The Determination of Radon Activities in Ground Water from Wisconsin Tills in Southwestern Ohio and Southeastern Indiana[1]

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ABSTRACT. **Two hypotheses have been suggested by previous workers to explain the relatively elevated radon activities of ground water in certain areas of southwestern Ohio and southeastern Indiana. First, radon may be produced close to or at the Ordovician-Silurian unconformity by the concentration of uranium and radium on iron and manganese oxides and hydroxides and on clay minerals at this zone of weathering. Second, radon may be formed from the radioactive decay of radium which is concentrated on iron-oxides in zones of higher hydraulic conductivity in the lower carbonate section of the Silurian System. In both cases, it has been proposed that the elevated radon activities result from either the application of radium-bearing phosphate fertilizers or from the inclusion of radium-bearing fragments of Devonian black shale in the till.**

The present study attempted to determine if ground water from different Wisconsin till units in southwestern Ohio contain significantly distinct radon activities due to the differing amounts of Devonian black shale contained in the drift. Duplicate samples were collected from 47 private wells and springs that produce water from different glacial units. The samples were analyzed for their radon activity using liquid scintillation methodology.

Data indicate that it is not possible to differentiate statistically between radon activities in water from wells that penetrate the Upper and Lower Shelbyville or the Crawfordsville tills. Instead, those wells and springs with high radon activities appear to be associated with production of water from horizons of unusually high hydraulic conductivity. Wells that produce water from glacial drift close to areas where carbonate rocks of the lower Silurian System subcrop also have a higher radon signature. These higher radon activities are believed to be associated with flow of ground water up into the drift from zones of high hydraulic conductivity in the lower Silurian System.

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INTRODUCTION

Radon-222 (hereafter radon) is a colorless, odorless, radioactive gas that is a daughter element in the decay chain of uranium-238. The immediate parent of radon is radium-226. Radon occurs naturally in elevated concentrations in the air of some homes and beneath the Earth's surface in soil gas and ground water. The alpha radiation emitted by radon and its polonium progeny is considered a significant health hazard by the United States Environmental Protection Agency (USEPA) because at elevated levels it causes lung cancer.

Radon activities in the air of at least 40% of the homes in the Dayton, OH, area frequently exceed the four picocuries per liter environmental-action level set by the USEPA (Paul and Lindstrum 1987). Few studies, however, have sought to define the geologic source of this radon.

Possible Origin of Radon in Southwestern Ohio

Harrell et al. (1993) postulated that the source of indoor radon in Ohio may be the till that blankets the upland regions in the glaciated regions of southwestern Ohio. They theorized that the Wisconsin glaciers eroded and transported large quantities of uraniferous Devonian black shale to the southwest as it flowed across outcrops of the easily eroded black shale near Bellefontaine in west-central Ohio. These black shales have an arithmetic mean of 26.7 ppm uranium (Harrell and Kumar 1988).

Harrell et al. (1993) further suggested that the extensive weathering of the carbonate clasts that comprise a high percentage of the till may be another source of the elevated indoor radon activities. The weathering of these limestone clasts leaves behind a concentration of insoluble residues (clays, iron oxides, and hydroxides) in the resulting soils. These weathering residuals, with their capacity to adsorb uranium and radium, could be a source of the elevated radon activities recorded in many homes within the glaciated region.

The importance of glacial drift as a generator of radon was further suggested in work by Duval (1982). His aerial radiometric contour map, showing variations in uranium concentrations in soils in Ohio, mimics closely the lobate nature of the recessional moraines that cover the glaciated portion of the state.

In an attempt to define more precisely the geologic source of elevated indoor radon activities in southwestern Ohio, Baldwin and Treick (1991) collected ground water from 157 private wells for radon analysis. Their work suggested that there is a distinct zone of elevated radon production in the lower Brassfield Formation, a carbonate unit that lies immediately above the Ordovician-Silurian contact. Gall et al. (1995), however, found that elevated radon activities occurred in ground water not only in the lower Brassfield Formation but also in the Dayton Dolomite, a carbonate unit approximately 18 m (59 ft) higher in the Silurian section in the

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Dayton area. They found that the zones of highest radon activities in ground water are also horizons of highest hydraulic conductivity and uranium concentrations in the bedrock. Thus they postulated that the flow of large volumes of water containing low concentrations of uranium and radium has resulted in the adsorption of these radionuclides on iron and manganese oxides and clays in these horizons.

