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MAPPING THE RADON POTENTIAL OF THE UNITED STATES: EXAMPLES FROM THE APPALACHIANS

Linda C.S. Gundersen and R. Randall Schumann
U.S. Geological Survey, Denver, CO 80225-0046, USA

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The geologic radon potential of the United States was recently assessed by the U.S. Geological Survey. Results indicate that approximately 33% of the U.S. population lives within geologic provinces where the average indoor radon levels have the potential to be greater than 4 pCi/L (147 Bq/m³). Rock types most commonly associated with high indoor radon include: 1) Uraniferous metamorphosed sediments, volcanics, and granite intrusives, especially those that are highly deformed or sheared. 2) Glacial deposits derived from uranium-bearing rocks and sediments. 3) Carboniferous, black shales. 4) Soils derived from carbonate rock, especially in karstic terrain. 5) Uraniferous fluvial, deltaic, marine, and lacustrine deposits. Different geologic terrains of the eastern United States illustrate some of the problems inherent in correlating indoor radon with geology. The Central and Southern Appalachian Highlands of the eastern United States have not been glaciated and most soils there are saprolitic, derived directly from the underlying bedrock. Regression analyses of bedrock geologic and radon parameters yield positive correlations ($R > 0.5$ to 0.9) and indicate that bedrock geology can account for a significant portion of the indoor radon variation. In glaciated areas of the United States such as the northern Appalachian Highlands and Appalachian Plateau, the correlation of bedrock geology to indoor radon is obscured or is positive only in certain cases. In these glaciated areas of the country, it is the type, composition, thickness, and permeability of glacial deposits, rather than the bedrock geology, that controls the radon source.

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INTRODUCTION

The relationship between geology and radon has been documented since the late 1970s, most notably in uranium exploration literature. Among the first to document a correlation between regional geology and indoor radon were Sachs et al. (1982). More recent studies have confirmed that this relationship is complicated and dependent on climate, terrain, bedrock composition, soil permeability, soil texture, and other parameters particular to the region's climate and geology. Geology controls the chemical composition of the rocks and soils from which radon is derived. It also controls the distribution of certain surficial deposits like glacial till and river alluvium. Climate exerts a strong control over the temperature of soils and their moisture content, thus affecting radon emanation and physical and chemical weathering of the soils and rocks.

Indoor radon assessments often rely on *a priori* factors such as bedrock geology or permeability to predict the potential of an area for radon. A regional-scale indoor radon assessment of the United States using such factors was recently completed as part of an interagency agreement between the U.S. Environmental Protection Agency (USEPA) and the U.S. Geological Survey (USGS). The purpose of this radon assessment was to help identify and prioritize the different radon areas of the country and supply information for the application of radon-resistant building codes. A simplified version of the geologic radon potential map of the United States is presented in Fig. 1. This map illustrates the radon potential of the country by the major geologic provinces relevant to the radon problem. It incorporates evaluations of radioactivity, soils, surficial materials, and bed

