

RADON EMANATION FROM SOIL OF KENOSHA, RACINE AND WAUKESHA COUNTIES, SOUTHEASTERN WISCONSIN

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ABSTRACT

The radon (²²²Rn) emanation from soil of Kenosha, Racine and Waukesha Counties was measured and related to the soil chemistry, inherent radioactivity and grain-size distribution. Our twelve study sites were in soil of the Horicon, New Berlin and Oak Creek Formations, lake plain sediment and where bedrock is close to the surface. Canisters containing activated charcoal were placed into holes bored into the soil. The hole openings were covered, and field radon emanations were collected for a period of over 50 hours.

Radon emanation concentration at all sites other than the shallow bedrock site was above 4 pCi/l; at three locations radon concentration exceeded 100 pCi/l. Sites where subsoil had high clay or silt plus clay content tended to have lower radon emanation concentration. Soil permeability may also influence whether relatively high inherent soil radioactivity results in high radon emanation concentration.

We found no significant relationship between radon emanation concentration and pebble lithology, atmospheric pressure, or uranium (²³⁸U) concentration determined by airborne gamma ray and magnetic surveys. Water-soluble phosphate concentration of surface soil were low; therefore, the natural radon concentration was probably not augmented by uranium-bearing phosphate fertilizer additions.

In an ancillary study, we found that radon emanation concentration was highest at the ground surface and decreased to nearly zero at a depth of 6.9 m. The higher radon concentration at the surface may be due to more conduits for soil gas movement present in the weathered zone than at depth.

INTRODUCTION

Radon (²²²Rn) is a chemically inert gas produced from the decay of radium (²²⁶Ra), and has a half-life of 3.8 days. Its decay produces a sequence of 4 short-lived progeny followed by lead (²¹⁰Pb) whose half-life is 19.4 years. The short-lived progeny attach to dust and aerosols and lodge in the lung tissue when inhaled. The decay of polonium (²¹⁸Po and ²¹⁴Po) gives off alpha particles which are the main source of radiation hazard when they decay in the lungs (Robkin, 1987).

Airborne radon concentration is usually expressed in picocuries per liter, pCi/l, a measure of the number of radioactive disintegrations per minute in a liter of air. The pCi/l measurements can be converted to S.I. units (Bq/m³) by multiplying by 37.18.

The ultimate source of radon is uranium in rock and soil. Typical uranium concentration in soil and rock is a few ppm (Bodansky, 1987). The crustal mean concentration of uranium is about 3 ppm;

granite and shale average about 5 and 3.5 ppm uranium, respectively; phosphate rock contains 50-100 ppm uranium (NCRP, 1976; Krauskopf, 1979). As radon atoms are liberated from geologic materials they become part of the soil gas. Soil gas can be drawn into basements of buildings via cracks in walls and floors in response to air pressure differences.

Soil gas movement depends on rock/soil permeability (Tanner, 1986). There is greater soil gas movement, and therefore a potential for high radon emanation rates when soil water content is low (Nevissi and Bodansky, 1987). Radon solubility in water ranges from 51.0 cm³ of radon gas (at 0°C, 1 atmosphere) per 100 cm³ of water at 0°C to 13.0 cm³ radon per 100 cm³ of water at 60°C (Chemical Rubber Company, 1988).

Our study was conducted following a survey of indoor airborne radon in Wisconsin homes (Wisconsin Department of Health and Social Services, 1987). According to the survey, an estimated 44.3% of