MATERIALS AND METHODS

The study area lies in southwestern Ohio and southeastern Indiana (Fig. 1). Water was collected from 41 private wells and six springs in an attempt to determine if different till sheets were the source of elevated radon values in ground water in this region. Where possible, water was collected from wells which penetrate different till units deposited by ice which entered southwestern Ohio from either the northeast or northwest. It was postulated that the sediment deposited from ice flowing from the northeast would have a higher percentage of black shale fragments and thus produce more radon because it would have crossed the uranium and radium-rich, Devonian, black-shale outlier near Bellefontaine. No nearby source of Devonian shale inclusions exists for tills deposited by ice that flowed into southwestern Ohio from the northwest. Well logs were utilized where available to determine subsurface conditions and water-producing horizons. Water was collected from springs wherever they were found.

Geology of Study Area

Thin limestone and shale units possessing low hydraulic conductivity and belonging to the Cincinnatian Series of the Ordovician System underlie much of the study area (Fig. 2). Wells cased to bedrock seldom yield more than 19 l per minute (5 gals per minute). Where usable quantities of water are produced, the water is derived from bedrock fractures or from a weathered zone at the top of the Ordovician section. Limited outcrops of the Brassfield Formation, a lower Silurian carbonate unit, overlie unconformably the upper Ordovician rocks along the northern edge of the study region.

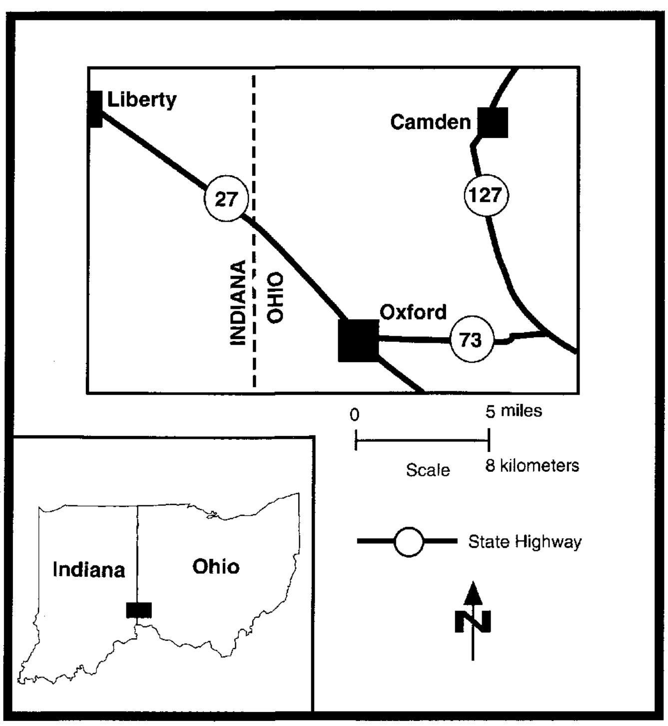

FIGURE 1. Location of study area.

Overlying the Ordovician-Silurian bedrock is non-indurated Pleistocene drift. As reported by Stewart and Miller (1987), the drift is predominantly of Wisconsin age (Fig. 3). The Fayette Till, the Connersville sands and gravel, the Shelbyville Tills, the sands and gravel deposited during the Shelbyville Interphase and the Crawfordsville Till lie unconformably on top of the Ordovician and Silurian section under much of the study area. The Fayette Till is a compact, dark gray, pebbly to sandy clay material. Hydraulic conductivity tests on the Fayette Till yielded a mean value of 4.5×10^{-8} cm/s. This low value limits the downward movement of ground water. Till fabric analysis suggests that the unit was deposited by ice which flowed into the area from the northeast.

The overlying Connersville sand and gravel deposit is a discontinuous, poorly sorted sand, silt, and gravel unit. Apart from the valley-train deposits of sand and gravel that fill the larger valleys in the area, the Connersville is the most important water-producing unit in the study area. It has a mean hydraulic conductivity of 4.9×10^{-2} cm/s.