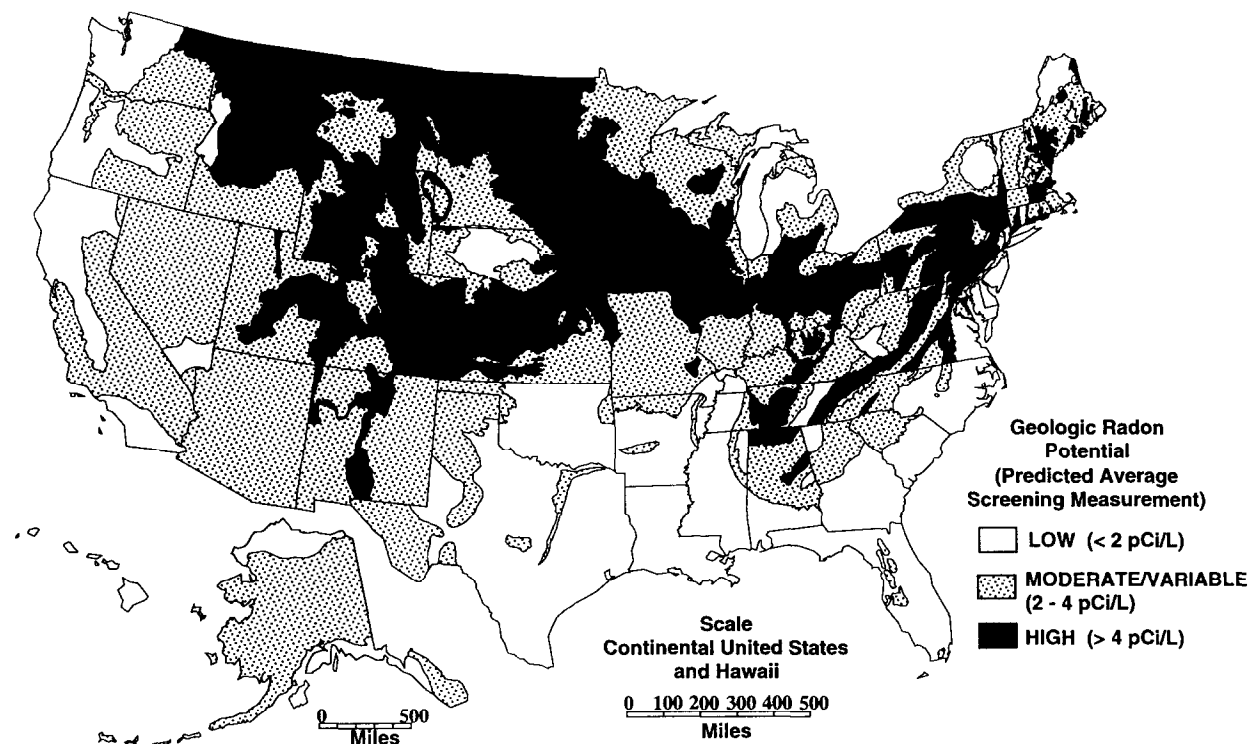


Fig. 1. Generalized Geologic Radon Potential Map of the United States.

rock lithology in the context of screening indoor radon measurements for the country (Gundersen et al. 1992; Schumann 1993).

The method used to identify the different radon areas shown in Fig. 1 is divided into two basic parts, a Radon Index (RI), used to rank general radon potential (Table 1), and a Confidence Index (CI), used to express the level of confidence in the prediction based on the quantity and quality of the data (Table 2). This methodology works best for areas delineated by geologically-based boundaries (geologic provinces) rather than political ones (state/county boundaries) in which the geology may be inconsistent across the area.

To calculate the radon index, five main categories are evaluated and a point value of 1, 2, or 3 is assigned to each category. Indoor radon data from the EPA/State Indoor Radon Survey were used for the 34 states for which data are available (Ronca-Battista et al. 1988; Dziuban et al. 1990) for the remaining 16 states, data from The Radon Project (Cohen and Gromicko 1988; Cohen 1990) and radon data from individual state indoor radon surveys were used. Indoor radon is evaluated using unweighted arithmetic means of the combined (basement and first-floor) indoor radon data. Aerial radioactivity data are from an equivalent uranium map

of the conterminous United States compiled using data from the National Uranium Resource Evaluation (NURE) program of the 1970s and early 1980s (Duval et al. 1989).

The geology factor is complex and actually incorporates many geologic characteristics. In the matrix, positive and negative refer to the presence or absence and distribution of rock types known to generate elevated radon in soils or indoors. The term variable indicates that the geology within the region is variable or that the rock types in the area are known or suspected to generate elevated radon in some areas but not in others due to compositional differences, climatic effects, localized distribution of uranium, or other factors. Geologic information indicates not only how much uranium is present in the rocks and soils but also gives clues for predicting general radon emanation and mobility characteristics.

The soil permeability refers to soil characteristics that influence radon concentration and mobility, including soil type, grain size, structure, soil moisture, drainage, and permeability. In the matrix, low refers to permeabilities less than about 0.6 in (1.5 cm)/h; high corresponds to greater than about 6.0 in (15 cm)/h in U.S. Soil Conservation Service soil percolation tests. One of

Table 1. Radon Index Matrix used to evaluate the geologic and other parameters for estimating the radon potential of the United States.

