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EVALUATION OF RADON EMANATION FROM SOIL WITH VARYING MOISTURE CONTENT IN A SOIL CHAMBER

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The EPA chamber (2 x 2 x 4 m long) was constructed to study convective and diffusive soil gas movement under known conditions of ^{226}Ra and ^{222} Rn concentration, moisture, density, soil constituent, and physical response to pressure variation. The radon emanation rates of soil are known to depend strongly on the moisture content of the soil. Because the moisture content varies greatly with depth in the EPA's soil chamber (from saturated at the bottom to nearly dry at the top), it is not possible to fully understand the radon distribution within the chamber without knowing the emanation rate as a function of moisture. Soil radon concentrations vary in the chamber from 7.4 kBq m^{-3}, near the soil surface, to 86.2 kBq m^{-3}, at the chamber bottom. This paper describes measurements of the emanation coefficient and diffusion of radon in soil contained in the chamber, using a wide range of moisture contents. In addition, equal amounts of well-mixed oven-dried soil were placed in 20 L aluminized gas-sampling bags, and, after approximately 1 month of in-growth, radon samples were taken, after which water was added, and another period of in-growth and sampling followed. The emanation coefficients and radon concentrations in the gas bag experiment were observed to increase with increasing moisture content and then decrease before reaching saturated conditions. The emanation and diffusion effects on the radon concentration soil gradient were identified for this sandy soil having approximately 200 Bq kg^{-1} radium and a soil density of 1682 kg m^{-3}. *Copyright ©1996 Elsevier Science Ltd*

INTRODUCTION

A complete understanding of ^{222}Rn production and movement through soil and foundation substructures is required to understand radon flow and entry rates into buildings. To better understand entry and design effective countermeasures, a better knowledge of the behavior of radon in soil is essential. Mathematical models describing transport and entry have been developed; however, validation of the models through comparison of simulation and measurements under controlled conditions is needed (Mosley 1992). To simulate conditions of the movement of radon gas through soil, a research chamber has been constructed (Menetrez et al. 1993) containing 16 m^3 of soil with elevated levels of naturally-occurring ^{226}Ra that can generate significant concentrations of radon. Diffusive and pressure-driven flow conditions are monitored along a two-dimensional plane intersecting the central length of the structure that is 2 m wide, 4 m long, and 2 m high. The project is expected to yield valuable information about how radon moves through soil and enters buildings, and consolidate the understanding of other areas of research, such as radon blocking barriers and pressure-driven flows.

Changes in soil radon concentrations are caused by many factors such as, variation in soil radium concentration, soil permeability, homogeneity in radium distribution, density, grain size, soil type, and moisture content (Cothern 1987). To correctly interpret the effects of driving forces on radon concentrations in the central plane of the soil research chamber, an understanding of the effects of moisture variation with depth is necessary.

The release of radon from the solid is a process known as emanation, and the quantitative ratio of radon released from the solid to the total radon formed by decay of radium, is the emanation coefficient (Cothern 1987). As radium decays and newly created radon radionuclides recoil from the parent mineral grain, the available water between grains absorbs the recoil energy and stops the progeny before they are embedded in adjacent particles. In addition, as moisture increases in the pore space, it enhances the direct recoil fraction by absorbing the remaining recoil energy, and the radon atom ends the recoil in the water-filled particle pore space, instead of embedded in an adjacent grain or the originating grain. After progeny are absorbed by the water, they are now free to diffuse through the pores (Tanner 1964). Increasing moisture increases the radon emanation coefficient (by promoting alpha recoil) and available free radon, until saturation is reached, diffusion decreases, and radon in the water-to-air interface decreases. At the point of soil saturation, radon emanation and diffusion decrease (Cothern 1987; Nielson and Rogers 1994).