The Shelbyville Till lies above the Connersville sand and gravel and is divided into two subunits (Goldthwait et al. 1981). The Lower Shelbyville is a blue-gray to oxidized brown, poorly sorted clayey to silty sand with a moderate hydraulic conductivity of 1.5×10^{-3} cm/sec. This unit thus allows ground water to move vertically through it until the water reaches an underlying relatively impermeable layer such as the Fayette Till or bedrock. The till has a northwest till fabric because it was deposited on the east flank of the White River Ice Lobe (Goldthwait et al. 1981). The Lower Shelbyville crops out as the surface till in the western portion of the study area (Fig. 4) and most likely lies beneath the Upper Shelbyville in the eastern and northeastern sections of the study area.

The Upper Shelbyville Till was deposited at the western edge of the Miami Lobe (Goldthwait et al. 1991). It has a northeast fabric and thus potentially a higher uranium/radium radon concentration. The Fayette Till also has a northeast till fabric; however, it is discontinuous through the study area and, as mentioned previously, is relatively impermeable. The Upper Shelbyville is a medium brown, loosely compacted, and sandy textured till (Goldthwait et al. 1981).

The Camden Moraine and the Crawfordsville Till lie above the Upper Shelbyville Till in the northeastern portion of the study area. During Crawfordsville time, the ice advanced from the northeast to a terminal position marked by the Camden Moraine. The till deposited during this phase is yellow brown to gray brown and

