

THE RADON EMANATION COEFFICIENT: AN IMPORTANT TOOL FOR GEOLOGIC RADON POTENTIAL ESTIMATIONS

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ABSTRACT

The fraction of radon atoms generated from radium decay that are released from into a rock or soil pore space is defined as the radon emanation coefficient, or emanating power, of the material. The emanation coefficient of rocks and soils typically varies between about 0.05 and 0.70, and an average value for soils is probably between 0.20 and 0.30. Geologic, pedologic, and climatic factors, including radium content, grain size, siting of radon parents within soil grains or on grain coatings, and soil moisture conditions, determine the soil's emanating power and radon transport characteristics. Soils derived from similar parent rocks in different regions appear to have significantly different emanation coefficients due to the effects of climate on these soil characteristics. Data on soil radium content, permeability, and moisture content are available for many areas of the United States. With these and data on emanation coefficients, quantitative predictive models for radon generation in rocks and soils at regional to national scales can feasibly be developed. To predict radon generation from soils at a town, neighborhood, or building-site scale, however, would require data at a greater level of detail than currently exists.

INTRODUCTION

Not all of the radon atoms generated by the radium contained in a rock or soil grain are actually released into pore spaces and mobilized. One of three things can happen to a radon atom after it is released by decay of a radium atom: (1) it may travel a short distance and remain embedded in the same grain; (2) it can travel across a pore space and become embedded in an adjacent grain; or (3) it is released into a pore space (Fig. 1). The fraction of radon atoms released into a rock or soil pore space from a radium-bearing grain is termed the "radon emanation coefficient", "emanating power", or "escape-to-production ratio" of the soil. The radon emanation coefficient of typical rocks and soils ranges from about 0.05 to 0.70 (Nazaroff and others, 1988), and it has been suggested that an average value for most soils may be about 0.22 (Rogers and Nielson, 1988). However, it is likely that soils derived from similar parent rocks in different regions may have significantly different emanation coefficients due to the effects of climate on the development of soils from their parent geologic materials. An understanding of these processes is essential to the development of accurate and useful predictive models for radon availability from rocks and soils.

The process of soil formation involves physical, chemical, and biological interactions with the air and water that pass through the soil, commonly resulting in the formation of different chemical compounds than those that existed in the parent material. Parent material composition (mineralogy and chemistry) determines not only the initial amount and distribution of radionuclides in the soil, but also how the rock will interact with climate to form the soil, thus determining the soil's radon emanating power and transport characteristics. As a soil develops, the interaction of the parent rock with climate determine important soil characteristics including grain size and shape, the siting of radon parent nuclides within soil grains, and soil moisture conditions.

When a radium atom decays to radon, the energy generated is strong enough to send the radon atom a distance of about 40 nanometers ($1 \text{ nm} = 10^{-9}$ meters)—this is known as alpha recoil (Tanner, 1980). For a radon atom to escape a soil grain, then, the radium atom must be within the recoil distance of the grain surface (which varies somewhat depending on the density of the material) and the direction of recoil must send the radon atom toward the outside of the grain. Grain size and shape are two of the important factors that control the soil's emanation coefficient because they determine in part how much uranium and radium is near enough to the surface of the soil grain to allow the newly-formed radon to escape into a pore space. The specific surface area (surface area-

to-volume ratio) of soil grains generally increases as grain size decreases, and it is also controlled by grain shape, so that a clay particle, with its small size and platy shape, would have a higher specific surface area, and thus, a higher emanation coefficient, than a larger or more spherical grain with the same radium content. If radium is uniformly distributed throughout the soil or mineral grains, the radon emanation coefficient is inversely proportional to the radius, i.e., grain size (Flügge and Zimens, 1939; Semkow and Parekh, 1990). Expressed in other terms, the radon emanation coefficient increases linearly with increasing specific surface area (Bossus, 1984).