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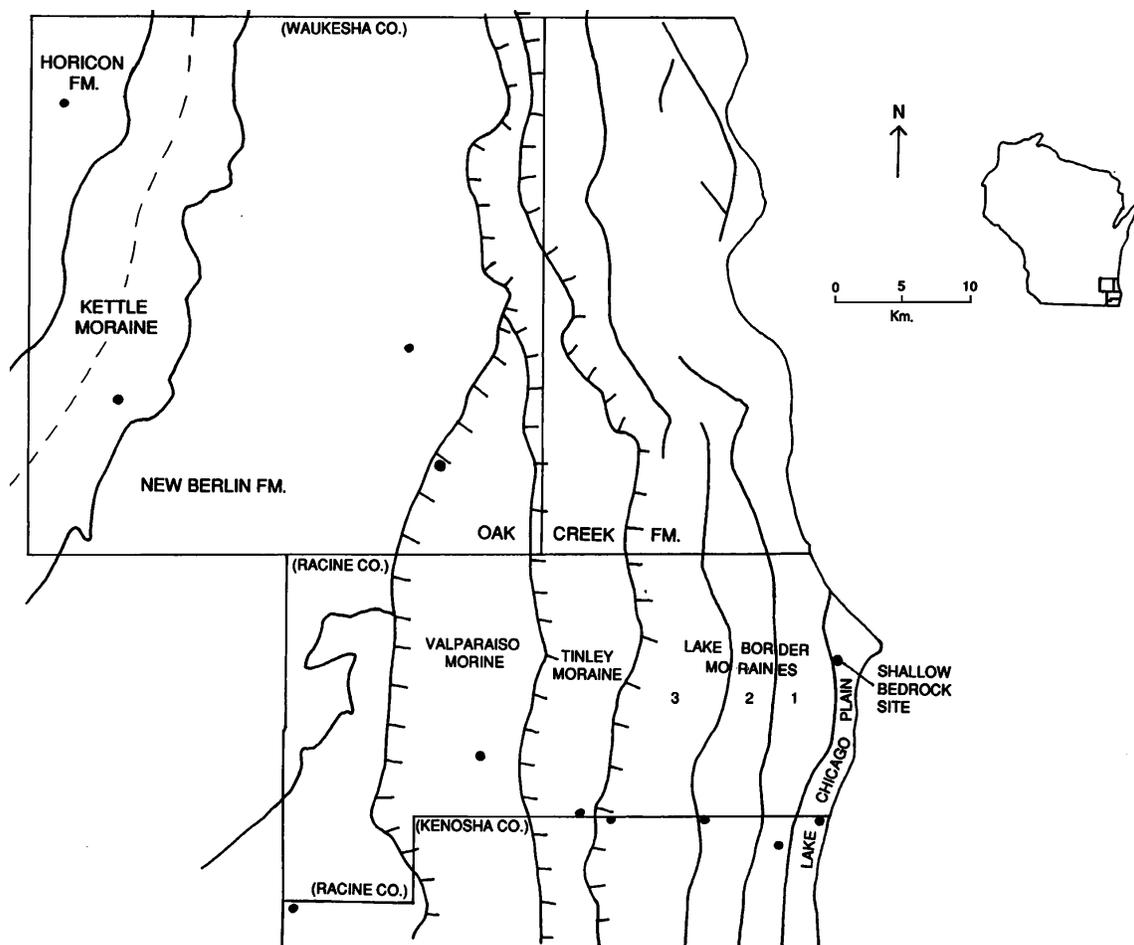


Figure 1. Map of sites in the study area in southeastern Wisconsin. Geology after Schneider (1983).

homes in an area of southeastern Wisconsin (including Kenosha, Racine and Waukesha Counties) have indoor radon concentration greater than 4 pCi/l, the Environmental Protection Agency's recommended limit for indoor air. The purpose of our research was to measure the radon emanation from soil and relate it to soil chemistry, inherent radioactivity and grain-size distribution. Our ancillary study was conducted to determine if the weathered zone of an unconsolidated formation contains higher radon emanation concentration than the unweathered zone.

METHODS

Ten of our twelve sites were located in the moraines of the Oak Creek, Horicon and New Berlin Formations (fig. 1). The moraines vary in thickness from 0 to 125 m and overlie Silurian dolostone bedrock (Schneider, 1983). The glacial material is weathered

to a depth of 3 to 4 m. Two more sites were near Lake Michigan in lake plain sediment and where dolostone bedrock is close to the surface. At the shallow bedrock site, the surficial deposit is lacustrine clay. As we were concerned that radon from uranium-bearing phosphate fertilizer additions could augment the natural radon concentration, wooded sites with medium to large trees were selected to ensure the site had not been recently farmed, and therefore not fertilized.

Twenty-cm-diameter holes were dug with a power auger to depths of 27 to 79 cm; most holes were 33 to 42 cm deep (table 1). The holes extended to the upper part of the B soil horizon. Soil samples were collected from the surface and from the bottom of each hole (herein referred to as "surface soil" and "subsoil"). To determine variability of the radon concentration over short distances, two adjacent holes, 4 to 7 m apart, were installed at each of the 8 sites in Racine and Kenosha Counties.