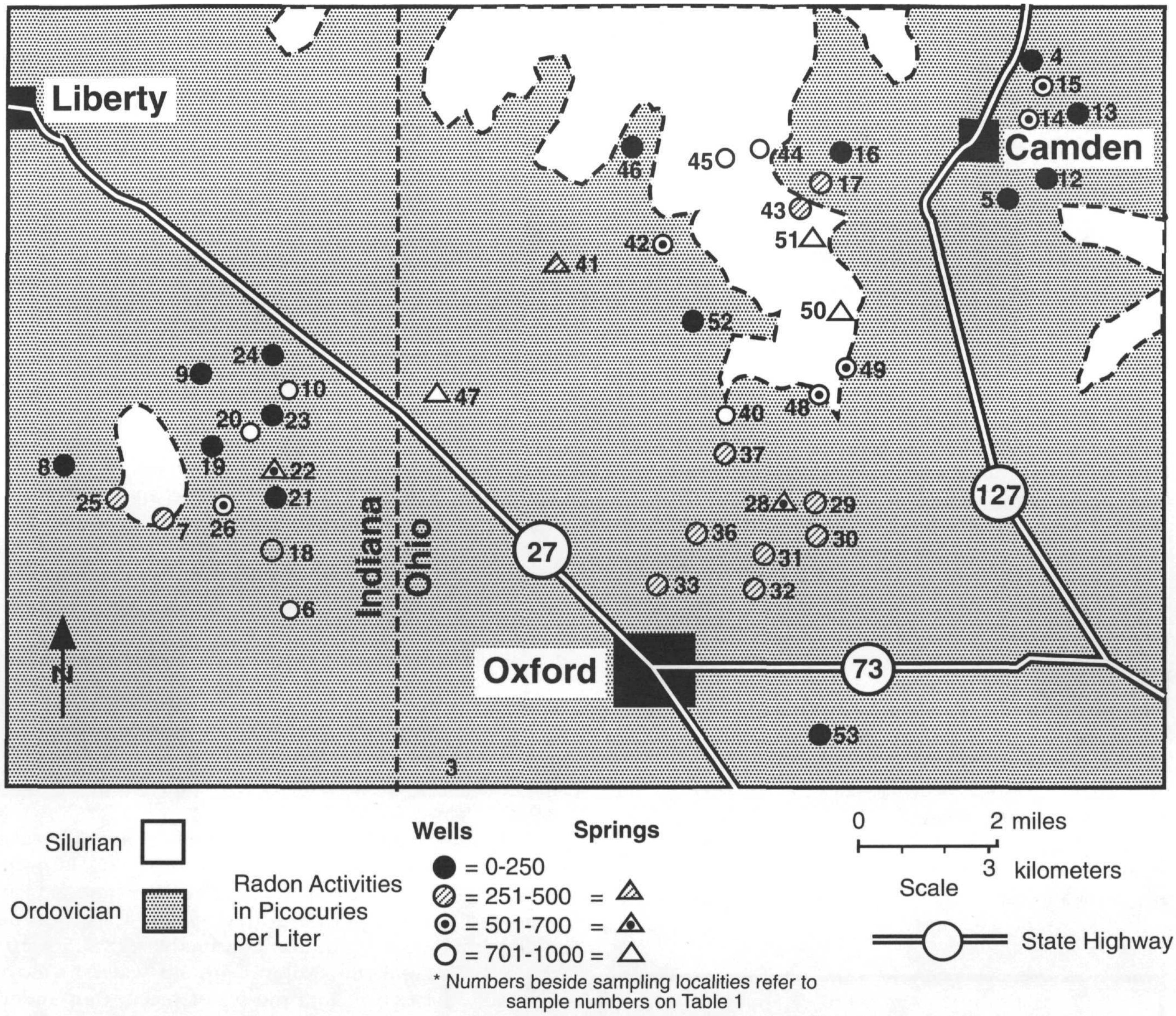

FIGURE 2. Bedrock geology and radon activities of well and spring water in study area (Modified from Gray et al. 1972).

is calcareous (Goldthwait et al. 1981). The hydraulic conductivity of this unit is unknown.

Field Collection

The area of study (shown in Fig. 4) was selected because the boundary separating the Upper and Lower Shelbyville Tills crosses the region. In addition, the Camden Moraine and the Crawfordsville Till also lie at the surface in the northeastern portion of the area. The study design assumed that a significant portion of the radon signature acquired by ground water in each subarea is imparted during movement of the water through the surface till unit. This assumption was necessary because of the inability to determine from driller's logs whether one or more drift units is yielding water to the well.

Appropriate wells and flowing springs were then selected and well logs were obtained where available from the Ohio Department of Natural Resources and the local Health Departments of Preble and Butler counties. Where possible, wells were selected that did not penetrate the drift-bedrock interface. Logs for wells in Indiana were not available for this study.

Details concerning the water supply were determined

WISCONSINAN	Camden Stade	Knightstown Phase
		Crawfordsville Interphase
		Crawfordsville Phase
		Shelbyville Interphase
		Shelbyville Phase
GLACIAL	*Connersville Interstade*	
	Fayette Stade	
	Sydney Interstade	
STAGE	Fairhaven Stade	
	New Paris Interstade	
	Whitewater Stade	

FIGURE 3. Wisconsin stratigraphy in study area (Modified from Goldthwait et al. 1981).