The presence of grain coatings, in which radium is concentrated on the surfaces of soil or mineral grains, increases the emanating power of soils relative to those in which radium is uniformly distributed throughout the grains (Semkow, 1990; Wanty and others, 1992). Iron and manganese oxides, aluminum, silica, and carbonates are common soil weathering products important to radon generation. Silica can form cements through leaching and reprecipitation, or can form aluminosilicate clays by combining with weathering products of feldspars and other minerals. Clays, iron and manganese oxides, and carbonates tend to preferentially sorb or complex uranium and radium, so they can control the distribution of radionuclides in the soil because most radium atoms exposed to soil pore spaces stay attached to the grain surfaces or are contained within cements or grain coatings. Megumi and Mamuro (1974, 1977) postulated that the relative increase in radionuclide contents in soil particles of decreasing size was related to complexing of soluble ions of uranium and radium with iron or other metallic oxides on grain surfaces and that surface adsorption plays an important role in the relative enrichment of radionuclides in smaller grains.

Soil moisture is significant for several reasons. Average soil moisture conditions, generally a direct function of climate, exert a strong control on the rate and intensity of soil weathering and development processes, which, in turn, affect the physical and chemical processes discussed above. A more direct and immediate influence of soil moisture on radon emanation is that of the thin film of water surrounding soil grains which slows radon atoms as they are ejected from the soil grain, increasing the likelihood that the radon atom will remain in the pore space rather than crossing the pore and imbedding itself in an adjacent soil grain (Tanner, 1980; Thamer and others, 1981). Even at wilting point (generally considered to be the lower limit of soil moisture availability to plants), most soils contain such a film of electrostatically-bonded water (Gardner, 1968). At higher soil moisture levels, the pore spaces become filled with more water than gas, and radon atoms are more likely to be retained in the pore water. This has no effect on the emanation coefficient, which is defined as the proportion of radon atoms that are liberated to the pore space regardless of the medium filling that space, but it would have a significant effect on radon concentrations in the gaseous phase and radon gas transport through soils. Partitioning of radon between the liquid and gaseous phases within soil pores was discussed in the context of observed soil-gas radon concentrations by Washington and Rose (1990).

REGIONAL DIFFERENCES IN RADON EMANATION COEFFICIENTS

A preliminary examination of data from field studies in which uranium or equivalent uranium in soils is compared with soil-gas radon concentrations (Gundersen and others, 1988a, 1988b; Schumann and Owen, 1988; Duval and others, 1990; Gundersen and others, 1991; Henry and others, 1991; Schumann and others, 1991; Gates and Gundersen, 1992) suggests that there are regional differences in the amount of radon generated by soils with similar radionuclide concentrations in different climatic zones (Fig. 2). In general, it appears that soils of similar initial composition in moister climates may have higher radon emanation coefficients than those in drier regions. This is likely due to climatically-controlled differences in soil weathering processes. Another possible explanation for the observed regional differences in soil-gas radon versus equivalent uranium (eU) is that different amounts of radon-parent radionuclides are leached from the surface soil layers in different climatic areas. Because the gamma-ray spectrometer used to measure eU primarily detects gamma-rays from the upper 30 cm of soil, these values may not necessarily correlate with the actual amount of radium in the soil at or near the depths sampled for soil-gas radon (0.75-1.0 m). Either or, most likely, both, processes may be affecting the observed regional trends shown in figure 2.

Moisture, temperature, seasonal timing of wetter and drier periods, and rate of soil development exert regional controls on physical and chemical soil properties, which, in turn, potentially affect the soil's radon emanation coefficient. If climate-related trends in radon emanation coefficients follow those of other soil characteristics, both north-south (primarily temperature-related) and east-west (primarily moisture-related) trends across the United States would be expected.

