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GENERATION AND MOBILITY OF RADON IN SOIL

TECHNICAL REPORT AND PROPOSAL FOR FURTHER FUNDING

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ABSTRACT

Research under the current DOE contract has (1) confirmed large seasonal and daily variations of Rn in soil gas, (2) developed models for the effects of temperature and moisture on air-water Rn partition, inhibited Rn diffusion from wet soil into sparse large air-filled pores and effects of diffusion into bedrock, (3) demonstrated that organic matter is a major host for 226Ra in soils and that organic-bound Ra largely determines the proportion of 222Rn emanated to pore space, (4) shown that in contrast 220Rn is emanated mainly from 224Ra in Fe-oxides, (5) detected significant disequilibrium between 226Ra and 238U in organic matter and in some recent glacial soils, (6) demonstrated by computer models that air convection driven by temperature differences is expected in moderately permeable soils on hillsides. Additional research is proposed on 238U-234U-230Th-226Ra disequilibrium in the same soil profiles, on field evaluation of air convection effects, and on the quantitative significance of air convection and moisture effects on the Rn levels in homes.

INTRODUCTION

This report summarizes research during the period June 1990 to July 31, 1992 under the current DOE grant DE-FG02-87ER60577, which expires February 28, 1993. It also proposes additional research on radon problems for a further 3-year period.

Objectives and Approach of Expiring Research Grant

1. To determine the processes that cause large seasonal and short-term changes in the radon (Rn) content of soil gases, and to develop methods of predicting and modeling these variations.
2. To evaluate the relation of Rn emanation coefficients to form of radium (Ra) and other U-series decay products, particularly the role of Ra in organic matter and Fe-oxides.
3. To evaluate the conditions in which convection of gas in

soil and bedrock may affect soil gas radon availability in houses.

4. To collaborate with other DOE researchers on evaluation of Rn flux into houses, using our well characterized soil sites.

To accomplish these objectives, a set of 13 sites representing several parent materials, climatic regimes, and drainage conditions in eastern and central U.S. (PA, NY, NC, TN, IL) have been investigated in detail. A complete soil description plus measurements of radon and thoron in soil gas, and radium, uranium, thorium, air permeability, diffusion coefficient and emanation coefficients of soils have been obtained at each site. Follow-up investigations to evaluate selected problems are continuing. Convection of air in soil is a current focus of attention.

Research Accomplishments

Objective 1. Processes causing seasonal and short-term variations in radon content of soil gases.

1. In extremely wet soils, we have observed unusually low values of Rn in soil gas, especially in winter. Mathematical relations have been developed for two phenomena that seem to explain this observation (Rose and Washington, 1992): decay of Rn during slow diffusion in water-filled pores (Figure 1), and downward diffusion of Rn toward bedrock (Figure 2). An effect similar to the latter is expected for very stony soils, such as our 14-83 site (see below). These relations should be incorporated into computer models for Rn in soils adjacent to houses; also, changes in soil moisture status may have major effects on radon levels in homes.

2. In order to gain additional information on the magnitude of temporal variations and their correlation with other variables, we have continued measurement of Rn, moisture, temperature and other properties at two sites. Figures 3 and 4 show that the large range of variations previously found has continued: At site 14-80 the range at 104 cm depth is a factor of 5, with low values in March and April of 30.3 to 63.3 kBq/m^3, and

high values of 156.8 kBq/m^3 in May. Greater depths show a similar pattern but smaller range. Based on profiles that show no gradient in Rn concentrations toward the surface below about 120 cm, the values at depths below 120 cm should be essentially unaffected by diffusion toward the surface, but the 104 cm values may be affected by changes in diffusive properties caused by changes in soil moisture. The very high Rn in May corresponds to a period of rapid decrease in soil moisture, caused by a combination of increased evapotranspiration by trees plus the beginning of a summer-long drought.