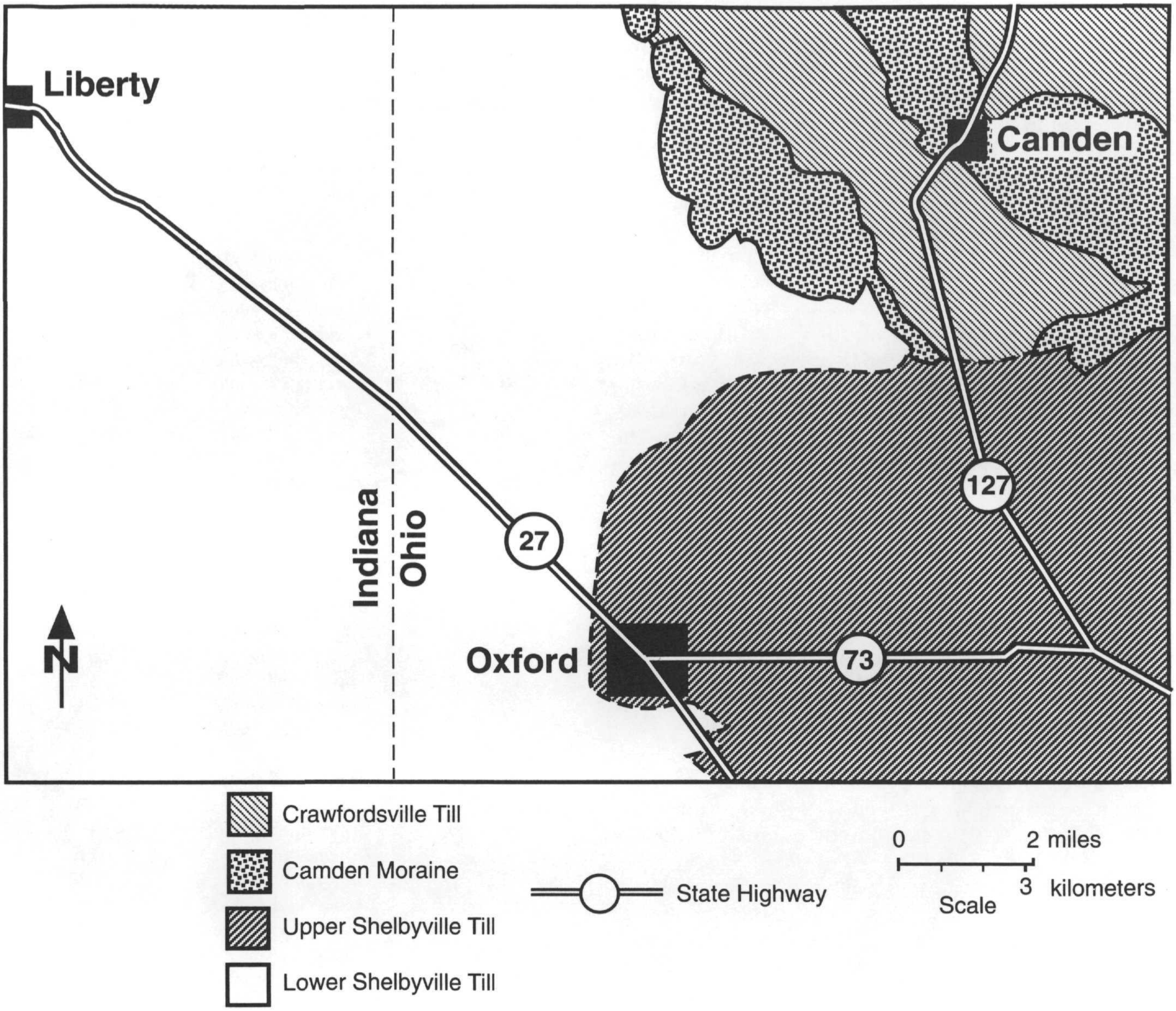

FIGURE 4. Surface tills in the study area.

from each home owner. These included the size of the holding tank, distance of faucet from the well, the type of treatment systems used in the home, and the amount of water used prior to sampling. This was done to determine the length of time it would take to purge stagnant water from the system to obtain a fresh, representative sample. As recommended by the U.S. Environmental Protection Agency (1978), stabilization of the water temperature as it flowed from the faucet was used as an indication that fresh water was being obtained from the aquifer.

After temperature stabilization was achieved, faucet flow was decreased and a funnel and hose apparatus was attached to the faucet. Water was allowed to slowly accumulate in and then overflow the funnel. This minimized agitation of the water and thus decreased the possibility of radon escaping from the sample. Ten ml of water were drawn slowly from the funnel into a 20 ml syringe and immediately injected slowly beneath a 10 ml volume of scintillation oil that had been premeasured into a 20 ml, quartz-scintillation vial. The vial was then sealed, shaken vigorously for 10 seconds to dissolve the radon in the oil and the time the sample was collected was recorded. This procedure is in accordance with that suggested by the USEPA (1978).

Forty-seven samples were collected from wells and flowing springs in the study area (Fig. 2). Duplicate samples were collected at each site to ensure reproducibility of the radon counts.

Sample Analysis

All samples were taken to the Geology Department at Miami University for analysis of radon activity on a Beckman 1900 liquid scintillation counter. A period of at least three hours passed before the samples were analyzed to allow the daughter decay products to equilibrate with the radon. Time between sample collection and liquid scintillation analysis was kept as short as possible, however, to minimize statistical counting error caused by decay of the radon. Radioactivity of each sample was determined for 50 min with the pulse height analyzer set from 0 to 1,000. This permitted the counting of all electric pulses no matter what the energy of the photo-electrons striking the photomultiplier tubes.