Factor	Increasing Radon Potential → → → → → → →		
	Point Value		
	1	2	3
Indoor radon (average)	< 2 pCi/L	2 - 4 pCi/L	> 4 pCi/L
Aerial radioactivity	< 1.5 mg/kg eU	1.5 - 2.5 mg/kg	> 2.5 mg/kg
Geology*	negative	variable	positive
Soil Permeability	low	moderate	high
Architecture Type	mostly slab	mixed	mostly basement

**Geologic field evidence (gfe) points*: GFE points are assigned in addition to points for the Geology factor for specific, relevant geologic field studies. See text for details.

Geologic evidence supporting:	High radon	+2 points
	Moderate	+1 point
	Low	-2 points
	No relevant geologic field studies	0 points

<i>Scoring:</i>	Rn potential category	Point range	Probable indoor Rn average
	Low	3-8 points	< 2 pCi/L
	Moderate	9-11 points	2 - 4 pCi/L
	High	12-17 points	> 4 pCi/L

Possible range of points = 3 to 17

Table 2. Confidence Index Matrix for evaluating the quality of geologic and other parameters used to estimate the radon potential of the United States.

Factor	Increasing radon potential → → → → → → →		
	Point Value		
	1	2	3
Indoor Radon Data	Sparse/no data	fair coverage/quality	good coverage/quality
Aerial Radioactivity	questionable/no data	glacial cover	no glacial cover
Geologic Data	questionable	variable	proven geol. model
Soil Permeability	questionable/no data	variable	reliable, abundant

<i>Scoring:</i>	Low confidence	4 - 6 points
	Moderate confidence	7 - 9 points
	High confidence	10 - 12 points

Possible range of points = 4 to 12

the best summaries of soil engineering terms and the national distribution of technical soil types is the National Soils Map (USGS 1970). Architecture type refers to whether homes in the area have mostly basements, mostly slab-on-grade construction, or a mixture of the two. To add additional weight to the geologic factor in cases where additional geologic evidence is available, Geologic Field Evidence (GFE) points are added or subtracted from an area's score. In some cases, geologic models and supporting field data reinforce an already strong (high or low) score; in others, they can provide important contradictory data.

Except for architecture type, the same factors are used to establish a Confidence Index (CI) for the radon potential prediction for each area. Architecture type is not included in the confidence index because house construction data are readily and reliably available. Indoor radon data are evaluated on the distribution and number of data points and on whether the data is consistent, randomly-sampled data or non-random volunteered vendor data. Aerial radioactivity data are given the highest score in unglaciated areas where the correlations with geology and radon levels are reliable, and fewer points in glaciated areas where the correlations are generally poor. For the geologic data factor, a high confidence score is given to an area where a proven geologic model for radon generation and mobility can be applied. Rocks for which the processes are less well known or for which data are contradictory are variable, and those about which little is known or for which no apparent correlations have been found are deemed questionable. The soil permeability factor is scored on quality and amount of data. Percolation test data and other measured permeability data are more accurate than grain size and drainage and score a higher confidence level.

As a result of the present study of the radon potential of the United States and numerous other geologic studies, some general statements can be made about which rocks and soils are radon sources. Rock types that are most likely to cause indoor radon problems include carbonaceous black shales, glauconite-bearing sandstones, certain kinds of fluvial sandstones and fluvial sediments, phosphorites, chalk, karst-producing carbonate rocks, certain kinds of glacial deposits, bauxite, lignite, certain kinds of coals, uranium-rich granitic rocks, metamorphic rocks of granitic composition, graphitic schist and slate, silica-rich volcanic rocks, many sheared or faulted rocks, and certain kinds of contact metamorphosed rocks. Rock types least likely to cause radon problems include marine quartz sandstones,

non-carbonaceous shales and siltstones, certain kinds of clays, silica-poor metamorphic and igneous rocks, and basalts. Exceptions exist within these general lithologic groups because of the occurrence of localized uranium deposits, commonly of hydrothermal origin causing veins in crystalline rocks or roll-front deposits in sedimentary rocks. Uranium and radium are most commonly sited within rocks in heavy minerals, iron-oxide coatings on rock and soil grains, organic materials in soils and sediments and uranium with phosphate and carbonate complexes in rocks and soils, and uranium minerals. Some physical features, most notably faults and shear zones, have been identified as sites of localized uranium concentrations and have been associated with some of the highest reported indoor radon levels (Gundersen 1991).

