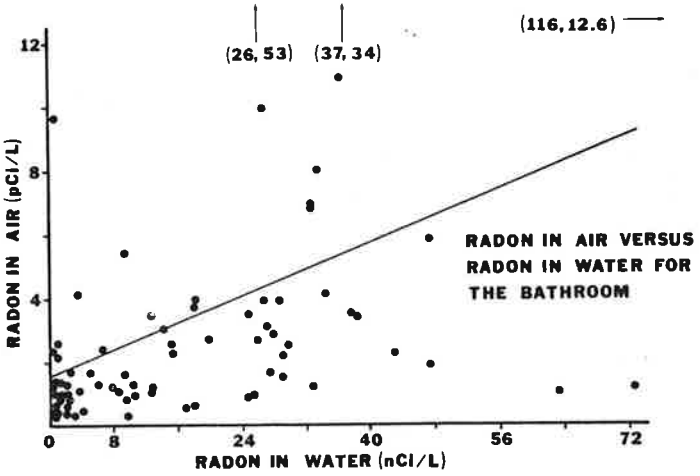

Fig. 2. Radon concentrations in bathroom air measured with track etch cups vs radon in water measured by the liquid scintillation method.

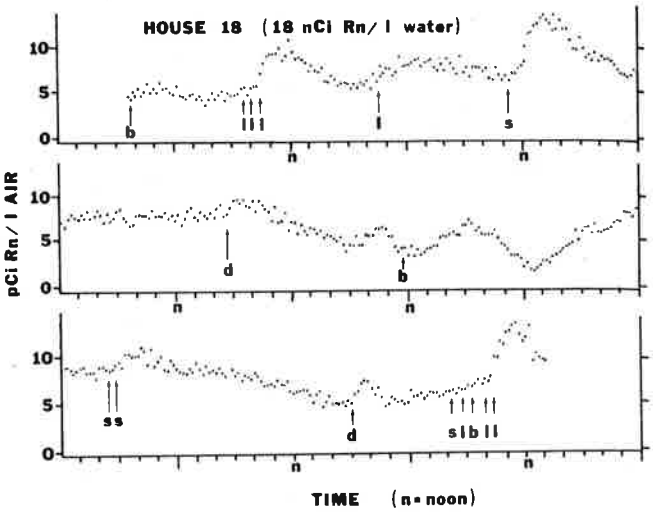

Fig. 4. Radon concentration in house 18 vs time of day for 7 days. Water uses marked as in Fig. 3, with b for bath and d for dishwasher use.

tions with water supply radon levels. Sources, such as soil gas, which contribute to the air radon levels recorded by the track etch method, are probably also correlated with radon in water.

Dynamic studies

Portions of the data from the continuously recording radon detectors are shown in Figs. 3–5. The times of various water usages are indicated and explained in the figures. Generally, a spike of radon shows up in the air shortly after a shower or the use of a washing machine or dishwasher, and sometimes a low spike can be seen after a bath was run. The times indicated for water uses are those given by the inhabitants of the houses, and are approximations of varying accuracy.

The data presented in Fig. 3 are for a typical house with a relatively low radon level in the water supply (6.5 nCi/L) and a low ventilation rate, while Fig. 4 shows data from a house with more radon in the water supply (18 nCi/L), a low ventilation rate, and a significant continuous background radon level on top of which the spikes from water use appear. Portions of data from two houses are presented in Fig. 5; those for house 7 (27 nCi/L) show little radon other than that associated with water use, and house 18 was exceptional in that it had the highest average radon in air, 87 pCi/L. For this tightly sealed solar house, the variations in the radon concentrations associated with showers or other water uses were not significant enough to show up against the high background. The source of the radon was an integral greenhouse; its strongly emanating floor was crushed gravel over granite bedrock, with no impermeable interface.

Table 1 presents a summary of the results for the 18 houses in which dynamic radon levels have been measured.

In determining the ventilation rates of the houses from the characteristics of the radon spikes associated

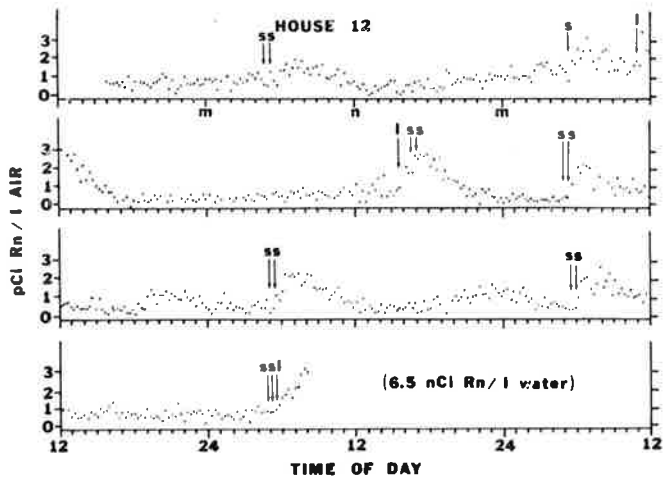

Fig. 3. Radon concentration in house 12 vs time of day for 7 days. Noon marked n. Showers are marked with letter s, and laundry with l.