The resulting counts per minute then were converted to picocuries per liter (pCi/l) based upon a radium standard provided by the USEPA Environmental Monitoring Systems Laboratory in Las Vegas, NV. This radium/radon standard was diluted with deionized water to an activity expected for radon in the study area and, after sitting for approximately 30 days to allow the radium to reach secular equilibrium with radon, the standard was sent to the USEPA laboratory in Montgomery, AL, to determine the actual radon activity. The arithmetic mean of the radon activities measured in duplicate samples collected at each sampling location was assumed to represent the radon activity in the ground water.

RESULTS

These mean radon values (Table 1) were statistically analyzed using an Analysis of Variance Test (ANOVA) to determine if variability of radon activities between groups was sufficient to differentiate statistically among water samples which had flowed through the Lower Shelbyville, the Upper Shelbyville, and the Crawfordsville tills. The ANOVA provides no statistical evidence that the mean activities of well water taken from each of the three units were significantly different (F-value = 1.16; $p = 0.32$).

Mean radon activities in well waters were plotted against the surface till at the site where the sample was collected (Fig. 5). This figure groups mean-radon values in ranges of 100 pCi/l. For example, there are five wells which penetrate the Upper Shelbyville Till and which produce water with radon activities between 200 and 300 pCi/l. The plot demonstrates the wide range of radon values that exists not only between the three till groups but also within any one till unit.

DISCUSSION

The high variability in radon activity in water from wells within each till unit may result from several factors. First, purge volumes and pumping rates varied for each well. Thus despite the care taken to collect water representative of the aquifer, it is difficult to be sure that this goal was always achieved.

Secondly, the study assumed that the radon signature was imparted to ground water by passage through the surface till unit. It was impossible to determine which drift unit actually was producing the water.

The controlling geologic variable, however, is most likely the variation in hydraulic conductivity near each well. These zones of high hydraulic conductivity also are horizons of preferential flow; thus they transmit large volumes of water carrying dilute concentrations of uranium and radium. Iron and manganese oxides, commonly observed in these more permeable sand and gravel units, effectively adsorb this radium and uranium thereby resulting in higher radon activities in the water upon the decay of the adsorbed radium.

TABLE 1

Radon activities of well and spring water in study area.

Water	Glacial Till	Radon Activity (pCi/l)
Well		
3	Lower Shelbyville	433
6	Lower Shelbyville	925
7	Lower Shelbyville	328
8	Lower Shelbyville	223
9	Lower Shelbyville	224
10	Lower Shelbyville	903
18	Lower Shelbyville	421
19	Lower Shelbyville	129
20	Lower Shelbyville	868
21	Lower Shelbyville	205
23	Lower Shelbyville	187
24	Lower Shelbyville	205
25	Lower Shelbyville	331
26	Lower Shelbyville	522
16	Upper Shelbyville	197
17	Upper Shelbyville	296
28	Upper Shelbyville	671
29	Upper Shelbyville	376
30	Upper Shelbyville	282
31	Upper Shelbyville	291
32	Upper Shelbyville	346
33	Upper Shelbyville	496
36	Upper Shelbyville	456
37	Upper Shelbyville	439
40	Upper Shelbyville	716
42	Upper Shelbyville	628
43	Upper Shelbyville	395
44	Upper Shelbyville	786
45	Upper Shelbyville	710
46	Upper Shelbyville	227
48	Upper Shelbyville	554
49	Upper Shelbyville	555
52	Upper Shelbyville	147
53	Upper Shelbyville	223
4	Crawfordsville	152
5	Crawfordsville	218
11	Crawfordsville	235
12	Crawfordsville	182
13	Crawfordsville	94
14	Crawfordsville	592
15	Crawfordsville	555
Spring		
22	Lower Shelbyville	544
47	Lower Shelbyville	748
28	Upper Shelbyville	671
41	Upper Shelbyville	391
50	Upper Shelbyville	817
51	Upper Shelbyville	903