In the following section we will examine the utility of geology, soil radon, and gamma-ray data in predicting indoor radon and some of the regional and climatic limitations on these types of data with examples from the eastern United States.

METHODOLOGY AND DATA USED

Data presented in this paper includes indoor radon measurements, soil gas radon grab samples, surface gamma-ray spectrometry, aerial gamma-ray spectrometry, and geologic observations of geology and soil.

Indoor radon measurements

Indoor radon values used in this paper are predominantly charcoal canister measurements made in the lowest livable areas of the home over a 2-7 d period of time. Data were collected by the author and others as cited in the text. These data are expressed in units of concentration as pCi/L (147 Bq/m^3).

Indoor radon data are expressed using both arithmetic and geometric means. When indoor radon data are viewed at broad scales such as nationally, regionally, or by state, the data tends to be log-normally distributed. When data are viewed for more homogeneous units such as geologic units, or smaller spatial units, such as cities and counties, the data tends to cluster around a value or several values and tends not to be log-normally distributed.

Gamma-ray measurements

Equivalent uranium (eU) data from surface gamma-ray measurements are used in this report and are expressed in units mg/kg. These data provide an estimate of the surficial concentrations of uranium or radium in

bedrock, soil, and surficial deposits. Equivalent uranium is calculated from the counts received by a gamma-ray detector in the wavelength corresponding to ^{214}Bi with the assumption that uranium and its decay products are in secular equilibrium. Three mg/kg eU corresponds to approximately 1 pCi/g of ^{226}Ra . Surface gamma-ray measurements were made using a multi-channel calibrated gamma spectrometer, gamma radiation was counted for five minute intervals at each ground location and the same machine was used in each study sited.

Radon in soil

Radon in soil gas was determined using a modified version of the method of Reimer (1992). A steel probe of approximately 8 mm diameter is driven into the ground to a depth of 0.75-1m. A grab sample of 20 cm³ is withdrawn through an air-tight septum in a removable cap fitted on top of the probe. Soil air enters the probe through 10 holes which pierce a recessed zone of the probe 1.3 cm from the end. Radon concentration, expressed in pCi/L, was determined using an alpha scintillometer several hours after sampling to eliminate thoron and for each sample to equilibrate. Sampling was performed usually during dry stable periods of time in the spring and summer by the author and others as sited in the text.

RESULTS AND DISCUSSION

Saprolite soils of the Northeast and Mid-Atlantic States and indoor radon

In the Northeastern and Mid-Atlantic States that are unaffected by glaciation, bedrock geology can be successfully used to predict indoor radon, especially in basement homes. Figure 2 presents frequency distributions of indoor radon and soil radon for several major metamorphic rock types in the Piedmont of Maryland (Gundersen et al. 1988). The soils of the Piedmont are saprolitic, meaning that they are derived directly from the underlying bedrock and still retain some of the original bedrock structure. The indoor radon data in Fig. 2 are predominantly basement charcoal canister data and the soil radon data were sampled at a depth of 0.75 m. The frequency distribution of indoor radon in homes varies for each rock type but is generally asymmetrically normal to log-normal. Radon in soil gas varies accordingly. Some rock types, like the gneiss and schist, are more heterogeneous than the phyllite and mafic rock, and this is reflected in the distribution of the