Ten-cm-diameter canisters containing activated charcoal with diffusion barrier in desiccant were used to measure the radon concentration. Prior to being placed in the hole, filter paper was taped over the top of each canister to keep soil out. The canisters were placed on small wooden boards which were inserted into the side of each hole. This allowed for air circulation around the canister and for drainage of any water that might enter the hole. The canisters were 15 - 37 cm below the ground surface (table 1). A plastic sheet, a wooden board, and soil were placed over the hole so that equilibrium between the soils and air in the hole would be established. The canisters were exposed to soil gas for over 50 hours, retrieved, sealed and taken to the Wisconsin Department of Health and Social Services, Section of Radiation Protection in Madison where the radon concentration was determined using gamma ray spectroscopy. Average atmospheric pressure during the time each can was exposed was determined using hourly weather records from the National

Weather Service Forecast Office in Milwaukee (table1).

The water-soluble phosphate content of surface soil at each site was determined by the Truog and Meyer method (Chapman and Pratt, 1961) and the ascorbic acid reduction method was used to determine phosphate concentration in each extract. Particle-size distribution of the subsoil was determined using screens (pebble- and sand-size fractions) and settling tubes (silt- and clay-size fractions). The particle-size divisions were: >2 mm = pebbles; 2 mm to 0.063 mm = sand; 0.063 mm to 0.004 mm = silt; and <0.004 mm = clay. The lithology of the pebbles and constituents of the sand were determined by microscopic examination.

The inherent radioactivity of subsoil from 6 holes was determined using the method described in Edgington and Lucas (1970). In summary, 160 g soil samples were analyzed for counting periods of 21 to 70 hours using a sodium iodide crystal to detect

Table 1. Hole and canister depth, duration of canister exposure, and average atmospheric pressure for each site.

Site	Canister number	Hole depth cm	Canister depth cm	Canister open		Canister closed		Exposure hr	Pressure mb
				Date	Time	Date	Time		
HORICON FORMATION									
Waukesha Co.	061	56	28	11/5/88	11:41 AM	11/7/88	2:22 PM	50.68	969.4
NEW BERLIN FORMATION									
Kettle Moraine, Waukesha Co.	130	36	18	"	10:55 AM	"	1:32 PM	50.62	969.3
Ground Moraine, Waukesha Co.	149	38	23	"	10:09 AM	"	12:56 PM	50.78	968.9
Pitted Outwash, Kenosha Co.	147	41	20	11/4/88	5:14 PM	"	12:37 PM	67.38	967.3
	131	79	23	"	5:10 PM	"	12:31 PM	67.35	967.3
OAK CREEK FORMATION									
Valparaiso Moraine									
Racine Co.	095	27	15	"	4:15 PM	"	11:45 AM	67.50	967.3
	089	29	17	"	4:22 PM	"	11:49 AM	67.45	967.3
Waukesha Co.	157	33	20	11/5/88	9:33 AM	"	12:21 PM	50.80	968.6
Tinley Moraine, Racine Co.	104	36	20	11/4/88	3:40 PM	"	11:13 AM	67.55	966.7
	055	27	15	"	3:44 PM	"	11:19 AM	67.58	966.7
Lake Border Moraines, Kenosha Co.									
3	009	64	37	"	3:09 PM	"	11:00 AM	67.85	967.0
	046	42	30	"	3:17 PM	"	10:53 AM	67.60	967.0
2	150	33	22	"	2:43 PM	"	10:30 AM	67.78	967.0
	033	36	20	"	2:48 PM	"	10:35 AM	67.78	967.0
1	004	33	23	"	2:13 PM	"	10:07 AM	67.90	966.7
	036	41	28	"	2:20 PM	"	10:00 AM	67.67	966.7
LAKE PLAIN SEDIMENTS,									
Racine Co.	052	71	38	"	1:54 PM	"	9:45 AM	67.85	966.7
	080	41	30	"	1:45 PM	"	9:37 AM	67.87	966.7
SHALLOW BEDROCK SITE,									
Racine Co.	038	53	34	"	1:00 PM	"	9:00 AM	68.00	966.5
	032	58	33	"	1:05 PM	"	9:06 AM	68.02	966.5

gamma ray emissions, and identify their nuclide source based on their energy.