SOIL BED CHAMBER

The chamber (2 x 2 x 4 m long) was filled with soil that contains 97.5% sand, 1% silt, and 1.5% clay, at a pH of 6.4 to 6.5. In excess of 20 000 kg of soil was used to fill the soil chamber, in increments of 0.5 m by the following procedure: The soil was trucked to the laboratory, manually mixed, and passed through a 1.9 cm metal screen to isolate debris and foreign objects. It was then spread evenly along the surface in increments of 0.5 m, and water was added to the soil until the soil surface was submerged. After 2 days, the water was drained, the soil density and packing density were measured (using a Troxler Nuclear Density Gauge), and the next layer was added. Typically, the soil bulk density averaged 1682 kg m^{-3} after the soil was loaded into the chamber in layers, flooded, and then drained. The moisture retention properties demonstrated that, after equal soil dry weights are packed at various densities and saturated by chamber flooding, the deviation in bulk density exhibited by soil samples is minimal (less than 3%), indicating that the sandy soil, being well drained, will attain near maximum density after chamber flooding, saturation, and drainage (48 h minimum). This was verified by soil laboratory analysis by the North Carolina State University, Soil Science Department (Menetrez et al. 1993).

The soil chamber was made of painted steel plate on the four sides and the bottom, and is ordinarily open to the atmosphere on top (except for experiments which involve covering the open top with a plastic film). Gas recirculating probes extend horizontally from the sides to the center of the chamber for sample collection (Menetrez et al. 1993). Moisture measurement in the chamber was performed by lowering a Troxler 200AP moisture probe through four vertical pipes (3.8 cm ID, schedule 40), sealed and secured at the chamber bottom. The moisture measurement probe was calibrated by Troxler, using a sandy soil and having a maximum or saturated moisture retention level of 35% (volumetric water content). The maximum moisture retention level of 35% (volumetric water content) was also measured in the sandy soil used in the soil chamber and all additional experiments. Moisture was measured by mass balance calculation (Nielson and Rogers 1994), or by use of the Troxler moisture probe.

The variation of radon concentration in soil, spatially and with changing moisture conditions in the soil chamber, is to be investigated. Quantitatively identifying the extent to which moisture is affecting the radon emanation and diffusive transport components of the soil chamber radon concentration gradient is the focus of this study.

EXPERIMENTAL RESULTS

Soil samples were analyzed by Acurex Environmental Inc., the University of Florida, Rogers and Associates Engineering Corporation, and North Carolina State University for various soil properties.

The soil radium concentration was measured at 185 to 222 Bq kg^{-1}. Moisture from the soil-packing process was allowed to drain to limit the level of saturated soil to approximately 6 cm above the chamber floor. The vertical moisture profile above the saturated area near the chamber bottom varies to air dry at the soil surface (Fig. 1).

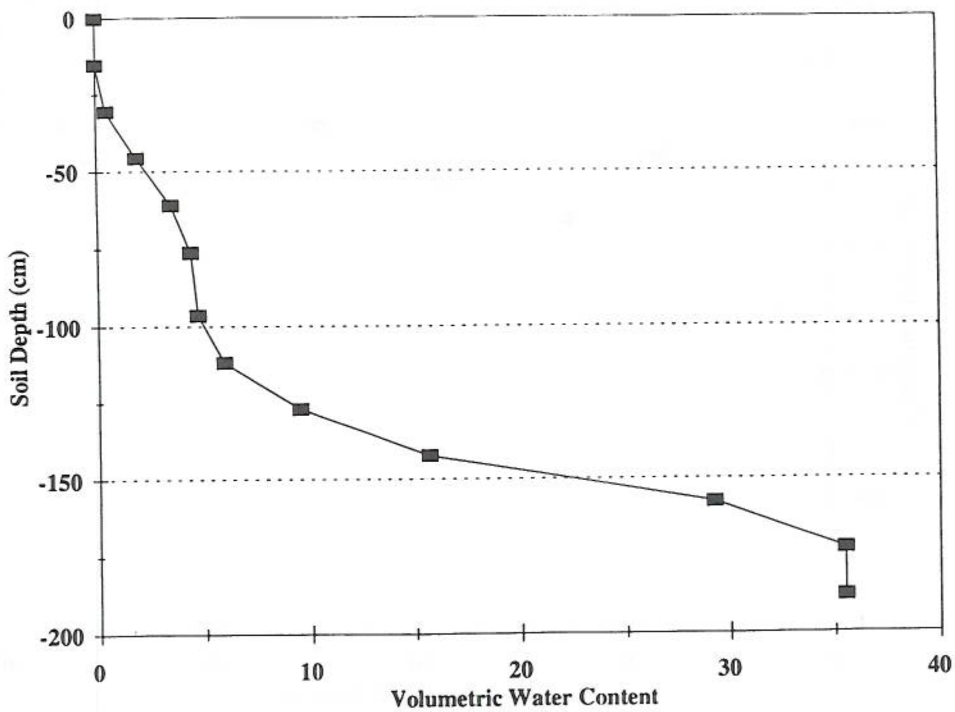

Fig. 1. Soil moisture gradient.