measurements. Figure 2 also shows that soil radon and indoor radon vary systematically with different rock types. Each rock type has a radioactivity signature that is distinctive for that rock type and applicable to other regions of the world with similar rocks, climate, and soil development. The composition and origins of the rock control to some extent the amount of uranium that will be in it. Gneiss and schist in this instance are metamorphosed and deformed sediments, possibly sandstones, shales, and conglomerate, whereas the phyllite is derived mostly from carbonaceous shale. Most of the high readings in both Figs. 2a and 2b are from soils derived from sheared rocks. Shearing of these rocks took place after the sediments had become lithified into rock and metamorphism had begun. Shearing imparts a foliation to the rock causing mineral crystals like quartz and feldspar to become elongated and reduced in size. During shearing, uranium minerals and minerals that contain uranium, like titanite and zircon, breakdown and the uranium migrates to the foliation with iron oxides in an oxidizing fluid and are then redeposited in the foliations.

If the data from Maryland are compared with similar rock types and saprolitic soil from the Reading Prong (Gundersen 1991), the pattern emerges of the relationship among indoor radon, soil radon, and rock type (Fig. 3). The data shown in this figure include the averages of over 1000 indoor radon measurements (basement, charcoal canister) and 1500 soil radon measurements (including the data from Figs. 2a and 2b). Mafic rocks are characteristically a low radon source. Rocks such as aluminous and feldspathic gneiss, schist, and phyllite vary but are generally moderate to high radon sources. Granites and sheared rocks are generally very high radon sources. Another factor reflected in Fig. 3 is permeability. At lower permeabilities, radon migration in the soil is impeded and at higher permeabilities radon migration is favored or may move by flow rather than diffusion. The permeability of saprolitic soils over shear zones is typically high because of the deformation structure imparted to the rock and mimicked in the soil. Further, the rock weathers along the deformation structure, opening the soil structure and increasing the permeability and radon emanation. Permeability of gneiss and schist is moderate compared to their sheared counterparts and soils may be very clayey because of the breakdown of mica and feldspar. Mafic rocks usually have low permeability because many of the minerals in mafic rocks (biotite, hornblende, pyroxene, and serpentine) weather easily and breakdown into clay.

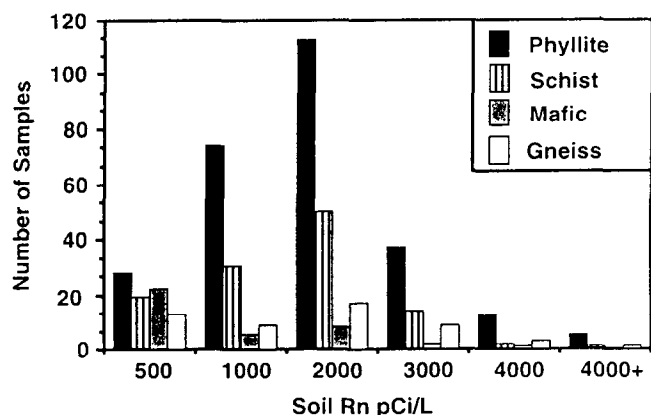


Fig. 2a. Soil gas radon for four major rock types in the Piedmont of Maryland.

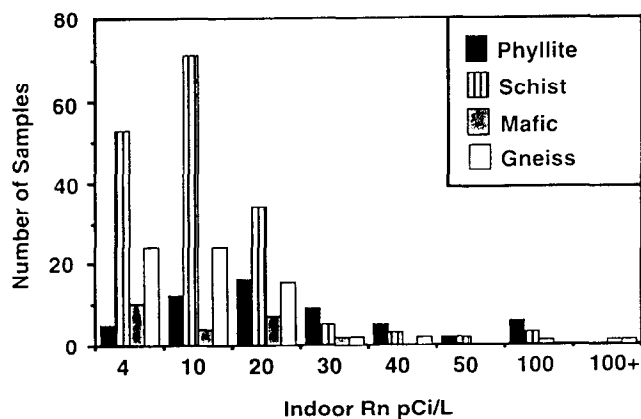


Fig. 2b. Indoor radon (charcoal canister) for four of the major rock types in the Piedmont of Maryland.