We compared the radon emanation concentration measured in our study to airborne gamma ray and magnetic surveys of southeastern Wisconsin (Geometrics, 1981, and High Life Helicopters, Inc./QEB, Inc., 1981). The gamma ray and magnetic surveys measured bismuth (^{214}Bi) concentration in the upper 46 cm of the ground and converted the measurements to equivalent ^{238}U . The ^{238}U distribution was determined from "pseudocontour maps." County boundaries and our study sites were located using latitude and longitude.

To determine whether radon emanation concentration changes with depth, an ancillary study was conducted. The study site was an 8-month old, approximately 9 m deep excavation in the Oak Creek Formation, lake border moraine 1 in Racine County. Steel drive-points were hammered 40 cm deep into undisturbed surface soil, the top of the unweathered till and to depths of 2.3, 4.6 and 6.9 m (vertical distance) below the top. The drive-points were open at the top and had a few small holes along the bottom 3 cm. A portable electric pump drew the standing air in the pipe and an equal volume of soil gas through a gas filter and into EDA Instruments, Inc. (Toronto, Canada) scintillation cells. Pumping time was 5 sec. Two samples were collected from the undisturbed surface soil and from depths of 2.3 and 6.9 m; one sample was collected from a depth of 4.6 m. The gas samples equilibrated for 2-3 hours before being placed into Radon Detector RDA-200 devices, manufactured by EDA Instruments, Inc. The scintillation cells have a ZnS phosphor coating which emits ultraviolet light when hit by alpha particles originating from radon and its decay products. The light flashes, detected by the RDA-200 photo-multiplier tube, are converted to electrical impulses and the accumulation of impulses is digitally displayed. We used a counting period of 30 minutes. The concentration of radon was calculated using an algorithm provided by the RDA operations manual.

RESULTS AND DISCUSSION

Radon emanation concentration was from 3.08 to 188.15 pCi/l (table 2). The highest concentration was in the Tinley Moraine, the lowest were at the shallow bedrock site.

To determine whether radon emanation concentration varies over short distances, we examined the results from the eight sites where two adjacent holes were installed. At two sites (Tinley and Valparaiso

Moraine in Racine County) the experiment set-up remained intact and no canisters were contaminated with soil or diluted with surface air. Adjacent holes differed by 12 and 21% in the Tinley and Valparaiso sites, respectively. Based on these results, it appears that there are only small variations in radon concentration over short distances.

However, at six other sites where two adjacent holes were installed, air dilution and/or soil contamination occurred at one of the two holes. The surface cover collapsed inward at four of the six sites (lake plain sediments, lake border moraines 1, 2 and 3 of the Oak Creek Formation). The radon emanation concentration at these sites varied 73 to 78% between the two adjacent holes. In each pair, the lower radon emanation concentration corresponds to the hole with the collapsed cover; the lower value may be a result of surface air diluting the radon emanations.

The board holding the canister became slanted or fell at two sites (New Berlin Formation pitted outwash and shallow bedrock site) and as a result, soil got into the canisters. In both cases, the radon concentration of the contaminated canister was higher than the uncontaminated ones.

Thus, out of 20 canisters, the radon emanation concentration from six canisters is not considered valid. Only radon concentration from the 14 canisters which remained uncontaminated and in place is considered accurate and have been included in the tables.

Sand was the dominant constituent of the subsoil at seven sites, the highest amount was 97.1% at the New Berlin Formation pitted outwash site (table 2, figure 1). Silt was dominant at four sites. At all but one site (lake plain sediments), silt was either the highest or second highest percentage component. Clay was the major constituent at one site (Valparaiso Moraine in Waukesha County).

Based on information from Tanner (1986), we expected high radon concentration where the soil is coarse and well-drained, and therefore permeable to soil gas containing radon. However, in our study, the relationship between the sand content of the subsoil and radon emanation concentration is inconsistent. For example, at the Horicon Formation site, subsoil contains 71.4% sand and radon concentration is 50.40 pCi/l. However, at the Valparaiso Moraine in Racine County where subsoil contains about the same amount of sand (70.6%), radon emanation concentration is much lower (13.77 and 16.69 pCi/l). We noted that the highest radon emanation concentration in our study (167.44 and 188.15 pCi/l at the Tinley Moraine) were at locations where silt is the dominant constituent of the subsoil.