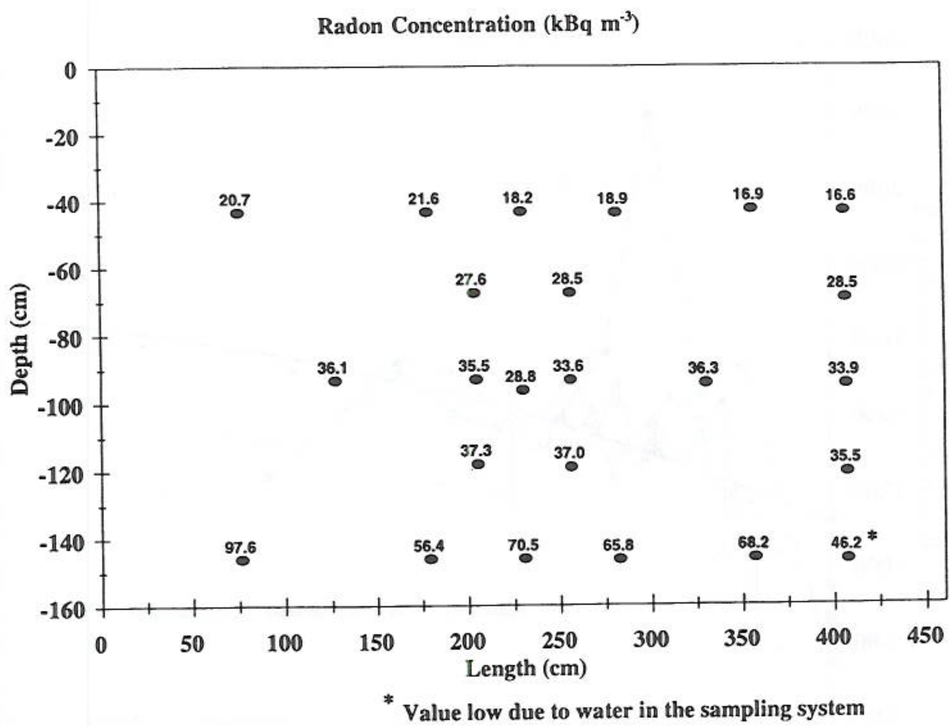

Fig. 2. Radon concentration in open soil gradient.

Steady-state radon concentrations were sampled across the central vertical plain of the soil chamber (Fig. 2). The radon concentrations increased with depth until saturated conditions interfered with sample collection (during sampling, the airflow stream contained significant amounts of water, making airflow difficult). Soil radon concentrations in the chamber were measured from 7.4 kBq m^{-3}, near the soil surface to

