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VARIATION OF RADON SOURCES AND INDOOR RADON ALONG THE SOUTHWESTERN EDGE OF THE CANADIAN SHIELD

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Radon source and indoor radon concentrations were measured in 240 homes. Waterborne radon concentrations ranged from 0.4 to 360 kBq/m^3, with a geometric mean value of 8.6 kBq/m^3 (230 pCi/L). Dry soil samples showed an average radon emanation of 16 Bq/kg (0.43 pCi/g) and an average radium content of 26 Bq/kg (0.7 pCi/g). In situ, surface soilborne ^{214}Bi concentrations ranged from 5 to 50 Bq/kg. Yearly average, indoor airborne radon concentrations ranged from 7 to 3800 Bq/m^3 (0.2 to 103 pCi/L) and varied significantly by season and by compartment. Radon concentrations in living spaces ranged from 7 to 2100 Bq/m^3, with an average value of 130 Bq/m^3 (3.6 pCi/L) and a geometric mean of 100 Bq/m^3 (2.7 pCi/L). Source and airborne radon concentrations are best described by log-normal distributions. Localized clusters of houses showed significant variation in source and airborne radon concentrations. Regional and geological clusters showed no significant variation in airborne radon, but did evidence some significant variation in source concentrations. Linear regression analysis of airborne radon in below-ground compartments and radon sources indicated significant correlation for emanative radon at individual sites, local clusters, and regional clusters and for ^{214}Bi concentrations in local clusters.

INTRODUCTION

Previous studies have shown that indoor airborne ^{222}Rn concentrations can depend upon geology, hydrology, meteorology, housing stock, and life style. (Gessell 1983; Nero 1983). Few studies have measured and analyzed all these important factors simultaneously in a large sample of homes. Yet agencies and individuals faced with the problem of trying to identify high-radon homes have often used simple, single factors as screening tools for planning and analysis. For example, major surficial geology and bedrock features have been used to assess radon potential in some states. One goal of the present study is to investigate the correlation between radon sources and indoor airborne radon in a substantial number of homes located in a variety of geological terrains. If laboratory-based source measurements or simple field tests are strongly correlated with indoor radon, then locating radon-plagued homes would become easier. In order to sample our surface geology and housing stock, 240 houses were surveyed between 1983 and 1987. These houses are scattered throughout a 3 x 10^5 km^2 area of Minnesota, Northern Wisconsin, and the Upper Peninsula of Michigan. The surface geology of the region is dominated by deposits from the Wisconsin glaciation occasionally punctured by Precambrian bedrock. Bedrock within the study area has the potential of containing significant deposits of uranium. In addition, the climate encourages energy-efficient housing and indoor living. The geological diversity, radon source potential, and housing patterns make the southwestern edge of the Canadian shield an attractive place to test and develop radon-prediction methods.

METHODS

Survey

The survey was organized geographically by town-sized areas, (approximate size: 100 km^2). These towns were selected on the basis of surface geology and a measurement of the local radon-related gamma flux. Volunteers were solicited through each town's high school physics teacher. This process yielded an average of five houses per town in 44 towns. At each house, the average indoor airborne concentration was measured in the lowest two living compartments for periods lasting from seven months to one year. In addition, samples of the water supply, subsurface soil near the house, and local radon-related gamma flux were taken. The homeowner completed a questionnaire that included questions on building location, materials, structural integrity, and ventilation.

Waterborne

Waterborne radon concentrations were measured using a small-sample, liquid scintillation technique (Prichard and Gessell 1977). The system was calibrated with RaCl solutions whose activity was known to within 10%. Although the samples were counted until the statistical uncertainty in the count was 5%, the radon concentrations drawn from the same source showed a variation of approximately 10%. The detection limit for the system used is 0.6 kBq/m^3 (15 pCi/L).

Soilborne

Three methods were used to characterize soilborne radon source strengths. Field measurements of the 1.6 to 2.6 MeV gamma flux gave a rapid, relative measure of the surface soil's ^{214}Bi concentration. The flux was measured with a shielded, carborne 7.6 x 7.6 cm NaI scintillator. This system was calibrated at two flat sites where the surface ^{214}Bi concentrations were estimated from bulk soil samples analyzed for residual ^{214}Bi concentrations by high-resolution gamma spectroscopy. The flux was measured during extended periods of stable weather in order to minimize meteorologically related variations. The reproducibility of this flux measurement is approximately 30%.

At each house, soil samples for laboratory analysis were collected from a depth of approximately 0.5 m within 1 m of the structure. Soilborne radium concentrations of aged, sealed samples were measured using a high-resolution gamma spectroscopy system. The system was calibrated with standard reference material (SRM4353) obtained from the National Bureau of Standards, whose radium activity is known to within 10%. Samples were counted until the statistical uncertainty in the count was approximately 10%. Multiple subsurface soil samples drawn from a 100 m long transection of a single site showed concentration variations of 25%. The detection limit for the system used is 1 Bq/kg (0.02 pCi/g).