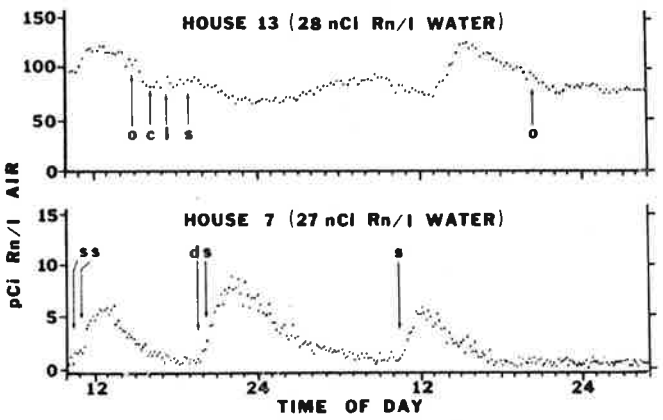

Fig. 5. Radon concentrations in houses 13 and 7 vs time of day. Water uses marked as in Fig. 3, with o for opening window and c for closing window.

Table 1. Summary of data for radon in 18 houses.

No.	Town	Date	Residents	Water Rn (pCi/L)	Mean Air Rn (pCi/L)	Rn Daughters Working Level	Air Rn from Water (pCi/L)	Ventilation Rate (Air Change/h)
1.	Orono	6/79	2	330	1.1	–	<0.3	<1.0
2.	Orono	7/79	3	330	0.6	–	0.3	–
3.	Orono	7/79	3	330	0.6	–	0.3	–
4.	Dedham	7/78	4	52,000	2.6	–	1.9	2.0 + 1
								– 0.5
5.	Prospect	1/80	4	25,000	9.	0.03	–	–
6.	Orrington	2/80	3	17,000	3.0	–	1.7	0.5 ± 0.1
7.	Dedham	5/80	4	27,000	3.2	0.008	3.2	0.4 ± 0.1
8.	Dedham	5/80	2	22,000	2.0	–	<1.5	–
9.	Dedham	5/80	4	25,000	2.0	–	<1.5	–
10.	Mt. Desert	10/80	2	6,000	1.5	–	–	–
	Mt. Desert	7/80	2	7,000	0.25	–	–	–
11.	Dedham	10/80	2	8,000	5.2	–	<1	–
12.	Dedham	10/80	2	6,500	1.5	–	0.7	0.5 ± 0.1
13.	Dedham	10/80	3	28,000	87	0.16	<5	<0.5
		10/80			201	0.26		–
						(Greenhouse)		
14.	Mt. Desert	10/80	2	20,000	3.0	–	–	–
15.	Mt. Desert	11/80	2	28,000	9	–	4.5	0.4 ± 0.1
16.	Dedham	11/80	4	31,000	35.	0.075	–	–
		11/80			–	0.30		–
						(Basement)		
17.	Clifton	11/80	5	35,000	4.6	–	–	–
18.	Mt. Desert	11/80	5	19,000	8	–	3	0.3 ± 0.1

with water use, slowly falling spikes will be associated with low ventilation rates, and vice versa. In analyzing spikes, the characteristic response of the radon detector must be known. Figure 6 shows the response of a radon detector to a short burst of radon from a shower in a house with a very high ventilation rate (house 4, 2 air changes/h). In the same figure are plotted relative instantaneous radon levels, determined by grab samples with Z cells taken at the same location as the radon detector (in the living room). The response of the radon detector is delayed, because of the time for diffusion of the radon through the foam cover of the detector and the buildup time of the 3-min polonium-218 alpha activity. The delay time for transport of the radon from the shower to the vicinity of the detector in the living room can also be seen in this figure. The decrease of alpha activity on the detector is associated with the 20 and 27 min (half-life) beta activities, ^{214}Pb and ^{214}Bi, which delay the ^{214}Po alpha particles.