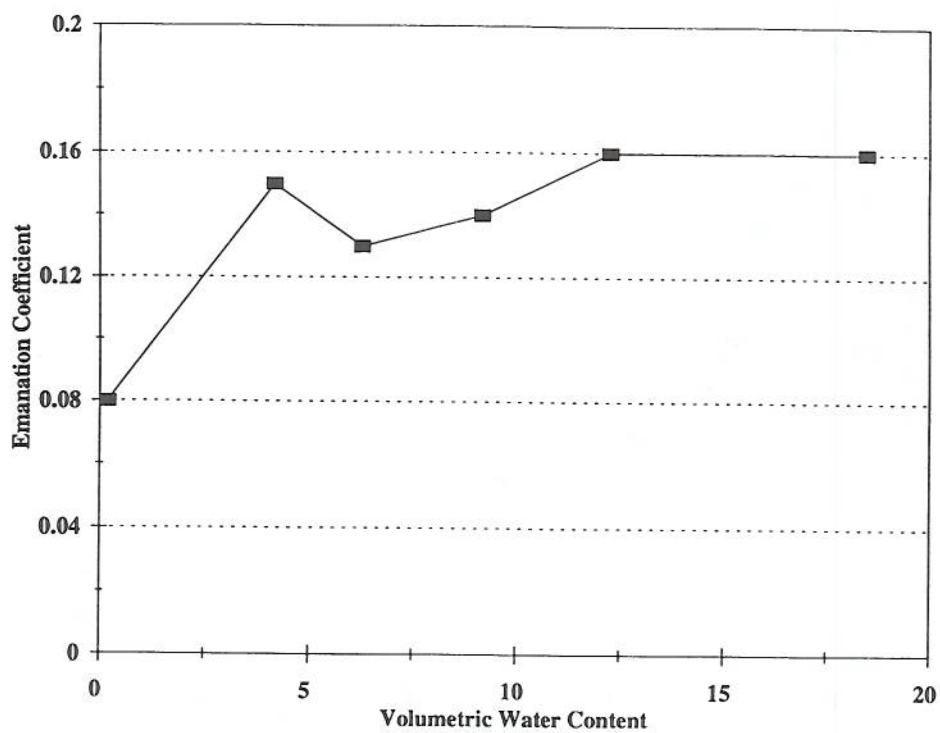

Fig. 3. Emanation coefficient with moisture gradient.

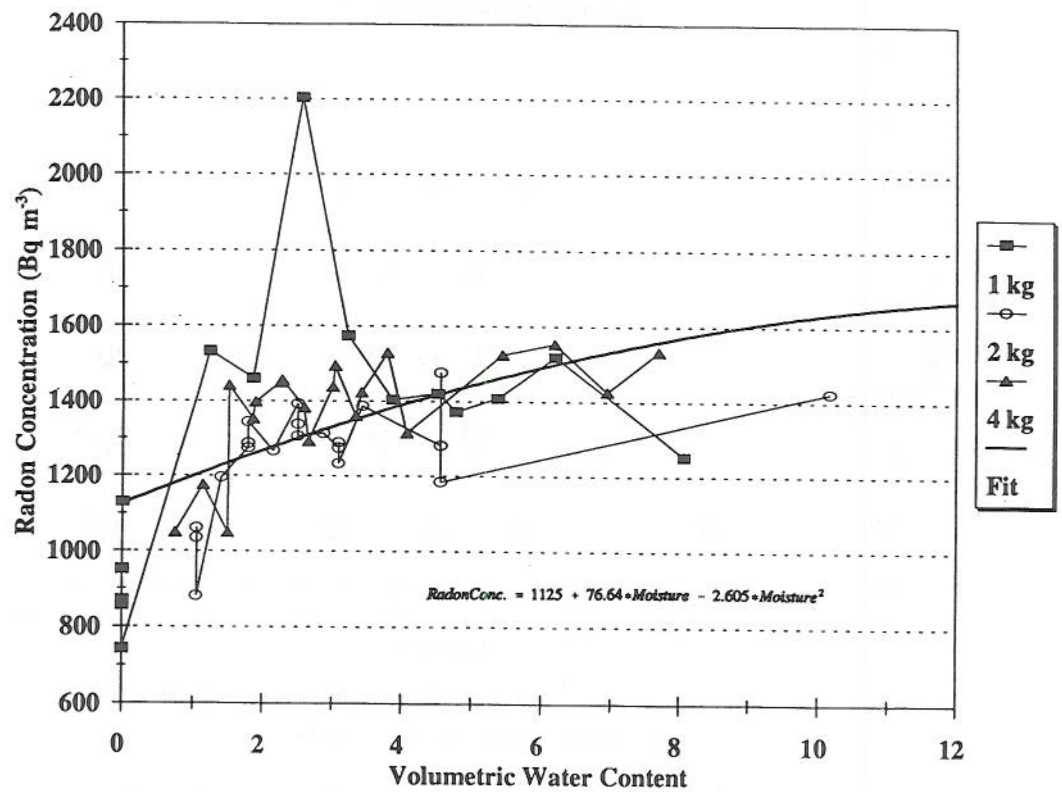

Fig. 4. Radon concentration with moisture gradient.

86.2 kBq m^{-3} at the chamber bottom. This variation was due to the moisture gradient, affecting the radon emanation coefficient, and to depletion of radon by diffusion (Nielson and Rogers 1994). Pressure differentials (measured at all 23 sampling probe sites along the center axis of the chamber) caused by the room HVAC system starting/stopping, or doors opening/closing, or atmospheric barometric changes, were measured (at approximately 2-10 Pa) and found to equalize throughout the open soil chamber within a few seconds. This fast response to equilibrate any pressure change eliminates pressure-driven flow as a source of radon movement, since the distance traveled in that amount of time is insignificant in comparison with the size of the chamber and variation inherent with the measurement of radon. This lack of a pressure gradient in the soil chamber eliminated the possibility of pressure-driven flow conditions.