An illustrative example of a dominantly moisture-controlled climatic trend is the development of calcium carbonate horizons. Although carbonate-bearing rocks occur across most of the United States, soils containing calcium carbonate are restricted almost entirely to the semiarid and arid West (Fig. 3). There are few carbonate-rich soils east of the line corresponding to roughly 500-600 mm (20-25 in) of average annual precipitation, largely because in areas with higher rainfall, the highly soluble calcium carbonate is carried out of the soil by percolating water (Jenny, 1941).

Iron oxides in soils are generally more common in areas with warm, dry climates, and temperature appears to exert a stronger control on the occurrence and relative abundance of iron oxides in soils than precipitation (Birkeland, 1984). However, the higher precipitation and denser vegetation of humid climates can also provide favorable conditions for iron accumulation because more water and humic acid are available to the soils, enhancing the rate and intensity of weathering. The midcontinent is a transition zone where iron oxides and calcium carbonate commonly occur together in soils. Because calcium carbonate and, especially, iron oxide coatings on soil grains can enhance the emanation coefficient, this kind of information could aid in predicting areas in which the radon emanation coefficients may be enhanced by climate-related soil processes, other factors being equal.

APPLICATION TO RADON POTENTIAL ESTIMATIONS

In theory, if one had sufficient information on radon emanation coefficients in different rocks and soils and their variation in response to climatic conditions, the emanation data could be used in concert with data on other pertinent soil characteristics to predict the amount of soil-gas radon generated by a particular soil ("radon availability"). Data on the other important soil characteristics affecting radon generation and transport are available for most of the United States at varying scales. The following discussion is intended to illustrate the main components of such an evaluation and their utility in predicting radon availability in soils. It is intentionally generalized and omits discussion of the inherent complexity of some of the factors. However, the level of complexity is in many cases directly related to the scale of the evaluation.

Data on the radium content of soils is of primary importance in estimating geologic radon potential of an area. A consistent source of these data is the aerial radioactivity surveys conducted as part of the U.S. Department of Energy's National Uranium Resource Evaluation (NURE) program of the 1970s and 1980s (U.S. Department of Energy, 1976). These data are now available in digital format on CD-ROM (Phillips and others, 1993). The NURE aerial radiometric data were collected by aircraft in which a gamma-ray spectrometer was mounted, flying approximately 122 m (400 ft) above the ground surface. The aeroradiometric data were smoothed, filtered, recalibrated, and matched with adjacent quadrangle data sets to compensate for background, altitude, calibration, and other types of errors and inconsistencies in the original data set (Duval and others, 1989). The data were then gridded and contoured to produce maps of eU with a pixel size corresponding to approximately 2.5 x 2.5 km (1.6 x 1.6 mi). This resolution obviously restricts use of this data set to relatively large-area evaluations. Other, more local-scale radiometric or radium-concentration data may be available from different sources in different areas.

Because gamma-ray spectrometers provide data on radium concentrations in approximately the uppermost 30 cm of soil, gamma-ray data may sometimes underestimate the radon-source strength in soils in which some of the radionuclides in the near-surface soil layers have been transported downward through the soil profile. Under these conditions the surface gamma-ray signal may indicate a lower radon source concentration than actually exists in the deeper soil layers, which are most likely to affect radon levels in structures with basements. The redistribution of radionuclides in soil profiles depends on a combination of climatic, geologic, and geochemical factors, and there is reason to believe that regionally-variable correlations of eU with actual soil radium and uranium concentrations at a depth relevant to radon entry into structures may be somewhat predictable. More local and regional studies of eU versus soil radium at depth are needed to determine whether a usable model can be established and what its parameters would be.

Data on soil permeability to air and soil moisture conditions are other critical components to radon potential estimations. Data on air-permeability of soils are rarely available, but data on permeability to water, along with information on typical soil moisture characteristics, are widely available and are commonly substituted. Data on permeability to water is an acceptable substitute for air permeability data except in cases in which excessive soil moisture is present. Water in soil pores inhibits gas transport by effectively decreasing the distance that radon can

move through the soil before decaying. In soils with high moisture contents, a significant proportion of the radon may be trapped in the water phase within soil pores (Washington and Rose, 1990), so less radon is available in the gas phase for transport and entry into buildings.