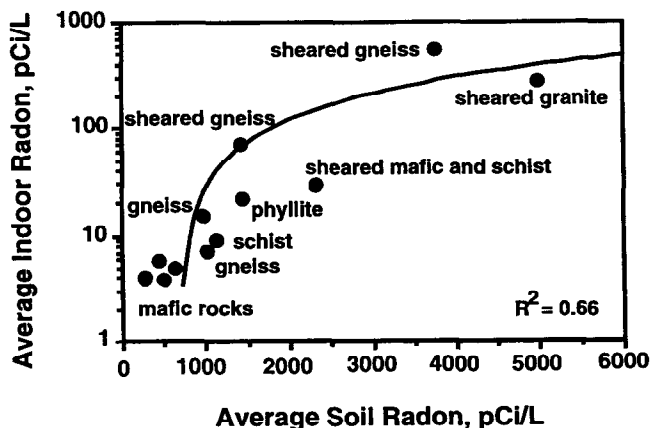


Fig. 3. Average indoor radon (charcoal canister) versus average soil radon for major rock units in the Piedmont of Maryland and in the Reading Prong of New Jersey and Pennsylvania.

Indoor radon in a glaciated area, examples from New York State

Soil radon (485 samples) and indoor radon (126 samples, charcoal canister) were examined in rocks from Orange County, New York, where glaciation has occurred. Here, the bedrock is diverse and consists of sedimentary rocks as well as metamorphic rocks similar to the metamorphic rocks in the previous examples. The bedrock under Orange County consists predominantly of shale, sandstone, conglomerate, and graywacke with several large areas of limestone, dolomite, mafic and granitic gneiss, and granite. The bedrock in Orange County does not have saprolitic soils developed on it. Various glacial deposits have been deposited over the area and are derived from local bedrock and rock sources to the north. These glacial deposits include kame, lake deposits, outwash, ground moraine, and recessional moraine. Moraines are ridges of till that form at the margin of a glacier and their thickness, permeability, and grain size varies over the area. Generally they are poorly sorted mixtures of gravel, sand, silt, and clay. Kame deposits are composed of gravel and sand left by rivers and streams flowing along margins or on top of the glacial ice. Glacial outwash silt, sand, and gravel is typical of the river valleys in Orange County. The glacial lake deposits are composed of fine sand, silt and clay. As can be seen in Fig. 4, a high correlation between indoor radon and soil radon is achieved when the measurements are grouped by glacial deposit and the measurements averaged. The glacial deposit types describe both the texture and permeability of the deposits while the soil gas defines the radon source. When the measurements are grouped by bedrock the regression only yields an $R=0.21$. Several studies of soil radon and

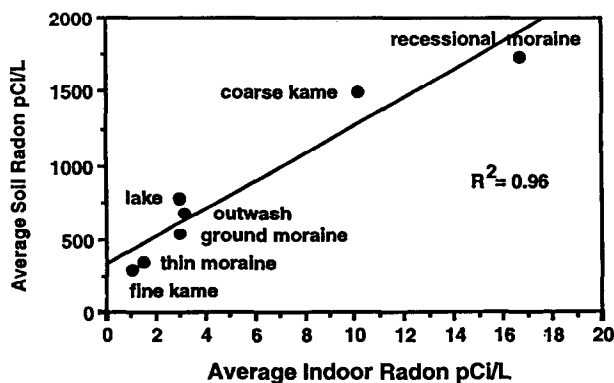


Fig. 4. Average indoor radon (charcoal canister) and average soil radon for glacial deposits in Orange County, New York.