Emanative soilborne radon concentrations were measured for dry soil samples using a liquid scintillation technique. Five milliliters of ground, compacted soil in a polyethylene vial was placed inside a 20 mL glass vial containing 5 mL of a standard toluene-based scintillation cocktail (NEN Liquiflour). The polyethylene vial was sealed with polyethylene tape to inhibit ^{220}Rn emanation. Samples were aged for one month and measured for a activity on a Beckman 100C liquid scintillation counter. The system was calibrated with RaCl solutions whose activity was known to within 10%. Although the samples were counted until the statistical uncertainty in the count was 5%, the radon emanation of samples drawn from the same source showed a variation of approximately 10%. Multiple subsurface soil samples drawn from a 100-m long transection of a single site showed concentration variations of 20%. The detection limit for the system used is 1.5 Bq/kg (0.04 pCi/g).

Airborne

Time-averaged, indoor airborne radon concentrations were measured by track registration material (CR39) enclosed in either 250 mL or 50 mL containers sealed with a microporous polycarbonate membrane (Alter and Fleischer 1981; Fleischer et al. 1975). In each house, a detector was placed near the house's center in the two lowest compartments, usually at a height of approximately 2 m. While each site was surveyed for at least seven months, many sites were monitored for a year. After exposure, the CR39 was etched in NaOH, and the resulting pits were counted. Both kinds of detectors were calibrated by exposure to known radon concentrations at the Technical Measurements Center (Grand Junction, Colorado). Although enough pits were counted to insure statistical uncertainties of 10%, replicate measurements of the same radon atmospheres showed as much as a 15% variation for typical exposures (20 kBq-day/m^3).

RESULTS

Statistical summaries of the waterborne, soilborne and indoor airborne radon concentrations are shown in Table 1 and Table 2. Cumulative probability distributions are shown for waterborne, emanative soilborne, and indoor airborne radon in Fig. 1 through 3. Figures 4 through 7 show the variation of the geometric mean of radon sources and airborne radon for town-sized ($10^2 km^2$) geographic clusters of houses. Error bars reflect the uncertainty in the mean. Measurements from single-house towns are displayed without error bars. Figures 8 through 10 compare concentrations for regional ($10^4 km^2$) clusters. Radon source and airborne radon concentrations for houses clustered by surface geological feature are shown in Fig. 11 through 13.

DISCUSSION

Radon Sources

Both waterborne and soilborne radon source strengths are approximately log-normally distributed in homes located along the southwestern edge of the Canadian shield (see Fig. 1, 2, and 3). The means of these distributions, listed in Table 1, are near the U.S. averages (NCRP 1975; Nero 1983). When houses are considered together in geographical clusters, significant variation is present for local clusters (Fig. 4 to 6).

Table 1. Statistical summary of radon sources.

RADON SOURCE	NO. OF SAMPLES	UNITS	MINIMUM	MAXIMUM	GEOMETRIC MEAN	GEOMETRIC STANDARD DEV.
Waterborne	387	kBq/m^3	0.4	360	8.6	2.8
		(pCi/L)	(20)	(9800)	(230)	
Soilborne						
Surface ^{214}Bi	10^4 km	Bq/kg	5	50	~10	
		(pCi/g)	(0.1)	(1.5)	(0.3)	
^{226}Ra	38	Bq/kg	9	52	26	1.5
		(pCi/g)	(0.2)	(1.4)	(0.7)	
Emanative Radon	232	Bq/kg	6	33	16	1.3
		(pCi/g)	(0.2)	(0.9)	(0.4)	

Table 2. Statistical summary of yearly average indoor airborne radon.

LOCATION	NO. OF SAMPLES	UNITS	MINIMUM	MAXIMUM	ARITHMETIC AVERAGE	GEOMETRIC AVERAGE	GEOMETRIC STANDARD DEV.
Living spaces	240	Bq/m^3	7	2130	133	98	2.06
		(pCi/L)	(0.2)	(58)	(3.6)	(2.7)	
Below-grade compartments	219	Bq/m^3	30	3800	197	140	2.13
		(pCi/L)	(0.8)	(103)	(5.3)	(3.8)	
Above-grade compartments	229	Bq/m^3	7	1420	107	76	2.19
		(pCi/L)	(0.2)	(38)	(2.9)	(2.0)	