At site 14-83, where very stony soil is developed on sandstone, the range at 157 cm depth is from 4.6 (1/10/92) to 53.3 kBq/m^3 (9/9/91), a range of about a factor of 10. The lowest values were periods in January and February when snow covered the ground; the highest values are in June-September. At 115 cm depth, the range is 9.8 to 45.1 kBq/m^3, a factor of 4.6.

This data confirms the previous measurements showing large temporal variation at depth. As shown on figure 5, most of the variability at site 14-80 can be explained by our previous model of the effects of moisture and temperature on radon concentration, and by changes in diffusion coefficient for the shallow depth. However, the variations at site 14-83 are much too large to explain by these mechanisms. Possible explanations are (1) air convection in the soil gas, and (2) changes in diffusion coefficient caused by increased moisture, in combination with diffusion into the numerous rock fragments in this soil, analogous to the proposal by Washington and Rose (1992) for diffusion into bedrock, as discussed above.

3. We previously obtained data at site 14-80 showing a correlation of ^{222}Rn ("Rn") with ^{220}Rn ("Tn", thoron) (Washington and Rose, 1992). However, the measurements for 1991-2 do not show such a correlation (Figure 6). The Rn-Tn correlation was used by Washington and Rose (1992) to argue against air convection as a cause of the seasonal variability at depth, since Tn variations would require air flow from depth within a few

minutes to account for variability of Tn (half-life 55 sec). The new data throws questions on this conclusion. Most of the apparent variability in Tn appears to be random counting error, so that single high or low values are not reliable (Figure 7). There does appear to be a general pattern of low values in summer vs. low in winter. The summer '91 to winter '91-92 Tn decrease may be explained by temperature/moisture effects, as for Rn. The winter '91 values are too low for this explanation, but are exremely noisy. We no longer consider that the Tn data are a good argument against air convection, especially at site 14-83, which is on a slope of about 5^o in relatively sandy and stony soil.

4. Bulk diffusion coefficients decrease by about an order of magnitude, and permeabilities by about 2 orders of magnitude within individual soil profiles, with the highest values generally at the surface. In some profiles the lowest values are in the B horizon. The diffusion and permeability coefficients correlate, and the diffusion coefficient can be predicted from the permeability, which can be estimated by simple measurements with a driven probe (Washington et al., manuscript; Washington, 1991).

Objective 2. Relation of emanation coefficients to form of Ra

5. Radium, the immediate parent of radon, exhibits high mobility in soil-forming processes, as might be expected from its similarities to Ca and other alkaline earth elements (Greeman, 1992). Vegetation contains large enrichments of Ra, relative to its parent uranium. In the soil, a high proportion of the Ra occurs in the exchangeable plus organic fractions (Figure 8), which has an average Ra/U activity ratio of about 25, with some values exceeding 1000 (i.e., Ra in this organic matter is unsupported). Soil minerals are significantly depleted in Ra relative to U.

Because of enrichment of Ra relative to U in organic-rich surface soils, the content of U in soils is not necessarily a good guide to the content of Ra (and Rn), and gamma activity of ^{214}Bi may not be representative of deeper soils.

Calculations show that although much of the Ra in organic matter of the A horizon originates from Ra contained in dead vegetation, the major part in deeper horizons is transferred from soil solutions into already dead organic matter, or into roots where it is fixed.

6. Radon emanation from soils correlates well with Ra in organic matter (Figure 9). Multiple regression of radon emanation for 26 soils vs. percentages of radium in the organic, Fe-oxide, sand, silt and clay components indicates that about 65% of the emanated Rn results from decay of ^{226}Ra in organic matter, with an emanation coefficient of about 40%. Most of the remaining Rn emanates from silt and clay grains, with an emanation coefficient of about 22%. Emanation from soils attacked by selective extraction reagents confirms this behavior.

The Ra enrichment in the organic fraction and the high contribution of organic matter to Rn emanation imply that construction or mitigation practices affecting soil organic matter, or regional differences in soils, may have a marked affect on Rn concentrations in soil gas and in homes. For example, the regional differences in soil Ra vs. soil-gas Rn detected by USGS geologists may reflect differences in the organic cycle of humid vs. semi-arid soils.