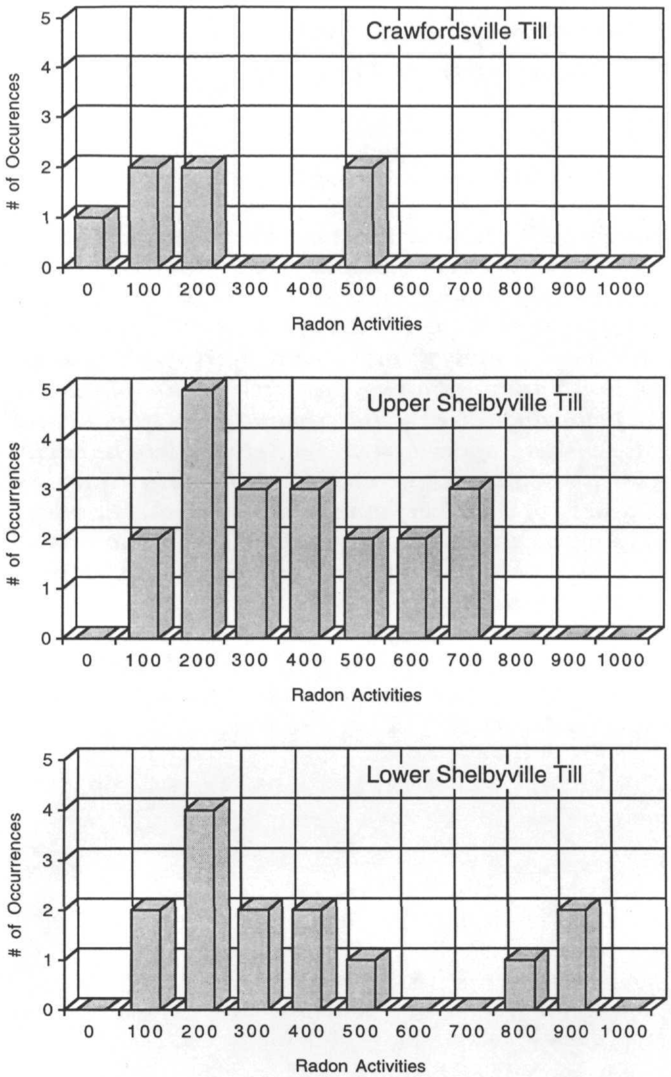

FIGURE 5. Histogram plotting radon activities in ground water against the surface tills at the well sites where the water was collected.

Two of the wells (sites 10 and 40) with radon activities in the highest range (701 – 1,000 pCi/l) were shallow hand-dug wells lined with field stone. These wells are likely producing water from shallow sand and gravel lenses of high hydraulic conductivity between till units, and, despite the large surface area of the standing water in the well through which radon could be degassed, these waters were still highly radiogenic.

Spring water also had anomalously high radon activities. The mean of six samples is 679 pCi/l compared to a value of 431 pCi/l for all the well samples. This finding, similar to that reported by Baldwin and Treick (1991), is supportive of the hypothesis that zones of elevated hydraulic conductivity in the aquifer contain elevated radon activities. These springs occur only where saturated zones containing high hydraulic conductivity are cut by the surface topography.

Work by Gall et al. (1995) supports the direct relationship between zones of higher hydraulic conductivity, elevated uranium concentration adsorbed on the rock matrix, and elevated radon activities in ground water. They measured the vertical variability in radon activity in two wells penetrating limestone aquifers in the Dayton, OH, area and found that the highest radon concentrations were always associated with horizons of highest hydraulic conductivity and uranium concentration in the rock matrix.

Variability caused by sampling or analytical errors is believed to have been minimal because radon activities of duplicate samples from each well were consistently in close agreement.

Although not confirmed statistically, visual inspection suggests that there is some correlation between well water and springs containing elevated radon activities and areas where the rocks of the lowermost Silurian system subcrop beneath the drift (Fig. 2). Baldwin and Treick (1991) found a similar relationship between high radon activities and spring and well water issuing from carbonate aquifers of lower Silurian age. It thus appears that there is some movement of ground water from these carbonate aquifers upward into the nonindurated, Pleistocene aquifers that blanket the region.

The probable source of radon is the decay of radium adsorbed on fragments of uraniferous Devonian black shale incorporated in the drift. Unresolved by this study, however, is the possible contribution of uranium and radium applied to agricultural fields as a contaminant in phosphate fertilizer.

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