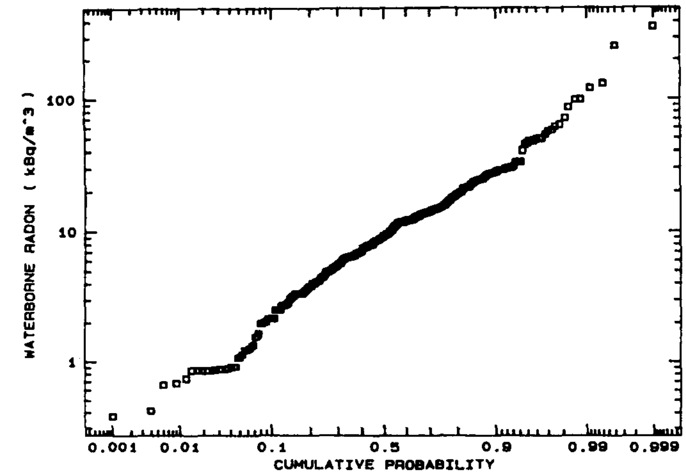

Fig. 1. Plot of observed waterborne radon concentrations as a function of the fraction of homes having that concentration or less. With these scales (log and probability), log-normally distributed data should lie along a straight line. Parameters or this distribution are listed in Table 1.

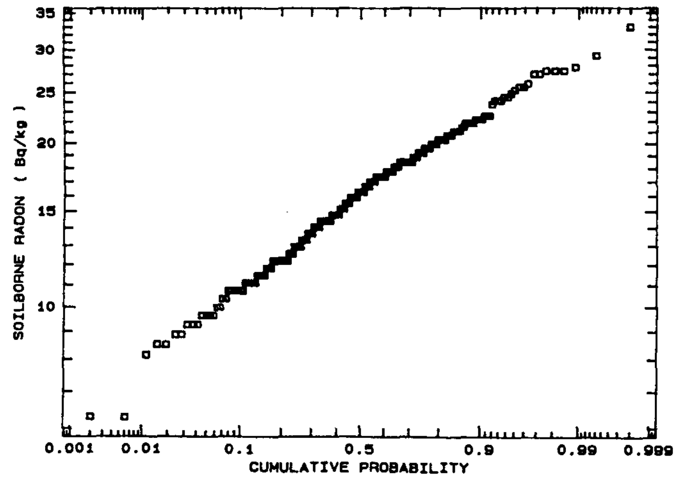

Fig. 2. Plot of observed emanative soilborne radon concentrations as a function of the fraction of homes having that concentration or less. With these scales (log and probability), log-normally distributed data should lie along a straight line. Parameters for this distribution are listed in Table 1.

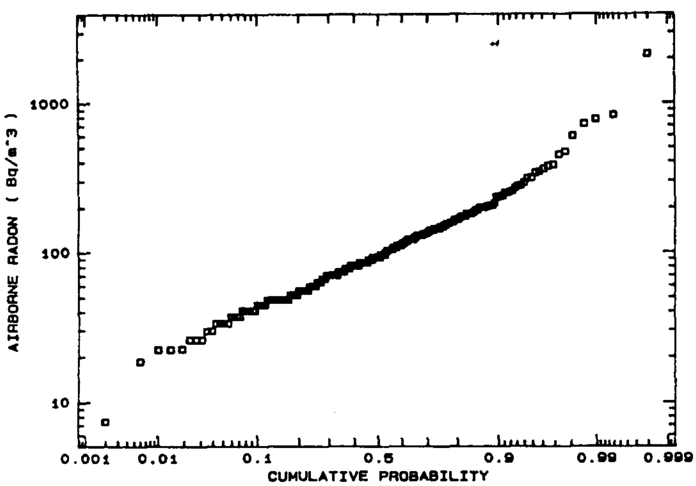

Fig. 3. Plot of observed indoor airborne radon concentrations in living spaces as a function of the fraction of homes having that concentration or less. With these scales (log and probability), log-normally distributed data should lie along a straight line. Parameters for this distribution are listed in Table 2.

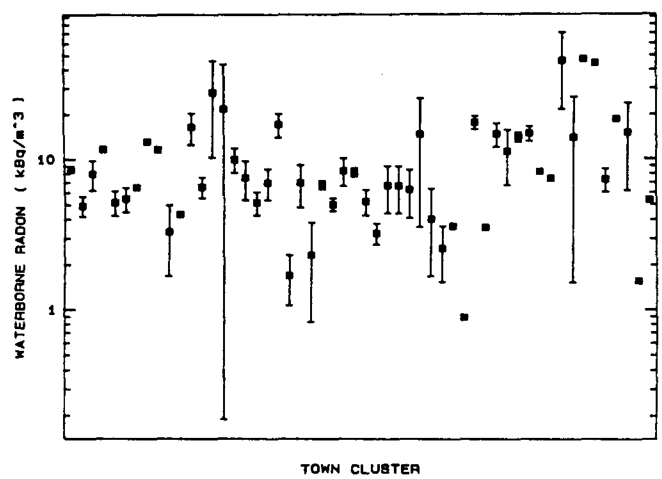

Fig. 4. Geographical variation of the geometric means of waterborne radon concentrations for town-sized (10^2km^2) clusters of houses. Uncertainties in the means are shown as error bars for those clusters that consist of more than one house. The first data point shows the geometric mean and standard error for all houses.