The emanation coefficient was measured for soil sampled from the soil chamber, and analyzed by Rogers and Associates Inc., using standard testing methods (Nielson and Rogers 1994), for various levels of moisture, from oven-dried to saturated (Fig. 3). The radon emanation coefficient increased from 8 to 17% (±1%), with increasing moisture.

Steady-state radon concentrations were measured by placing soil samples (of the same mixed soil used in the soil chamber), oven-dried and weighed, in 20 L aluminized gas bags with approximately 8 L of air. The five bags were sealed and radon in-growth was allowed for a minimum of 1 month to reach near-equilibrium. Radon concentrations were measured by recirculating air from the gas bags through four scintillation cells. After 4 h, the cells were counted in triplicate hour counts, resulting in 12 concentration measurements per bag condition. The average of the 12 measurements was used to represent the radon concentration at each moisture concentration. To repeat the test for various moisture conditions and amounts of soil (to check for consistency in radon generation per unit of soil), the five bags had measured amounts of water or soil added, and the cycle was repeated. The resulting measurements of radon concentration were made for moisture conditions from dry to saturated, and at three separate soil mass contents. The series of gas bag measurements contained soil masses of 1, 2, and 4 kg. The radon concentrations measured for the normalized soil masses are listed in Fig. 4. The best fit of the radon concentrations as a function of moisture is also graphically displayed by quadratic curve fit ($R^2 = 0.624$). Radon concentrations tend to increase with increasing moisture content, reach a plateau, and then decrease. At saturation, radon measurements (listed in Fig. 4) indicate that radon concentrations decrease to less than that of the dry soil, indicating that the ability of the soil to generate radon gas and exchange it to the air above the soil/water mixture has decreased significantly along with the air/water interface.

The soil chamber was completely sealed from the ambient atmosphere by enclosing the top of the soil surface with 0.9 mils (0.0009 in.) polyethylene plastic taped to the chamber sides. After 1 month, radon levels had reached their maximum concentrations, and equilibrium conditions were achieved without any pressure-driven flow effects. This plastic cover on the soil chamber eliminated the loss of radon from the chamber by diffusion and any minor ambient dilution. The resulting concentrations (Fig. 5) represent radon as a function of emanation, without the effects of the dilution of radon, diffusing out of the soil bed.

DISCUSSION

In summary, the identification of: 1) the soil chamber moisture profile; 2) the soil chamber radon concentration profile; 3) the soil emanation coefficient with increasing moisture concentration; and 4) gas bag radon concentration with increasing moisture concentration, including the best fit of radon concentration as a function of moisture, has been listed in Figs. 1, 2, 3, and 4, respectively. In addition, soil chamber equilibrium radon concentrations without dilution reduction effects are identified in Fig. 5.

The procedures described above isolate the effects, and allow for the prediction of radon emanation and radon concentration in the soil chamber, as a function of moisture, with and without the effects of dilution (Fig. 6). For example, radon emanation increased up to 50% with increasing moisture (Fig. 3); in radon concentration experiments performed with the gas-bag samples, radon increased 50% (Fig. 4); and in radon concentration samples taken in the closed soil chamber, radon increased with soil depth and increasing moisture (Fig. 5). The samples taken in areas of soil approaching saturation (the bottom of the soil chamber) are higher than the samples taken from the upper soil areas by proportions approaching 37%. In these examples, increasing moisture resulted in increased amounts of radon concentrations up to the levels approaching saturated soil, where radon concentrations decreased abruptly, due to a sharp decrease in soil permeability. Unlike the radon emanation coefficient, which increased