At locations where subsoil had high clay content, radon emanation concentration tended to be low. The three lake border moraines illustrate this relationship. As clay content increases from lake border moraine 3 to 1, radon emanation concentration decreases. At the shallow bedrock site, where the surficial deposits are lacustrine clays, the radon emanation concentration was the lowest of all 12 sites, 3.08 pCi/l. At this site, the soil was saturated when we did our field work. Therefore, the qualitative inverse relationship between clay content and radon emanation concentration may relate to the fact that clay, particularly if wet, inhibits radon movement (Tanner, 1986).

The two sites in the Valparaiso Moraine are about 25 km apart (fig. 1), yet radon emanation concentration at the two sites (three holes) is similar (13.77 and 16.69 pCi/l in Racine County, 11.43 pCi/l in Waukesha County). This is surprising considering that there were differences in grain-size distribution between the two sites. In Racine County the Valparaiso Moraine subsoil contains 70.6% sand, however, in Waukesha

County it is silty and clayey (89.9% silt plus clay).

To determine how radon emanation concentration, inherent soil radioactivity, and subsoil permeability are related, we looked at the data for the six sites where inherent soil radioactivity was measured (table 3). Inherent soil radioactivity measurements are expressed as counts per 1000 seconds of radium and bismuth (^{226}Ra , ^{214}Bi). The $^{226}\text{Ra}/^{214}\text{Bi}$ ratios are very similar for the six sites tested (table 3). The constancy of the ratios may be a basis for inferring that radon production is proportional to the ^{226}Ra and/or ^{214}Bi counts. If we assume radon production is approximately proportional to either its parent nuclide, ^{226}Ra , or one of its daughter nuclides, ^{214}Bi , then (of the six sites tested) the highest radon production is at the Valparaiso moraine in Waukesha County, and the lowest is at the Horicon Formation site.

At the Valparaiso Moraine in Waukesha County, inherent soil radioactivity is relatively high and radon emanation concentration is relatively low. The subsoil at this site contains 89.9% silt and clay. The low per-

Table 2. Radon concentration, water-soluble phosphate concentration and subsoil grain-size distribution for each site.

Site	Canister number	Radon pCi/l	Water-soluble phosphate ppm	Subsoil grain-size			
				>sand (%)	sand (%)	silt (%)	clay (%)
HORICON FORMATION, Waukesha Co.	061	50.40 ± .76	—	3.6	71.4	17.1	7.9
NEW BERLIN FORMATION							
Kettle Moraine, Waukesha Co.	130	123.61 ± 1.11	—	6.9	56.4	24.1	12.6
Ground Moraine, Waukesha Co.	149	47.78 ± .76	2.65	5.8	39.8	38.5	15.9
Pitted Outwash, Kenosha Co.	147	17.48 ± .58	0.25	.1	97.1	2.4	.4
	131	41.80 ± .92**					
OAK CREEK FORMATION							
Valparaiso Moraine							
Racine Co.	095	13.77 ± .54	0.25	.1	70.6	24.5	4.8
	089	16.69 ± .55					
Waukesha Co.	157	11.43 ± .46	1.25	1.4	8.7	41.2	48.7
Tinley Moraine, Racine Co.	104	188.15 ± 1.51	0.25	.9	28.6	55.1	15.4
	055	167.44 ± 1.34					
Lake Border Moraines, Kenosha Co.							
3	009	54.98 ± .88	2.0	1.9	42.7	39.8	15.6
	046	14.53 ± .62*					
2	150	35.03 ± .70	0.50	.2	35.5	45.0	19.3
	033	8.01 ± .48*					
1	004	25.54 ± .66	4.50	2.2	32.9	40.0	24.9
	036	16.53 ± .58*					
LAKE PLAIN SEDIMENTS, Racine Co.	052	116.09 ± 1.16	2.85	5.6	87.2	4.1	3.1
	080	24.67 ± 3.58*					
SHALLOW BEDROCK SITE, Racine Co.	038	3.08 ± .45	0.75	.6	19.2	53.6	26.6
	032	3.26 ± .47**					

*Hole cover collapsed

**Soil in canister

Table 3. *Inherent soil radioactivity, radon concentration and grain-size distribution for six sites. Background counting lasted 91.8 hr.*