To determine ventilation rates from the data for the radon bursts, we have calculated solutions to the Bateman equations for the theoretical time dependence of the alpha-emitting radon daughter activities on the Wrenn detector scintillator. An exponentially decreasing radon concentration in the room due to ventilation is assumed. The delay for the diffusion of the radon through the foam cover of the detector has been treated as an additional first-order differential equation in these calculations, with λ (diffusion) determined, from detector response data such as in Fig. 6, to be 5/h. Theoretical detector response curves calculated in this manner for several ventilation rates are presented in Fig. 7. For high ventilation rates, the curves fall off at a rate determined chiefly by the 20 min ^{214}Pb and 27 min ^{214}Bi precursors of the 0.03 ms (half-life) ^{214}Po alpha emitter, the radon in the air around the detector having been eliminated by ventilation before the collected daughters decay. For low enough ventilation rates, the differing rates of decrease predicted for activity on the detector permit significant discrimination among the ventilation rates. Fits of some calculated curves to data for radon bursts are shown in Fig. 7. Those house ventilation rates which could be reliably determined from the dynamic data are given in the last column of Table 1.

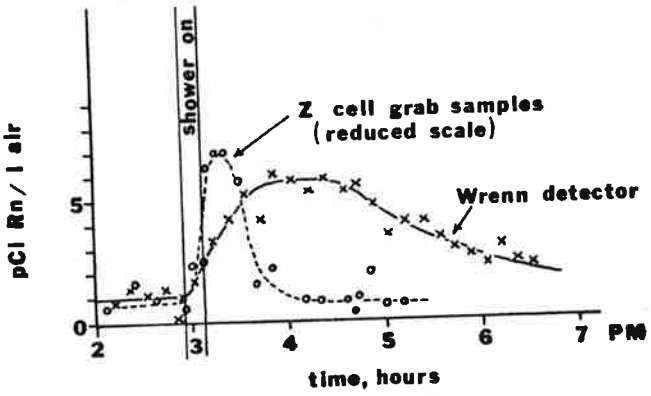

Fig. 6. Radon concentration after shower as measured in the living-room air with grab samples (Z cells) and a Wrenn electronic radon detector.

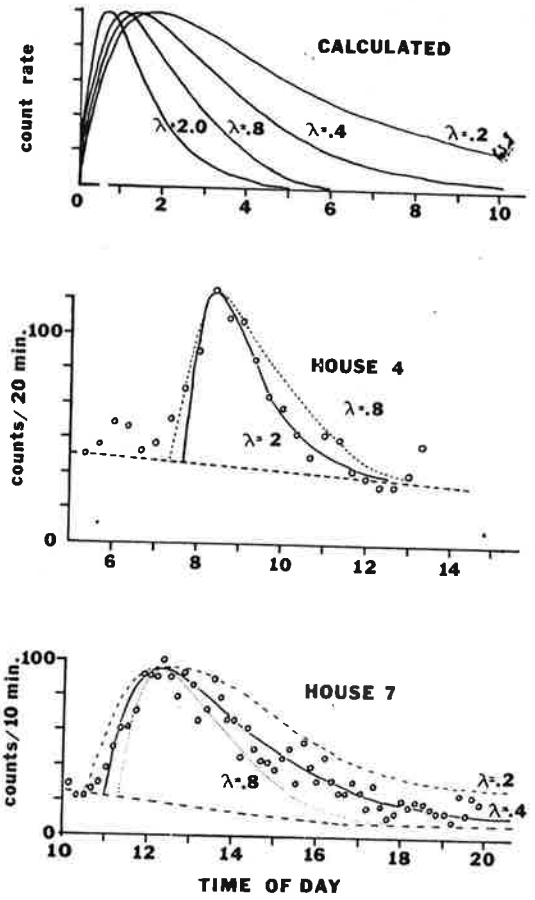

Fig. 7. Calculated theoretical responses of a Wrenn detector to exponentially decreasing radon concentrations as a function of ventilation rate, in air changes per hour, and fits of calculated curves to data for houses 4 and 7.

For most of the houses, the average concentration of radon in the air due to the water supply source can be determined from the data for the component of the radon appearing in jumps at times of water uses logged by residents. The averages of the components appearing in bursts at these times are given in the eighth column of Table 1. In addition to this component, there are continuous, slowly varying, and weak emissions of radon from water used in sinks, baths, and toilets.