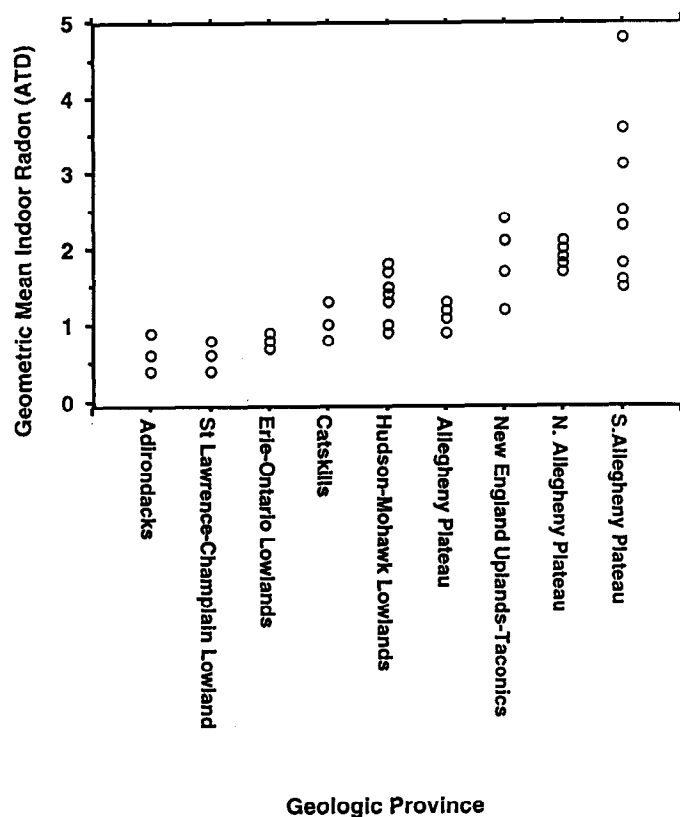


Fig. 5. County geometric mean of alpha-track detector (ATD) data for New York State versus the dominant geologic province of each county.

indoor radon in New York (Kunz et al. 1987; Laymon et al. 1990) and in New Jersey (Gates et al. 1990) also demonstrate clearly that glacial deposits are better predictors of indoor radon and radon sources in soil than bedrock geology. In the northern midwestern states, Schumann (1992) has shown positive correlations between soil gas and different kinds of glacial deposits and used glacial deposits to characterize the radon potential of this part of the United States (Schumann 1993).

Using the geologic concepts presented above, statistical analysis of indoor radon for an entire state can be made in the context of political boundaries such as counties. The state of New York has been conducting radon studies since the late 1970s. Since 1985, the state has had a diverse radon program including information and outreach, radon testing, training for contractors in radon mitigation and detection, and technical and financial assistance (Laymon et al. 1990). As part of their testing program, New York State now has a database of over 50 000 random and non-random charcoal canister indoor radon measurements and over 2000

indoor radon measurements using alpha-track detectors (Perritt et al. 1988). Figure 5 shows the indoor radon geometric mean of the alpha-track detector data for each county plotted by geologic province. The geologic province was determined using generalized maps of geologic provinces for New York State that were refined using bedrock and glacial deposit characteristics that capture the character and magnitude of the radon source.

Each of the geologic provinces has a distinctive bedrock geology overlain by glacial deposits. The Adirondacks are underlain by low-uranium anorthosite and charnokite rock and various metasedimentary and meta-volcanic rocks. These bedrock units are overlain by sand and gravel glacial deposits derived locally and from sandstone to the north. The Allegheny Plateau has been divided into three parts: a southern portion, dominated by uraniferous black shales overlain by gravel and cobble kame and outwash; the central Allegheny Plateau, dominated by shales, limestones, and siltstones overlain by locally derived till; and the Northern Allegheny Plateau, a mixture of siltstone, sandstone, limestone,

Table 3. R-factor analysis for the county geometric mean of indoor radon charcoal canister data (CC); the average equivalent uranium of each county by area; the county geometric mean of alpha-track detector (ATD) indoor radon data, and geologic province. Indoor radon data are from 54 counties in New York State, compiled by the New York State Health Department (1985-1992).