Site	Count time hours	²²⁶ Ra* Counts per 1000 sec minus background	²¹⁴ Bi** background	²²⁶ Ra/ ²¹⁴ Bi	Radon pCi/l	Grain-size distribution	
						>sand+ sand (%)	Combined silt+ clay (%)
HORICON FORMATION, Waukesha Co.	69.5	2.62	5.28	0.50	50.40 ± .76	75.0	25.0
NEW BERLIN FORMATION Kettle Moraine, Waukesha Co.	26.5	4.35	10.34	0.42	123.61 ± 1.11	63.3	36.7
OAK CREEK FORMATION Valparaiso Moraine Racine, Co.	50.4	3.17	7.73	0.41	13.77 ± .54 16.69 ± .55	70.7	29.3
Waukesha Co.	27.1	10.90	26.36	0.41	11.43 ± .46	10.1	89.9
Lake Border Moraine, Kenosha Co. 2	24.5	10.33	19.86	0.52	35.03 ± .70	35.7	64.3
1	20.6	8.81	20.29	0.43	25.54 ± .66	35.1	64.9

*180-192 Mev channel

**604-616 Mev channel

meability soil may have restricted the movement of soil gas containing radon, resulting in a relatively low radon emanation concentration. A different relationship occurs at the Kettle Moraine and Horicon Formation sites, where inherent soil radioactivity is relatively low but radon emanation concentration is relatively high. There, the subsoil is more permeable with 75.0% and 63.3% pebble- and sand-size particles. The radon emanation concentration we measured may originate from sediment or bedrock deeper than the hole we installed. The high soil permeability may cause radon gas movement from depth to the surface.

For some of the six sites, the data were inconsistent. For example, we noted that the inherent soil radioactivity and subsoil grain-size distribution of the Valparaiso Moraine Racine County and Horicon Formation sites are about the same. Yet the radon emanation concentration is more than three times higher at the Horicon Formation site than at the Valparaiso Moraine Racine County site.

Based on our comparison of radon emanation concentration, inherent soil radioactivity, and subsoil permeability data for the six sites where inherent soil radioactivity was measured, we believe there is a relationship between the three factors. However, the relationship could be obscured at some locations by factors not included in this study.

Water-soluble phosphate content of the surface soil was generally low, less than 5 ppm (table 2). A few ppm of water-soluble phosphate are considered background concentration in soil (Chapman and Pratt, 1961). Therefore, it is unlikely that the land has been heavily fertilized with rock phosphate. Phosphate is probably not a significant source of radon in our study

unless there are high amounts of insoluble phosphate compounds present.

The dominant pebble (>2 mm) lithologies in the subsoil were quartz, granite/gneiss and carbonate (table 4). The sand was >90% quartz at all sites. Cobbles encountered during hole construction were mostly carbonate. Because granite and shale are more likely to contain higher concentration of uranium than other rock (Krauskopf, 1979), we expected higher radon emanation concentration at sites where they are the dominant pebble lithology. We used Pearson correlation coefficients to determine if there is a statistical relationship between radon concentration and lithology, and found that there were no significant linear correlations between radon concentration and percent of each lithology.

Average atmospheric pressure while the canisters were exposed only varied 2.98 mb among the 14 valid data points. Atmospheric pressure can influence soil gas movement. Therefore, Pearson correlation coefficients were used to determine if there is a relationship between average atmospheric pressure and radon concentration. The r^2 value is virtually zero ($r^2 = 2.4 \times 10^{-7}$); therefore there is no relationship.

Figure 2 is a map of ²³⁸U concentration as determined by airborne gamma ray and magnetic survey. Statistical uranium anomalies, marked on the map with short dashes, can result from a number of situations, such as the true concentration of uraniumiferous minerals, differential soil and vegetation cover, and others (Geometrics, 1981). However, the pattern of uranium anomalies in southeastern Wisconsin could also be related to urban and industrial development. Pearson correlation coefficients were used to deter-

Table 4. Subsoil pebble lithology distribution for each site. The value "n" is the number of pebbles examined.