Permeability of soils to water is typically expressed in U.S. Soil Conservation Service soil surveys in terms of the soil's percolation rate in inches per hour, that is, the amount of time needed for an amount of water corresponding to one inch depth to soak into a one-foot square pit in the soil. There is no direct conversion between soil-water percolation units and actual units of soil permeability, i.e., cm s^{-1} or cm^2 . However, soil permeabilities greater than 6.0 in/hr may be considered high, and permeabilities less than 0.6 in/hr may be considered low in terms of soil-gas transport. Soils with low permeability may generally be considered to have a lower radon potential than more permeable soils with similar radium concentrations. Again, more detailed work is needed to properly establish and define a meaningful correlation between infiltration and permeability, and(or) more soil-air permeability data are needed.

SUMMARY

The radon emanation coefficient is defined as the fraction of the total number of radon atoms generated from radium decay that are released into the pore spaces of rock or soil. A soil's parent rock mineralogy and chemistry control the amount and distribution of radionuclides in the soil. Climate is a primary control on soil formation and with parent composition, determines the soil's emanating power and radon transport characteristics. In theory, if reliable, quantitative information on soil radium content, permeability to air, and radon emanation coefficient are available, an estimate of radon production in the soil can be made. Existing data on radium contents are either highly localized or available at relatively generalized scales. Soil-air permeability must usually be estimated from water percolation data, although the scale of available soil maps is typically considerably more detailed than that of radium-content data. Although detailed theoretical information on radon emanation coefficients is available, little field data on emanation coefficients of different soils, in different climatic zones, is currently available. With the collection of sufficient emanation data to allow formulation of empirical models that explain variations of emanation coefficients with climate and soil type, preliminary predictive models for radon availability can be formulated and applied to existing geologic, climatic, soil, and indoor radon data at a relatively large scale. Although the utility of such predictions on a regional, state, or national scale is without question, it remains to be seen at what minimum scale the collecting of site-specific data for predictions remains cost effective relative to building radon mitigation into structures and performing post-construction testing.

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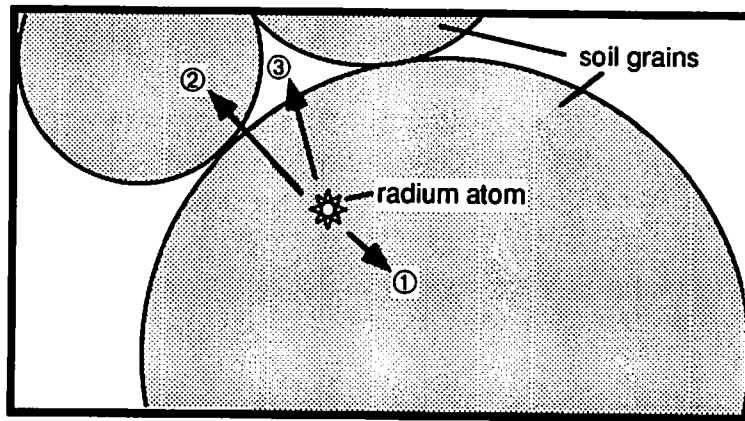


Figure 1. Schematic diagram illustrating three possible cases resulting from radium decay: 1) the newly-formed radon atom is imbedded in the original soil grain; 2) the radon atom is imbedded in an adjacent soil grain; 3) the radon atom is ejected into a pore space (after Tanner, 1980).