On a regional scale, the source concentrations show less variation (Fig. 8 and 9). To study the geological variation of radon sources on a large scale, houses have been clustered together by glacial lobe (Des Moines, Superior, Wadena, Rainy) or into a bedrock cluster when the glacial overburden is less then 10 m thick. As Fig. 11 shows, waters drawn from wells finished either in Superior lobe till or bedrock tend to have significantly higher waterborne radon. The variation of soilborne emanative radon (Fig. 12) apparent in regional clusters does not appear to be present in these larger geologic clusters.

Waterborne radon concentrations at most houses in the study were low enough that their contribution

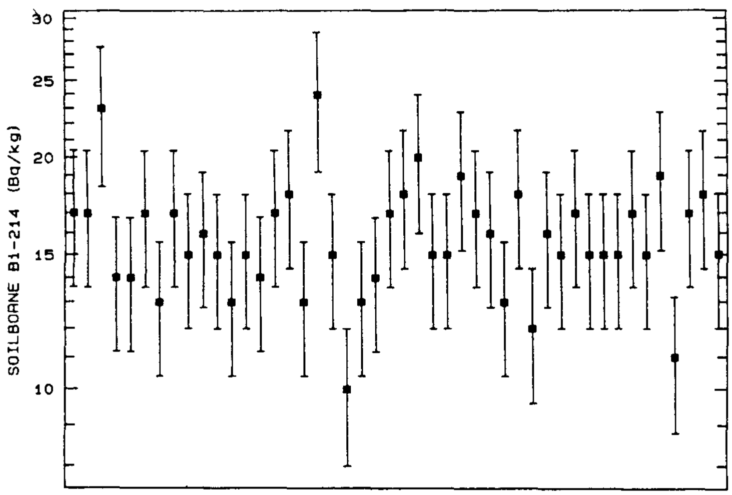

Fig. 5. Geographical variation of the geometric means of surface soil ^{214}Bi concentrations for town-sized (10^2km^2) clusters of houses.

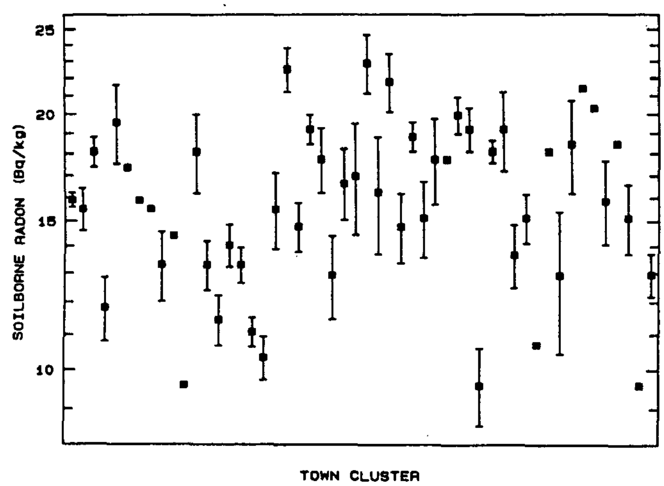

Fig. 6. Geographical variation of the geometric means of emanative soilborne radon concentrations for town-sized (10^2km^2) clusters of houses. Uncertainties in the means are shown as error bars for those clusters that consist of more than one house. The first data point shows the geometric mean and standard error for all houses.

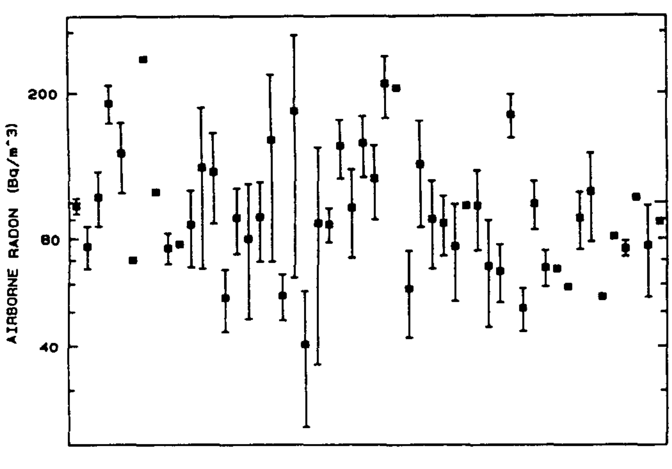

Fig. 7. Geographical variation of the geometric means of indoor airborne radon concentrations in living spaces for town-sized (10^2km^2) clusters of houses. Uncertainties in the means are shown as error bars for those clusters that consist of more than one house. The first data point shows the geometric mean and standard error for all houses.