7. Emanation of thoron (^{220}Rn) has also been measured on 62 soil samples. Based on regression analysis and limited measurements on selectively extracted samples, about half the thoron emanation is from silt- and clay-sized particles, and about half is from Fe-oxide coatings on soil particles. The host for thoron differs from radon (^{222}Rn) because of the short half life of thoron precursors, so that long-lived ^{232}Th is the effective parent. Based on this data, the relative emanation of radon and thoron may differ considerably in soils of different types and regions.

8. Many young soils developed on glacial deposits appear to have been leached of U much more rapidly than Ra, leading to excesses of Ra over U in the deeper soil as well as at the surface.

9. In view of the variability found in Ra/U activity ratios of soils and soil fractions, a project to measure ^{234}U and ^{230}Th in the same soil samples was started June 15 by Graduate Assistant Y.J. Chang under the direction of W. A. Jester and A. W. Rose. We are currently acquiring equipment and testing techniques for this work, using the procedure of Anderson and Fleer (1982) as modified by W.C. Burnett at Florida State University. The intent is to test our hypotheses regarding Ra unsupported by ^{238}U and ^{234}U in the soils.

Objective 3. Air convection in soils

10. A Ph. D. student, Weixing Guo, working on the production of acid mine drainage has developed a computer model for thermally driven air convection in spoil piles at surface coal mines. He has generously collaborated in calculating air convection in soil on hillsides, with results shown on Figures 10 to 12. The models assume a uniform permeability of 10^{-7} cm^2, which is similar to permeabilities measured by Washington (1991) at sites 14-80 and 14-83 (Figure 13). The bottom and sides of the model for Figures 10 and 11 are no-flow boundaries and sides have no heat flow; the bottom and top surfaces have fixed temperatures differing by 5 and 15°C with the bottom warmer than the top, as might occur during cold weather in winter (the temperatures are set above freezing to eliminate problems of ice forming from moisture in the air). In most runs the upper surface is open to air flow but in Figure 12 it is closed as it might be just after a hard rain, or with an ice zone at the surface.

The modeled pattern of air convection is in agreement with previous theoretical and observed treatments of convection in sloping or irregular bodies (Bories and Combarnous, 1973). For example, Sturm and Johnson (1991) document air convection in snow in Alaska, and also develop theory showing flow velocities up to 2 mm/s. They point out that in addition to sloping permeable slabs, convection is also likely in media with planar top surfaces but irregular basal surfaces, as might be expected for soils overlying irregular bedrock. Weeks (1987) demonstrates flow driven by temperature gradients in permeable tuff at the Nevada Test Site. Gammage et al. (1992) demonstrate air convection in hilly karst terranes in response to temperature differences between temperatures of outdoor air and soil or rock.

As can be seen from Figures 10-12, air convection at rates of 0.5 to 5 x 10-6 cm/s is found on the hillsides. In flat areas, no convection occurs, as predicted from the Rayleigh criterion (Schery and Petschek, 1983; Washington and Rose, 1992). The velocities along slopes amount to 3 to 40 cm/day, or up to about 2 m in one Rn half-life. In view of the fact that higher permeabilities were measured in some Pennsylvania soils (Figure 13), and that macropores and cracks may also allow easier flow, it appears that air convection is very feasable as a means of Rn transport, and may explain some of the variability observed at our field sites, as well as the variability among houses on similar soils and geology.

11. We are currently attempting to verify the effects of the calculated models by measuring thermal and transport effects on hillsides. At one site on a steep talus slope during a period of little wind, air was clearly flowing out at the base of the slope, and temperatures measured for this air were as low as 9^{o}C compared to an air temperature of 22 to 24^{o}C.