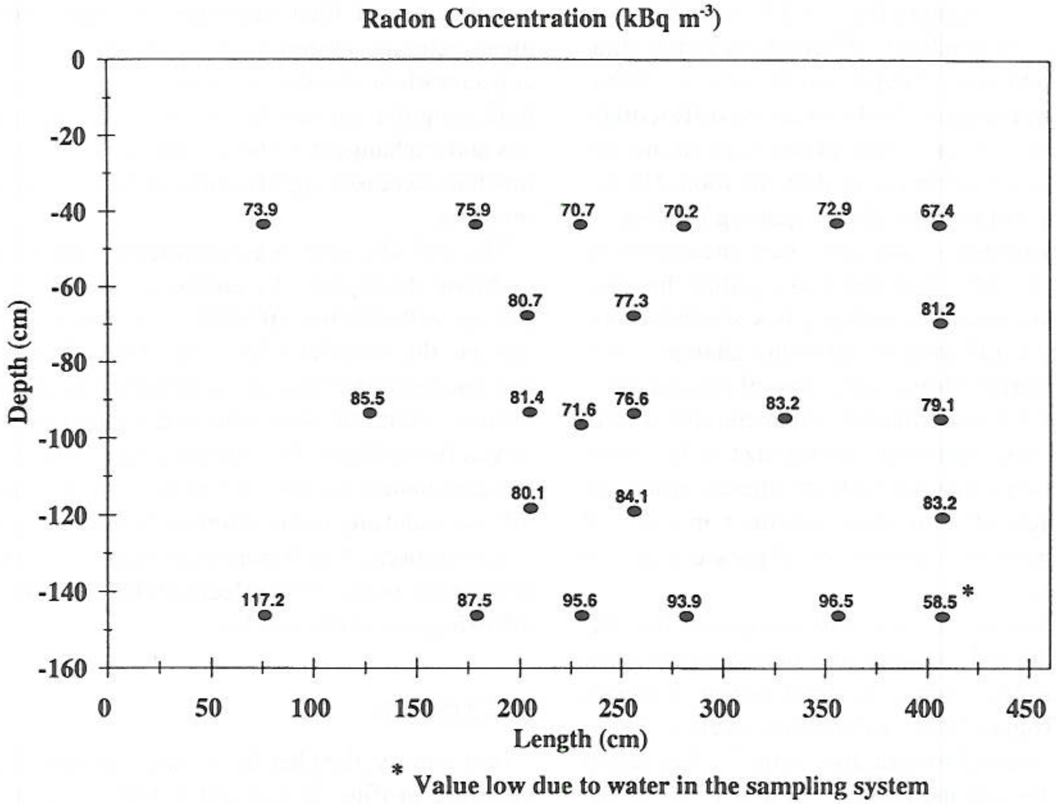

Fig. 5. Radon concentration in closed soil chamber.