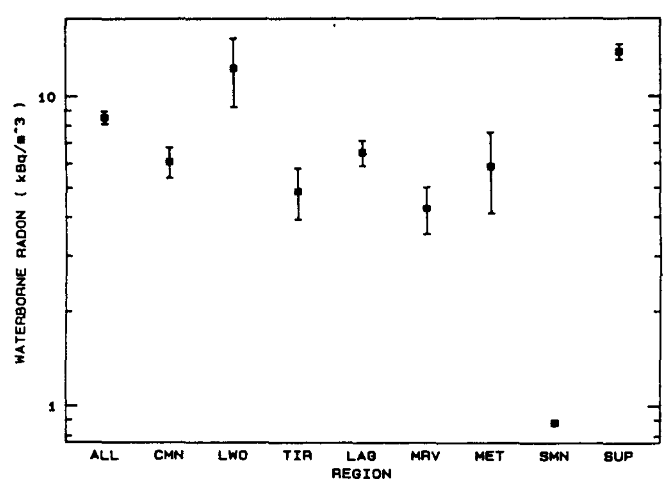

Fig. 8. Geographical variation of the geometric means of waterborne radon concentrations for regional (10^4km^2) clusters of houses. Uncertainties in the means are shown as error bars. The first data point shows the geometric mean and standard error for all houses.

to the airborne radon is expected to be minor (0.5 to 2%). (Nazaroff et al. 1987). No significant correlation was found between indoor airborne and waterborne radon concentrations for any cluster size (see Fig. 14). Thus, while waterborne radon may be an important contributor to airborne radon in a few individual houses, it is a poor indicator for airborne radon for the average house in our area.

In situ, surface ^{214}Bi concentrations in soils did show a positive correlation for town-sized clusters (99.9% probability) with indoor airborne radon in below-grade compartments (see Fig. 15). Variations within town-sized clusters of both ^{214}Bi and airborne radon concentrations limit the quantitative exploitation of this correlation. Surface ^{214}Bi concentrations may be useful as a screening tool for locating town-sized areas of elevated indoor airborne radon.

The radium content of 38 soil samples averaged 26 Bq/kg (0.7 pCi/g). Thus, it appears that the radium content of our soil is near the U.S. average (NCRP 75).

Soilborne, emanative radon concentrations have a reasonably strong positive correlation with indoor radon for sites (99.5% probability), towns, and regions (95% probability) (see Fig. 16 and 17). The variations within clusters of emanative soilborne radon and indoor airborne radon again limit the quantitative use of this correlation. It may be possible to combine the emanative radon measurement with physical soil characteristic measurements, like soil permeability, to improve this measurement's predictive power.

Bedrock, particularily granites, has often been cited as being closely associated with regions of elevated indoor radon. Since granitic rock lies under much of the study area, the data have been examined for bedrock effects. Approximately 25% of the houses in this survey have near-surface bedrock (10 m deep or less), and half of these are near granitic bedrock. The below-grade, indoor airborne radon is not much greater in houses situated over bedrock (including granite) than in houses built on glacial till. Towns

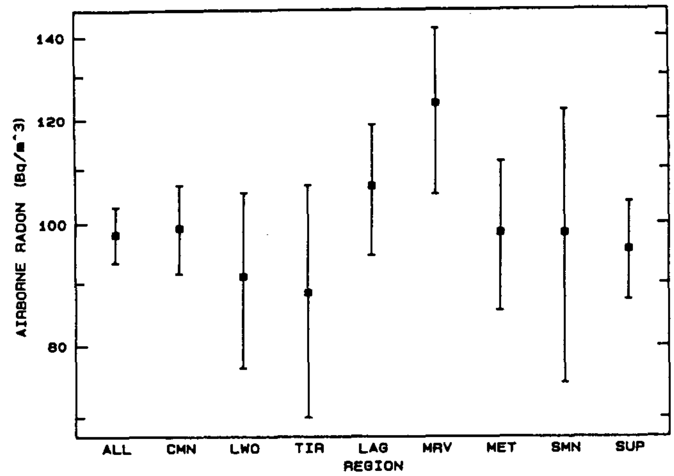

Fig. 10. Geographical variation of the geometric means of indoor airborne radon concentrations in living spaces for regional ($10^4 km^2$) clusters of houses. Uncertainties in the mean are shown as error bars. The first data point shows the geometric mean and standard error for all houses.