Objective 4. Collaboration with other researchers

12. Surface barrier radon detectors loaned to us by Dr. Donald Thomas (Univ. of Hawaii) have been used to measure short-term variability in Rn at site 14-80. Variations by a factor of about x4 over periods of a few days are recorded (Figures 14, 15), with abrupt changes related to rainfall or melting events. Significant diurnal effects are evident during spring but not in December. Barometric effects are not obvious. Since this data is from a depth of about 0.6 m, a likely cause of variability is changes in diffusion coefficient due to changes in soil moisture.

13. In collaboration with Don Thomas, six surface barrier Rn detectors have been emplaced at three sites located about 1m, 10 m and 50 m from a house in a clayey soil developed on limestone in State College. Temperature, moisture (by neutron moisture

gauge) and soil gas Rn in samples collected from tubing emplaced in the soil, and Rn in the house are also being measured. Rn values measured by the detectors vary widely, apparently in part reflecting soil moisture. However, a major part of the variability is yet to be explained.

Research Proposed for 1993-1996

The objectives for proposed research during 1993-1996 are as follows:

1. How do Ra and Rn in soil depend on the form and behavior of their ancestors ^{234}U and ^{230}Th?
2. Under what conditions can thermally driven air convection in soil have significant effects on Rn transport?
3. Under what conditions do soil moisture and soil air convection affect Rn in homes, and how are these variables relevant in mitigation?

The major variables controlling Rn generation and transport in soils and into houses are relatively well understood, based on research in recent years, but the potential for air convection on slopes and in other soil bodies of non-planar shape has not been seriously evaluated. We believe this topic deserves careful investigation, because it could be responsible for major variation from house to house. Similarly, this process is relevant to many other topics, such as dispersion of volatile contaminants, use of soil gases for petroleum and mineral exploration, and oxygen content, oxidation rate and bacterial populations in unconsolidated materials.

Similarly, the complexities of moisture variations and their effects on diffusion and permeability coefficients, air-filled porosity, and water/air partitioning of Rn in soils, remain to be fully implemented into models incorporating geological, pedological and construction complexities around homes. Comsiderable improvements in prediction, construction and mitigation should be possible with improved understanding of moisture phenomena.

At present the prediction of regional variations in Rn hazard utilize either U values or gamma measurements of ^{214}Bi.

It is clear from our work that Ra and U are not necessarily closely correlated. The research on ^{234}U and ^{230}Th should lead to better predictions of regions and localities where the U vs. Ra correlation is disturbed.

Goal 1 - U-Decay Series Nuclides in Soils

Radium-226, the parent of Rn, is itself a product of alpha decay of ^{230}Th ($T_{1/2}$=80,000 yr), which in turn is the decay product of ^{234}U ($T_{1/2}$=250,000 yr). These half lives are long compared to the rate of soil formation, so chemical redistribution and depletion of ^{230}Th and ^{234}U can markedly disturb the simple correlation of Ra with ^{238}U, the parent of the series. Several researchers (Rosholt et al., 1966; Hanson and Stout, 1968) have shown marked disequilibrium among these nuclides in the U decay series.

In our NY-1 profile developed on granitic rocks in the Adirondack Mts., silt and clay in the B and C horizons have ^{226}Ra/^{238}U activity ratios of 1.15, 1.27, 2.43 and 1.65. Similarly, the Ra/U of total soil in the B horizon in our IL-1 profile is 1.23 to 1.44 (no partial extractions are available for this profile. Both of these profiles are developed on glacial till and loess. We hypothesize that U is rapidly leached from the finely ground till particles, but that Th, being much less soluble, is retained and supports ^{226}Ra for the 10-20 thousand years since these glacial deposits were formed. The strongly weathered NC-1 profile also shows unsupported Ra in the deeper soil.

We are currently developing the analytical ability to determine ^{230}Th and ^{234}U by alpha spectrometry, and will then analyze samples from these three profiles plus a selection from other profiles to test the above origin for the glacial materials and to investigate the origin of other Ra patterns. Samples will be those already collected from the 12 carefully documented profiles. Professor Jester and Graduate Assistant Y.J. Chang will be mainly responsible for the analytical work on this project.