Correlation Matrix

	Geom. Mean CC	eU mg/kg	GM ATD	GeoProv#
Geom. Mean CC	1.000	0.500	0.966	0.712
eU mg/kg	0.500	1.000	0.558	0.542
GM ATD	0.966	0.558	1.000	0.745
GeoProv#	0.712	0.542	0.745	1.000

54 observations were used in this computation.

and minor shale overlain by lake clays and gravel deposits. The Erie-Ontario Lowland is dominated by lake clays. For the purpose of this study, the Hudson Mohawk Lowland province also includes the Tug-Hill Plateau and consists of limestone, sandstone, and minor shale overlain by locally derived sand and gravel deposits. The New England Upland includes metamorphosed rocks of the Taconic Mountains, Hudson Highlands, and Valley and Ridge. The Catskills province includes Devonian sandstones and siltstones overlain by till; it has been grouped with similar Triassic sandstones and siltstones to the south for the purpose of this study.

The geologic provinces were assigned numbers in simple numeric order (from 1 to 9) according to their general geologic radon potential. Table 3 is a correlation matrix showing the R-factor values for a simple regression analysis among the geologic province, the county geometric mean for both the alpha-track detector and charcoal canister measurements, and the average equivalent uranium for the county as calculated from the spectral gamma data acquired during the Department of Energy National Uranium Resource Evaluation (NURE) program (Duval et al. 1989). The alpha-track detector and charcoal canister data have a high positive correlation with each other and with the geologic province. Correlations with eU are also positive.

A multiple regression analysis using county geometric mean of the alpha-track detector data as the dependent variable and the geologic province and eU as independent variables was also performed. A plot of the county geometric mean for the alpha-track detector data calculated from the multiple regression analysis versus the actual county geometric mean for the alpha-track detector data is shown in Fig. 6. The geologic variables alone can account for a significant portion of the variability in the geometric mean. The correlation weakens

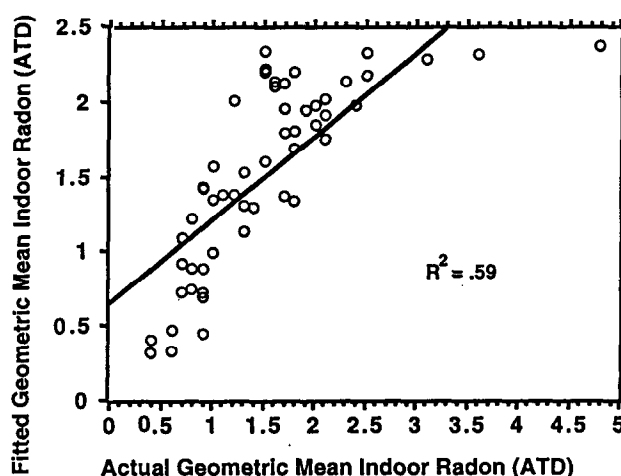


Fig. 6. Plot of county geometric mean of ATD data calculated by multiple regression analysis (Fitted ATD) versus the actual county geometric mean of the ATD data.

with increased indoor radon. Part of the failure of the geologic variables at higher geometric means is related to the variability of the southern Allegheny Plateau deposits and the subsequent variability of the geometric mean of these counties. Splitting out of the counties by permeability and type of glacial deposit, especially those counties in, and south of, the Horseheads Moraine, improves the higher end of the correlation slightly. This multiple regression analysis shows that geologic parameters cannot only be used as indicators of radon potential, but with refinement, can be used to predict radon in the current housing stock.

The indoor radon readings for the glaciated areas of New York are generally lower than those from the Piedmont and Reading Prong. Two factors probably influence this difference (other than the use of arithmetic means, which will be influenced by anomalies in the data set and can be higher than geometric means).

One possible factor is the difference in the maturity of the soils from glaciated to unglaciated areas. Saprolitic soils have been formed by weathering of bedrock material and tend to have more radium in grain coatings than locked in mineral sites, greatly enhancing emanation. Glacial deposits are not as weathered, and radium has not been significantly redistributed. Those glacial deposits that have the highest indoor radon tend to be those that are poorly sorted and have high permeability. The presence of a fine grained fraction of rock material in a coarse, permeable glacial deposit tends to yield the highest emanation. Another possible factor is that the sheared rocks in the Piedmont and Reading Prong are enriched in uranium and contain much higher uranium (as much as 50 mg/kg) than any of the undeformed crystalline rocks or glacial deposits.

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