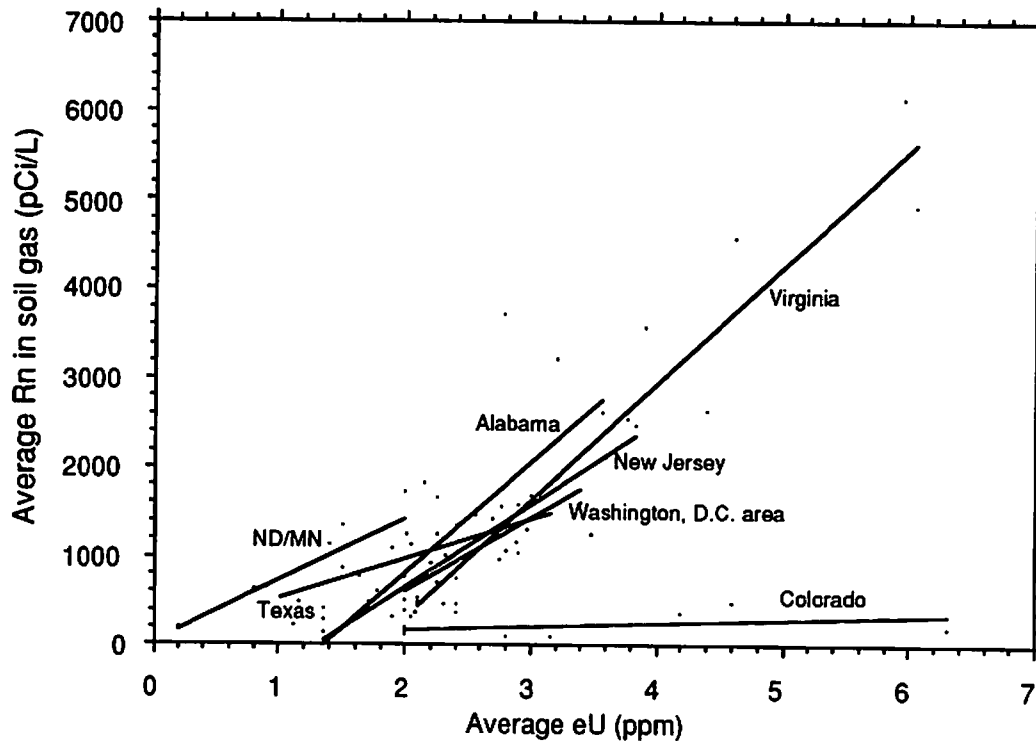


Figure 2. Plot of average equivalent uranium, grouped by rock type, versus average soil-gas radon, for different climatic areas of the United States. The differing slopes of the correlation lines are due to regional differences in emanation coefficients, leaching of different proportions of radium from surface soil layers, or a combination of both effects. Data from Duval and others, 1990; Gates and Gundersen, 1992; Gundersen and others, 1988a, 1988b; Gundersen and others, 1991; Schumann and Owen, 1988; and Schumann and others, 1991.