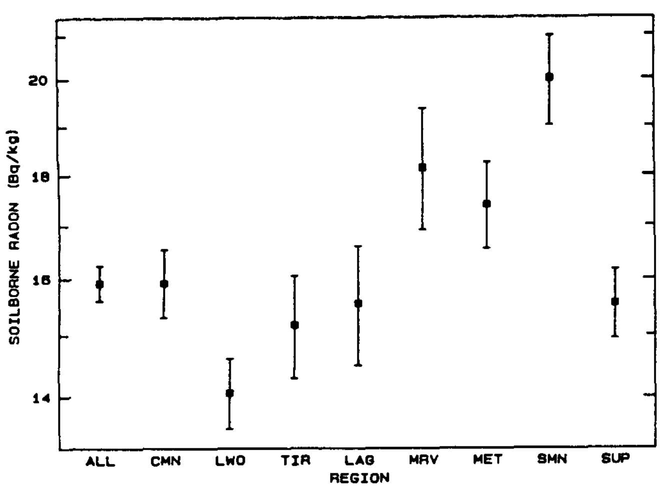

Fig. 9. Geographical variation of the geometric means of emanative soilborne radon concentrations for regional ($10^4 km^2$) clusters of houses. Uncertainties in the means are shown as error bars. The first data point shows the geometric mean and standard error for all houses.

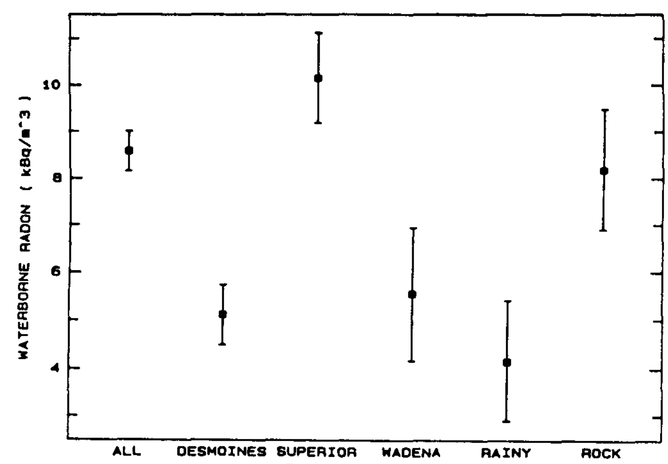

Fig. 11. Variation of the geometric means of waterborne radon concentrations for houses clustered by major surface geological feature. Uncertainties in the means are shown as error bars. The first data point shows the geometric mean and standard error for all houses. Clusters are named for lobes of the Wisconsin glaciation. Rock labels those houses located over bedrock within 10 m of the surface.

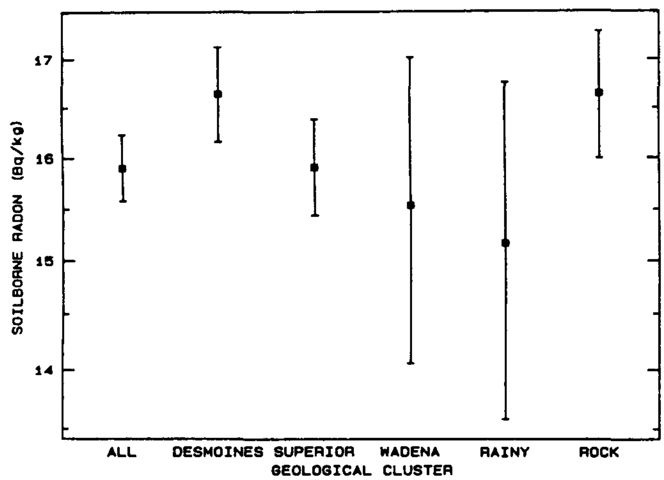

Fig. 12. Variation of the geometric means of emanative soilborne radon concentrations for houses clustered by major surface geological feature. Uncertainties in the means are shown as error bars. The first data point is the value for all houses. Clusters are named for lobes of the Wisconsin glaciation. Rock labels those houses located over bedrock within 10 m of the surface.