Goal 2 - Air Convection in Soils

Based on the observations of thermally driven air convection in snow (Sturm and Johnson, 1991), in tuff at the Nevada Test site (Weeks, 1987), in karst openings (Gammage et al, 1992) and in mine spoil (Cathles and Apps, 1975; Guo, personal communication), it is clear that this process can occur in nature where a marked difference between temperature of outdoor air and a porous medium is maintained. Treatments dealing with soil concern only horizontal tabular cases and conclude that convection is not reasonable for the temperature gradients and permeabilities of soils. However, the calculated models of Figures 10 to 12 clearly show that convection can occur under slopes, which are common in soils and unconsolidated surficial materials. Similarly, convection may occur around bedrock highs which maintain a different temperature than adjacent soil.

We propose to investigate this in the field and to extend the computer modeling shown above. An initial site in sloping sandy soil near Penn State was rejected for study by the state Game Commission, the owners of the land, but we are now investigating similar sites under other ownership. At this site, we will emplace tubing for obtaining air samples, Alpha-Nuclear Rn detectors for measuring short-term Rn variability, and thermocouples for temperature disturbance. Tracer gas experiments using N_2O or other gases will be conducted during winter (when temperature decreases upward) and summer (when temperature decreases downward). The soil profile will be excavated and described, and permeability measurements made. The results will then be modeled to compare observations with a computer model.

A similar but less detailed study will be conducted at the talus site for which preliminary results are described above.

The computer model will be extended so that we can test other sets of conditions, such as a constant heat-flow lower boundary, inhomogeneous soil, and irrgular lower boundary.

Goal 3 - Studies of homes affected by air convection and soil moisture effects

In order to evaluate the effects of these two variables, two types of studies will be conducted: detailed investigations of homes selected to show significant effects from these processes, and surveys of Rn in groups of homes classified according to these variables.

Specifically, we intend to select one or more houses at the base and/or top of slopes underlain by permeable material and measure Rn variation in the house and the Rn variability, temperature and other variables in soil gas near the house. The data will be analyzed for correlation and compared with models of convection. Similarly, a house in wet soil will be studied to examine Rn flux as compared to houses in soil that is similar but dry. It appears that the Washington house under current study with Don Thomas may meet the criterion of "wet", since holes adjacent to the house seem to accumulate water, and the basement is relatively low in Rn compared to many in the region.

Surveys using alpha-track detectors will be conducted of groups of houses to test for patterns in Rn values correlating with hilltops vs. hill bottoms, and similar effects, in order to investigate the significance of the convection effect in areas where the soil/overburden is reasonably well characterized.

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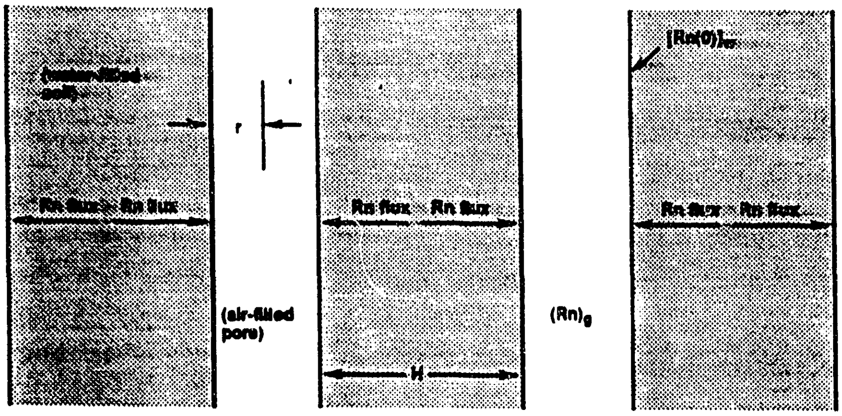