Site	n	Pebble lithology (%)				
		Quartz	Granite/gneiss	Mafic	Carbonate	Shale
HORICON FORMATION, Waukesha Co.	40	60	28	—	10	2
NEW BERLIN FORMATION						
Kettle Moraine, Waukesha Co.	16	38	31	—	31	—
Ground Moraine, Waukesha Co.	61	13	44	23	20	—
Pitted Outwash, Kenosha Co.	1	—	—	—	100	—
OAK CREEK FORMATION						
Valparaiso Moraine						
Racine Co.	10	10	70	—	10	10
Waukesha Co.	3	33	67	—	—	—
Tinley Moraine, Racine Co.	21	29	14	—	57	—
Lake Border Moraines, Kenosha Co.						
3	30	3	17	3	77	—
2	3	—	—	—	100	—
1	16	44	37	—	19	—
LAKE PLAIN SEDIMENTS, Racine Co.	23	4	18	—	78	—
SHALLOW BEDROCK SITE, Racine Co.	12	—	—	8	84	8

mine if there was a relationship between the radon emanation concentration at our sites and the ^{238}U concentration. The r^2 value is 0.002; therefore there is no relationship. We noted that the higher uranium concentration roughly match the boundaries of the Oak Creek Formation. The inherent soil radioactivity measurements generally correspond to the airborne-measured uranium concentration when they are compared qualitatively.

In our ancillary study we looked at changes in radon emanation concentration with depth in Oak Creek Formation till. The radon emanation concentration was highest in the surface soil (39.3 pCi/l) (a second value of 0.7 was discounted as air contamination). At the top of unweathered till, the radon emanation concentration was 17.2 pCi/l, and radon concentration progressively decreased with depth. At a depth of 2.3 m, radon emanation concentration was 13.2 and 5.0 pCi/l; at a depth of 4.6 m, 0.7 pCi/l; and at a depth of 6.9 m, 0.2 and 0 pCi/l. The higher radon emanation concentration at the surface than at a depth may be due to openings in the weathered surface zone. Soil gas containing radon is likely to flow more freely there than in the relatively dense, unweathered till.

CONCLUSIONS

Radon emanation concentration at the locations we studied appears to be primarily related to subsoil permeability. Based on our work, we believe that high silt and clay content in the subsoil restricts soil gas (containing radon) movement. Even locations with high inherent soil radioactivity may not have high radon

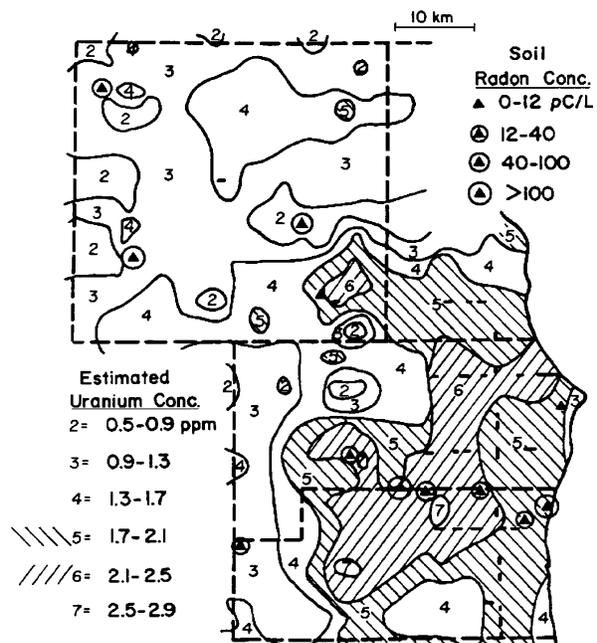


Figure 2. Airborne-measured uranium (^{238}U) concentration in the upper 46 cm of the ground (Geometrics, 1981; High Life Helicopters, Inc./QEB, Inc., 1981), and radon concentration at each site for Kenosha, Racine, and Waukesha Counties.

emanation concentration due to low soil permeability. Likewise, subsoil with high sand content would permit radon to move from depth to the surface; this could result in relatively high radon emanation con-

centration even where inherent soil radioactivity is relatively low. At some of our sites the relationship between radon emanation concentration and soil permeability is more apparent than at others. We recognize that many factors influence radon emanation concentration and that it may be not possible to isolate the relationship between radon emanation concentration and any one, or combination of, factors.

We found no significant relationship between the radon emanation concentration and subsoil pebble lithology, atmospheric pressure when the canister was exposed, and ^{238}U concentration of the soil as determined by airborne gamma ray and magnetic survey. In our ancillary study we found that radon emanation concentration decrease with depth. This may be a result of more pathways for soil gas movement in the weathered surface zone than in unweathered till at depth.

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