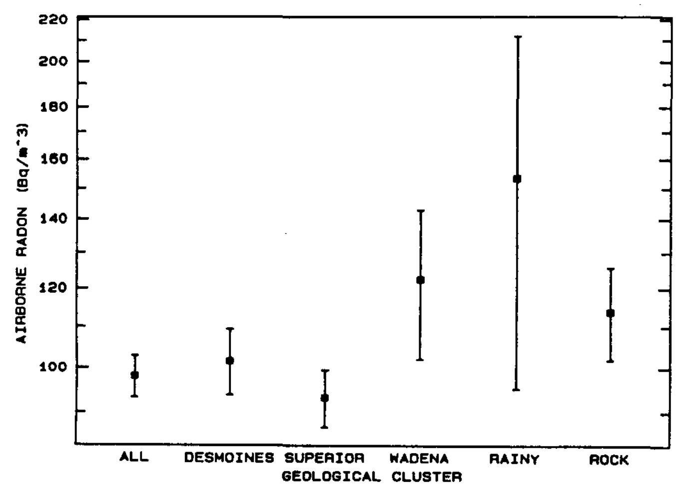

Fig. 13. Variation of the geometric means of indoor airborne radon concentrations for houses clustered by major surface geological feature. Uncertainties in the means are shown as error bars. The first data point is the value for all houses. Clusters are named for lobes of the Wisconsin glaciation. Rock labels those houses located over bedrock within 10 m of the surface.

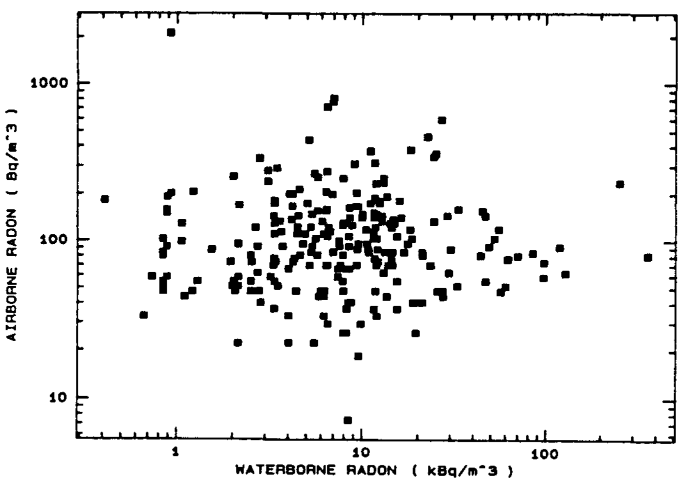

Fig. 14. Plot of indoor airborne radon concentrations in living spaces vs. waterborne radon concentrations in individual houses.

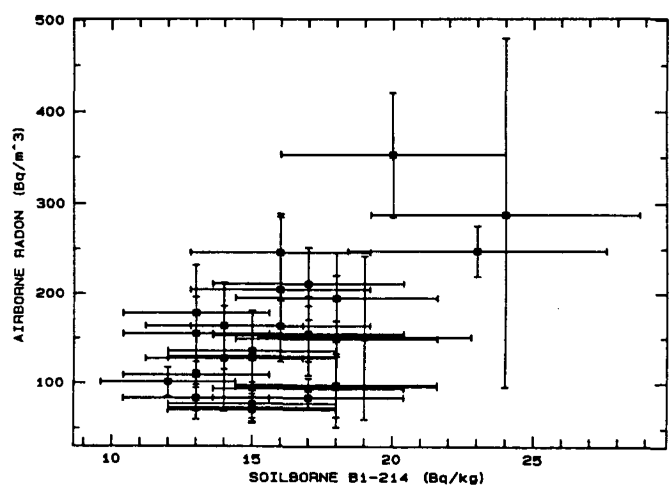

Fig. 15. Plot of indoor airborne radon concentrations in below-grade compartment vs. soilborne ^{214}Bi concentrations for town-sized clusters.

with elevated radon occur in both near-surface bedrock and glacial-till terrains. Some strong, highly localized anomalies may be associated with bedrock structures in conjunction with highly permeable soil. For example, the neighborhoods in Moose Lake with bedrock on the surface have significantly lower radon than a neighborhood on the south side of town where the bedrock is well covered by glacial outwash but fractured by a basalt intrusion. Thus, surface bedrock near a home is a poor indicator of indoor airborne radon in this study area.

Indoor Airborne Radon

Indoor airborne radon concentrations are log-normally distributed in homes along the southwestern edge of the Canadian Shield (see Fig. 3). The yearly average radon concentrations in living spaces have a geometric mean that is three times higher than the aggregate value of other U.S. regions (Nero et al. 1986) The geometric standard deviation of our distribution is smaller than that aggregate distribution (2.1 vs. 2.8). Thus, our arithmetic average is 2.5 times higher than the aggregate. Approximately 30% of the houses had yearly average radon concentrations in living spaces that exceeded the minimum U.S. E.P.A. action level, and 1% of the houses exceeded 740 Bq/m^3 (20 pCi/L).