Schematic diagram of two planar air-filled pores separated by water-saturated soil.

$$R_{Rn} = \frac{[Rn]_g}{[Rn]_s}$$

Solution:

$$= \frac{P_t l_w (H + 2r)}{Hr \coth (H/2l_w) + P_t H K_T l_w - 2r K_T l_w (1 - P_t)}$$

l_w = Rn diffusion length in water $=(D/P\lambda)^{0.5}$

P_t = Total porosity

K_T = Rn partition coeff. for water/air (a function of T)

Rn_s = Rn in single-phase pore space at infinite distance from crack

F = Fraction of pores that are water-filled

Q = $1/(F(K_T-1)+1)$ coth = hyperbolic cotangent

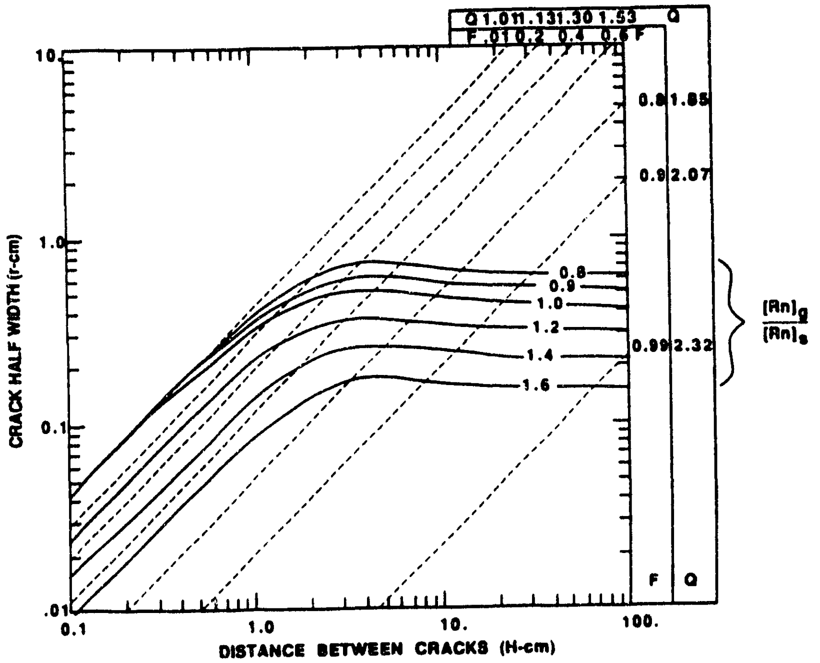
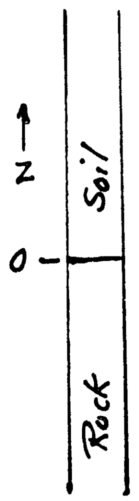


Figure 1. Rn_g/Rn_s (curves labeled 0.8 to 1.6) as a function of crack half-width and distance between cracks, for 3.5°C, P_t=0.47, D_{int}=0.1, as found for site 14-80, B horizon. Larger effects are expected if D and P_t are actually smaller in the water-filled medium, as is very likely. (After Washington and Rose, 1992)

$$(\mathrm{Rn}(z))_r\lambda_{\mathrm{Rn}} = \frac{(\Phi_s - \Phi_r)D_{b,s}l_r}{(D_{b,s}l_r + D_{b,r}l_s)} \exp\left(\frac{z}{l_r}\right) + \Phi_r \qquad (21)$$



$\phi = \frac{\mathrm{Ra}\lambda Ed}{P_t(F(K_T-1)+1)}$

D_b = Radon diffusion coefficient(bulk)

λ = Radium or radon decay const.

l = Rn diffusion length (see Fig. 1)

r,s rock, soil

E = Rn emanation coefficient

Ra = activity of Ra in soil or rock

d = Density of soil or rock

P_t = Total porosity

F = Fraction of pores saturated with water

K_T = Rn partition coeff (water/air)