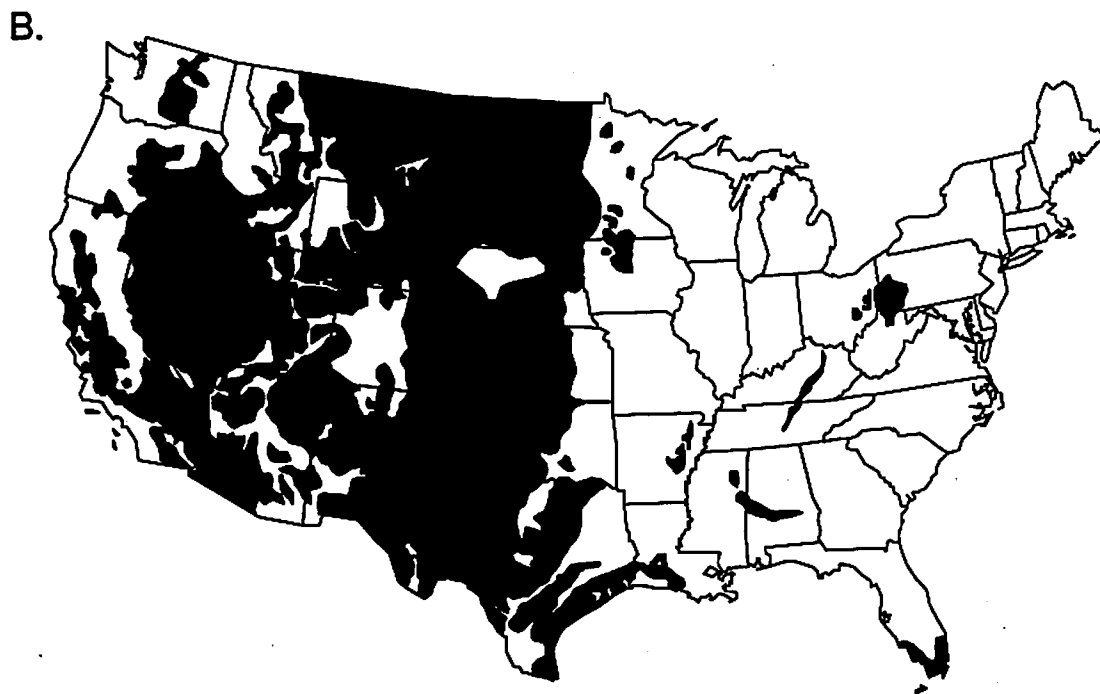
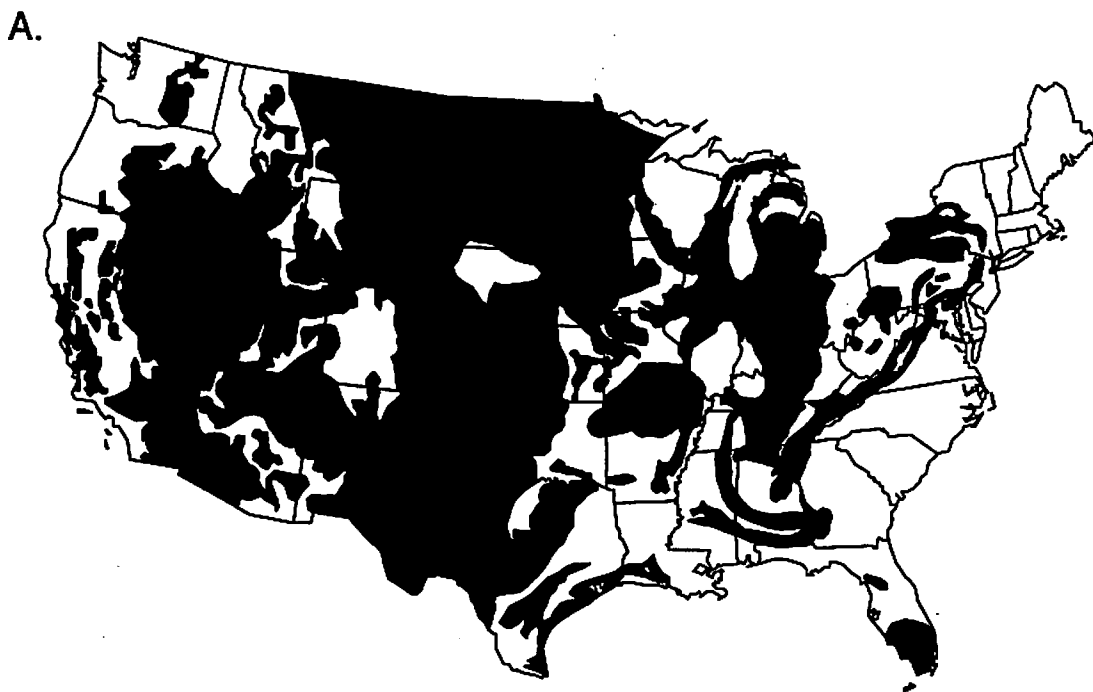


Figure 3. A—Approximate distribution of rocks and surficial deposits of the conterminous United States that contain calcium carbonate. B—Approximate distribution of soils containing significant accumulations of calcium carbonate. After Jenny (1941).