The outgassing of radon from static water and small water uses cannot be distinguished from the continuous radon levels from other sources, so their contributions have been estimated indirectly. Radon emanation rates for static and agitated water in representative surface-to-volume geometries have been measured in the laboratory by monitoring the radon concentration in the water with the liquid scintillation method. The water radon concentration decreases exponentially at a constant rate which depends on the agitation. For water used in showers and dishwashers, the decrease in radon concentration has been directly determined by measuring the concentrations in the input and discharge water. The results for the loss of radon from water in various uses are presented in the third column of Table 2.

From the information summarized in Table 2, including statistics (Kreissl, 1978) for average per capita water consumption rates for various uses, the slowly and weakly emanating water sources for airborne radon (sinks, toilets, and baths) can be estimated to be about one-third as strong as the sources which liberate radon in large, observable bursts (showers, laundries, and dishwashers). Thus, the values in column eight of Table 2 for average levels of airborne radon liberated from water in observable bursts should be increased by about 33% to obtain the total average radon concentration in air due to radon in the water supply.

In Fig. 8, the levels of airborne radon from water, normalized for a water concentration of 10 nCi/L for each house, assuming direct dependence of airborne radon from water supplies on water radon concentrations, is plotted versus ventilation rate. The figure indicates that the air radon concentrations depend inversely on ventilation rates. After correcting this amount, because the total airborne radon from all water sources will be about 33% higher than the component appearing in bursts which is plotted in Fig. 8, there are seen to be 0.8 pCi Rn/L of air per 10 nCi Rn/L of water in a house with ventilation rate 1.0 air change/h.

Total radon concentrations in air for the 18 houses have been plotted as stars versus water radon concentrations in Fig. 9. Open circles in the figure are the average radon air concentrations due to water supply use and coming in noticeable bursts, these averages being normalized, according to an assumed inverse ventilation rate dependence, to those expected for a ventilation rate of one air change per hour. With a 33% increase for slowly and weakly emanating water sources of airborne

Table 2. Outgassing of radon from water use.

Water Use	Liters/ Person Day (Kreissl, 1978)	Fraction of Radon Lost	No. of Measurements This Study	pCi Rn Liberated (Person day pCi/L Water)
Dishwasher	19	0.98	1	18
Shower	20	0.65	4	13
Laundry	40	0.9 (estimated)	0	35
Sink	20	0.3 (estimated)	1	6
Bath	18	0.3 (estimated)	0	6
Toilet	35	0.3 (estimated)	0	12
				90 TOTAL

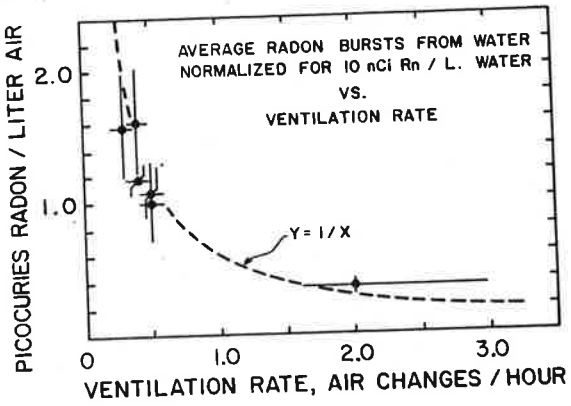

Fig. 8. Average radon in air from water use pulses, normalized to 10 nCi Rn/L of water, vs ventilation rate.

radon, the rate of air radon concentration dependence on water radon concentration is again 0.8 ± 0.2 pCi Rn/L of air per 10 nCi Rn/L of water.

The estimated exposures from airborne radon from all sources range from 0.01 to 14 working level months per year for the residents of these 18 houses when corrections are made for increased ventilation in the summer months.

Correlation

To investigate the significance of exposures to high levels of radon in water and corresponding high levels of radon in air, the correlation (Hess *et al.*, 1979) between age-adjusted cancer rates for the 16 counties in Maine (Mason, 1975) and county averages for radon concentrations in water, as estimated from more than 2000 water samples (Hess *et al.*, 1979) was calculated. The numbers of cases of lung cancer were 3682 for males and 656 for females from 1950 to 1969. The data for the correlation calculation are shown in Fig. 10. Age-adjusted lung cancer deaths per 100,000 person years for