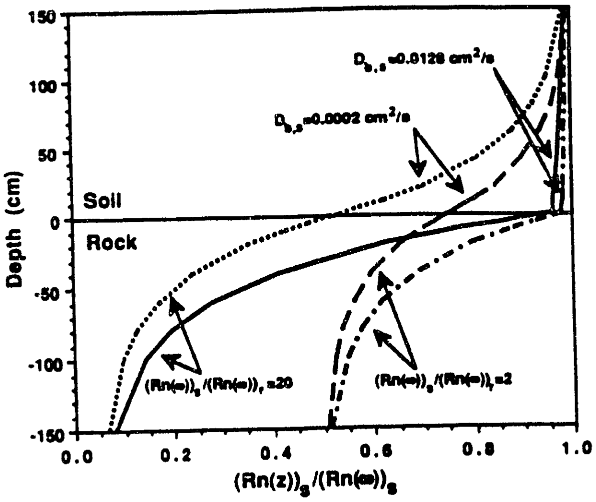


Figure 2. Deviation of Rn_z from Rn at infinite distance from the soil-rock interface, for D_b=0.0002 in the rock and two values of D for soil, differing because of moisture. Under wet conditions where D is similar for the two media, but porosities are very different, the Rn concentration is markedly depleted for 10's of cm from the bedrock. (After Washington and Rose, 1992).

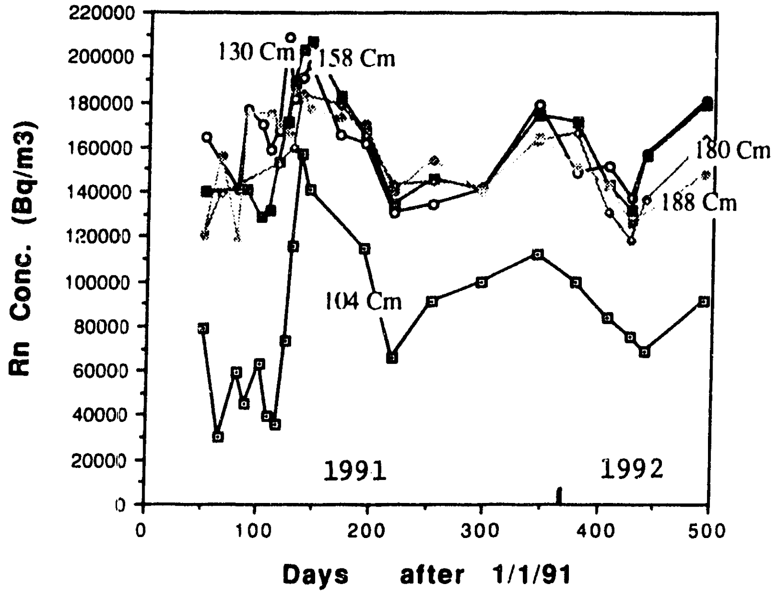


Figure 3. Rn activities vs date at site 14-80 for the period Feb. 1991 to May 1992, for four depths. Samples were extracted from permanently emplaced tubing and measured with a Lucas-cell type of detector. Note the large variability with time, with a seasonal pattern. Summer 1991 was very dry.