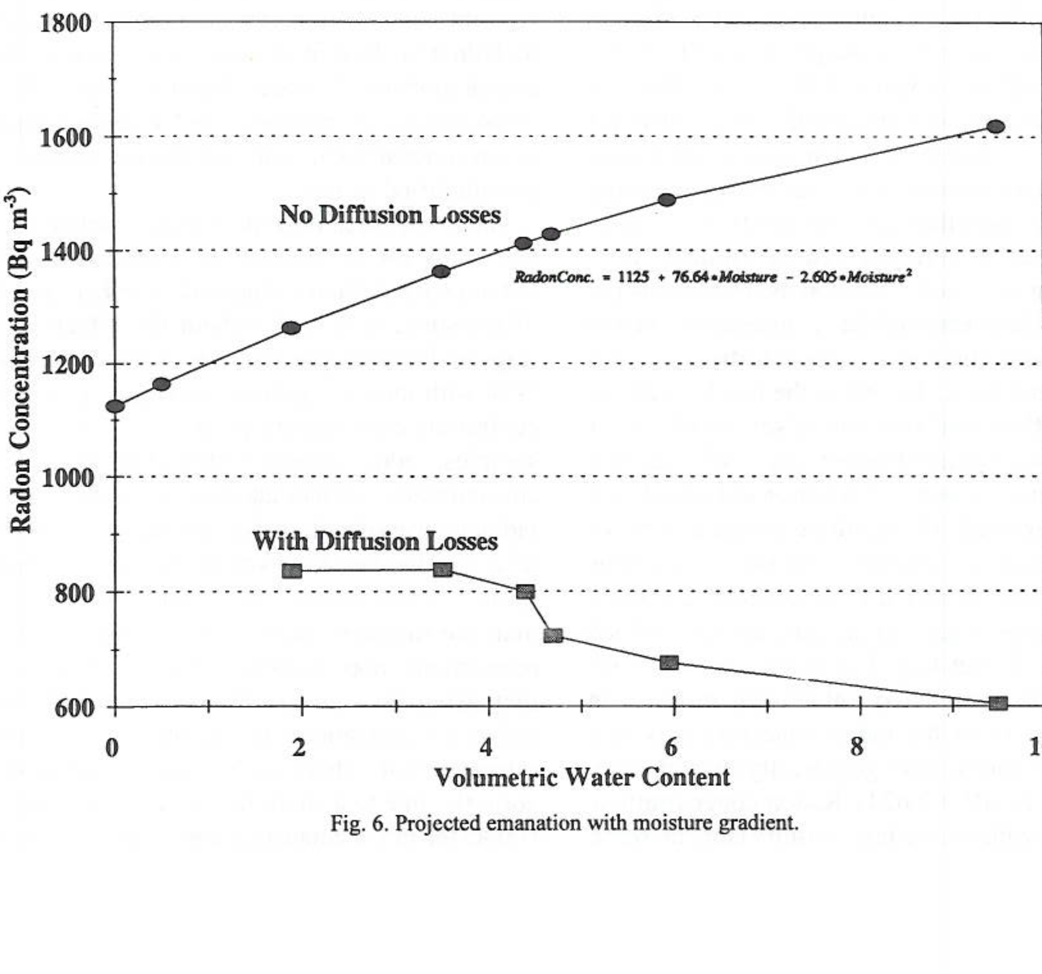

Fig. 6. Projected emanation with moisture gradient.

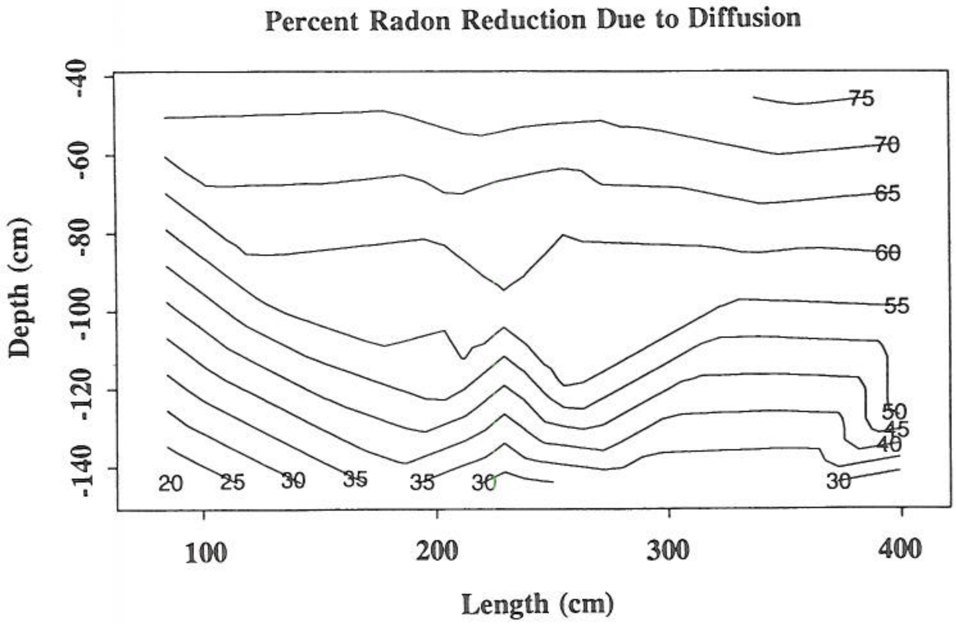

Fig. 7. Percent radon reduction due to diffusion.

to a level and maintained that plateau to saturated soil conditions, radon concentrations decreased with saturation.

A prediction of the radon concentration as a function of moisture with and without losses due to diffusion is illustrated in Fig. 6. A comparison of radon concentrations from the soil chamber taken while the top was open to the atmosphere and those concentrations taken while the chamber was closed (Fig. 7), account for reductions in concentration approaching 75%. The percent reduction in concentrations (represented in isobars) for the central vertical plane of the soil chamber is shown in Fig. 8. This reduction is the result of radon diffusing out of the soil. The effect of diffusion is significant throughout the soil bed, as exemplified by the fact that, at a soil depth of 2 m, a 35% reduction is apparent.

Moisture significantly affects emanation and diffusion and must be taken into account when predicting migration rates and concentrations. Understanding the relationships of radon generation and migration through soil, is valuable in advancing the knowledge of radon and how to better deal with it in limiting human exposure.

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