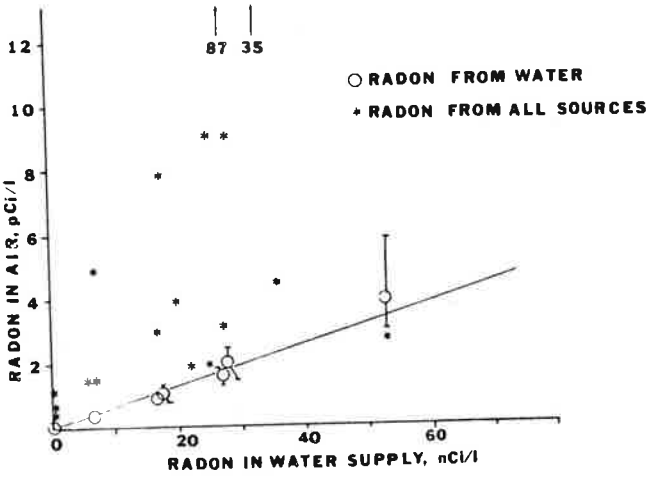

Fig. 9. Mean values of radon concentrations, air vs water, for 2–7 days during the heating season. The asterisks are for total radon observed, while the circles indicate average values due to water use pulses, normalized to 1 air change/h.

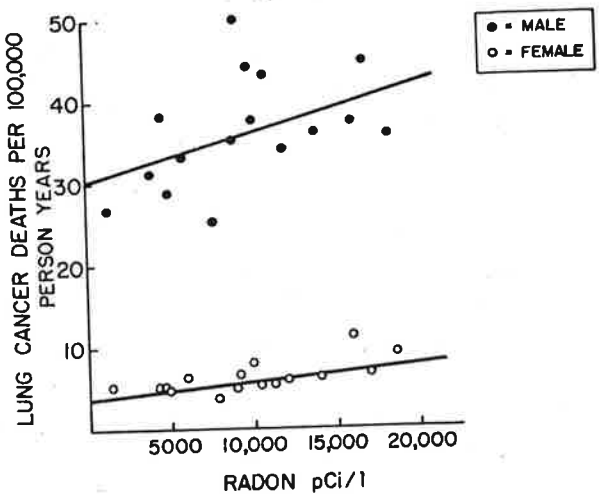

Fig. 10. Age adjusted lung cancer rates for males and females, in deaths per 100,000 persons per year, for 16 Maine counties, vs water radon concentrations for the counties.

males and for females in each country are shown versus areally averaged water radon concentrations for the counties. The regression line for males has a slope of 6.0 cases per 100,000 person years per 10,000 pCi Rn/L of water and an intercept of 30 cases per 100,000 person years. The correlation coefficient r equals 0.46, with a 0.05 probability of being spurious. The regression line for females has a slope of 2.0 cases per 100,000 person years per 10,000 pCi Rn/L of water, and an intercept of 3.8 cases per 100,000 person years. For females, the correlation coefficient r equals 0.65, with less than 0.01 probability of being spurious. Significant correlations were also found for all cancers together versus county averages for radon in water. The methods used parallel those of Segal (1962), who studied background radiation in New England.

Sources of airborne radon in houses other than water may be correlated with radon in water. Radon may diffuse from aquafer minerals into houses along with other soil gases, and locally obtained construction materials may have radon emissivities correlated with radon concentrations in well water.

Correlations of this sort cannot by themselves show causality. Among the causes of lung cancer are cigarette smoking and exposure to asbestos, as well as radiation exposure. A complete epidemiology study of lung cancer patients and their exposures to these agents is needed to show the magnitude of risk from each agent.

Conclusions

The average radon level measured with the track etch cups in the living areas of the houses was higher than 3 pCi/L air, which is the recommended criterion for remedial action for houses built on inactive uranium processing sites. The radon levels measured in air are significantly correlated with concentrations of radon in the water supplies for the houses, with a regression line

suggesting about 1.07 pCi Rn/L of air per 10,000 pCi Rn/L of water.

Dynamic studies with continuously recording radon detectors show bursts of radon released into the air at times of showers and uses of dishwashers and laundry machines. Ventilation rates can be derived from this data, which permit the conclusion that the component of airborne radon from radon in the water supplies depends inversely on ventilation rate and direction on concentration in the water supply, with 0.8 ± 0.2 pCi Rn/L air per 10,000 pCi Rn/L water at a ventilation rate of 1.0 air change/h.

Estimates of average levels of radon in the water supplies of the 16 Maine counties, based on over 2000 measurements, are significantly correlated with lung cancer rates in the counties. Smoking, asbestos, and other kinds of air pollution are known to be significant contributors to lung cancer, and it may well be that the correlations are influenced by geographical patterns for such factors. Geographical data to remove the effects of these additional variables from the correlations are not available. Under these conditions the correlations should only be interpreted as suggesting an association, and conclusions cannot be drawn without additional epidemiology studies of more comprehensive design.

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