The typical house in this study is a single-story, wooden, ranch-style home built on a full concrete-block basement. The basement is used as a living space. The house is well insulated, heated with fossil fuels and wood with forced air circulation. The typical house has a below-grade airborne radon concentration that is relatively constant throughout the year. The first floor concentration is 70% of the below-grade concentration during the heating season (October through May) and around 50% during the summer. Considerable variation (a factor of 10) of radon concentrations has been observed within a compartment of one of the houses that was studied extensively. Another home of different construction located nearby showed virtually uniform radon distribution throughout. Several houses that have been monitored for more than one year have shown yearly variations as great as a factor of two. However, the average yearly variation was closer to 30%.

Within the houses in a town, radon usually varied by less than a factor of two. All compartmental measures of indoor radon (living spaces, below-grade, above-grade) varied significantly from town to town. As Fig. 7 shows, the mean living-space radon concentration in a town varied from town to town by as much as a factor of four. However, there is little variation of indoor radon in either the regional or geological clusters used in this analysis (see Fig. 10 and 13). Some intermediate-sized geographic or geologic regions, such as the western Minnesota River valley, mid-continent rift valley, or the Iron Range, may have elevated indoor radon. A firm conclusion on intermediate-sized regions would require additional sampling.

CONCLUSIONS

The area along the southwestern edge of the Canadian shield contains waterborne and soilborne radon concentrations that are just slightly above the U.S. average and are log-normally distributed. The average indoor radon concentration in our living spaces is three times higher than the U.S. average. Indoor airborne radon concentrations are log-normally distributed in individual houses. Although indoor airborne radon is reasonably uniform in all regions contained within the study area, significant variation is present at a smaller geographic scale, including towns and neighborhoods.

Waterborne radon is not a significant source of airborne radon for the average home in the study area and shows no significant correlation with airborne radon. Significant correlations were observed between emanative soilborne radon, surface ^{214}Bi concentrations, and below-grade airborne radon, which makes these measurements useful indicators for locating

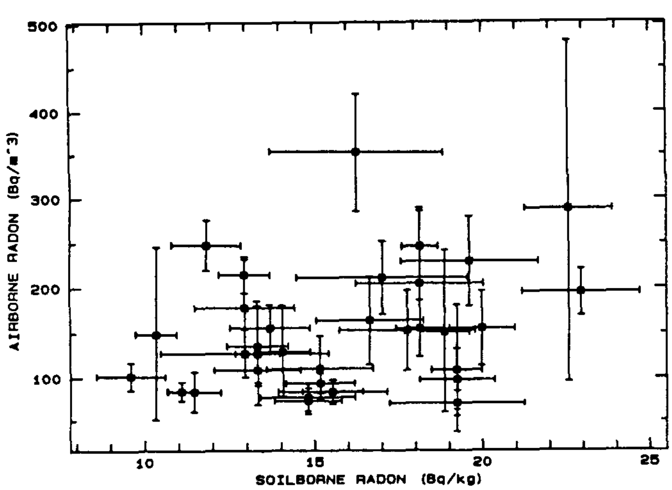

Fig. 16. Plot of indoor airborne radon concentrations in below-grade compartments vs. soilborne emanative radon concentrations for town-sized clusters.

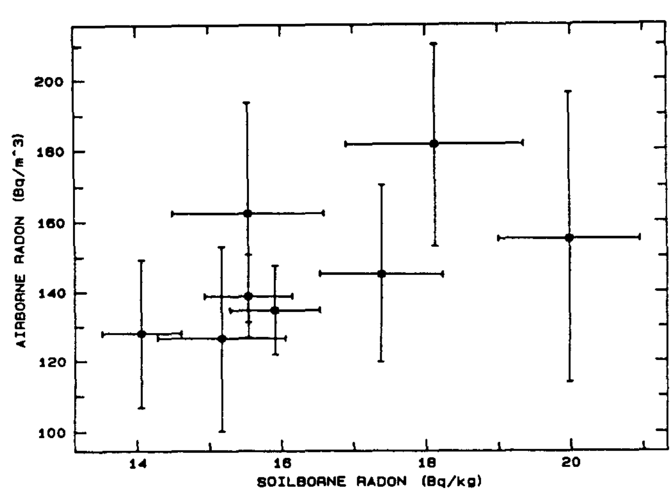

Fig. 17. Plot of indoor airborne radon concentrations in below-grade compartments vs. soilborne emanative radon concentrations for region-sized clusters.

areas of locally elevated indoor airborne radon. The relatively large variation of both soilborne source strengths and indoor airborne radon concentrations within clusters limits the quantitative predictive power of laboratory-based, single-source strength measurements. The failure of some characteristics (regional identification, geological classification, bedrock type, and waterborne radon) to predict indoor airborne radon concentration should be noted by those who use these characteristics as radon screening tools.

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