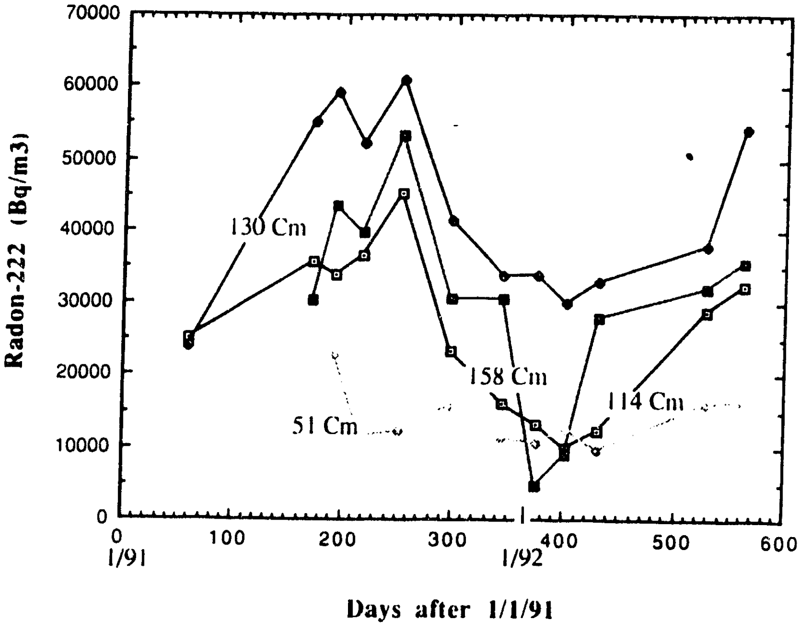


Figure 4. Rn activities vs. date at site 14-83 for the period Feb. 1991 to July 1992, for four depths. Samples collected and analyzed as above. Seasonal variation is very large, as found in 1988.

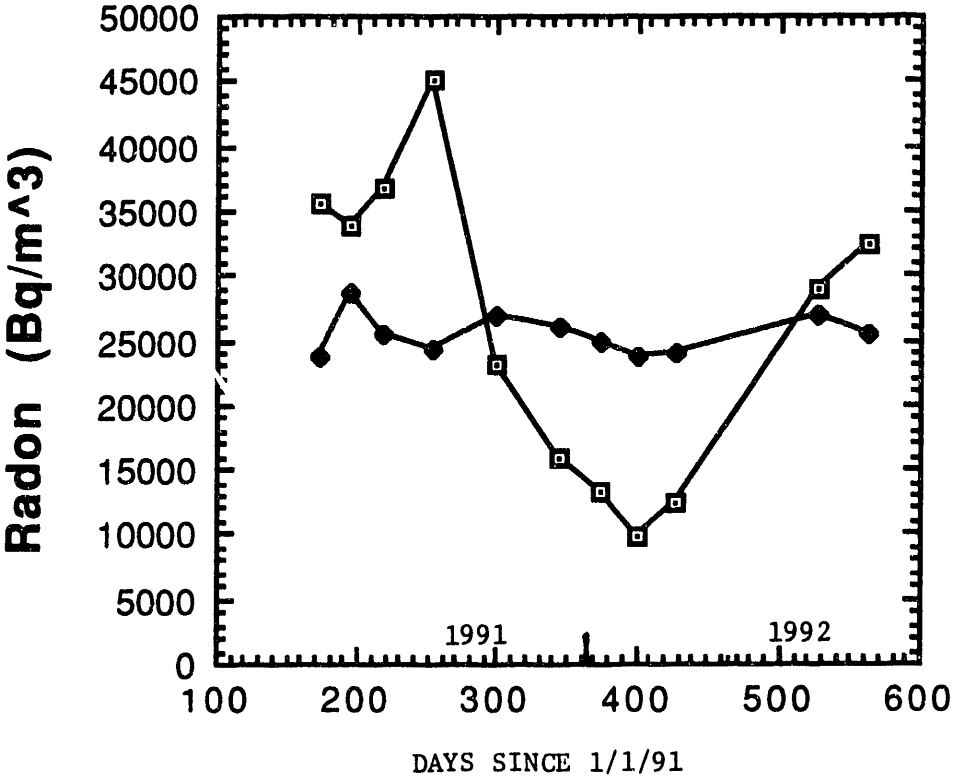


Figure 5. Comparison of observed Rn at site 1483 for 1991-92 at 114 cm depth (open squares) with calculated Rn based on Ra, emanation coefficient, porosity, moisture saturation and temperature (solid diamonds). The seasonal variations are much larger than can be explained by existing theory, and may be caused by air convection or diffusion into rock fragments.

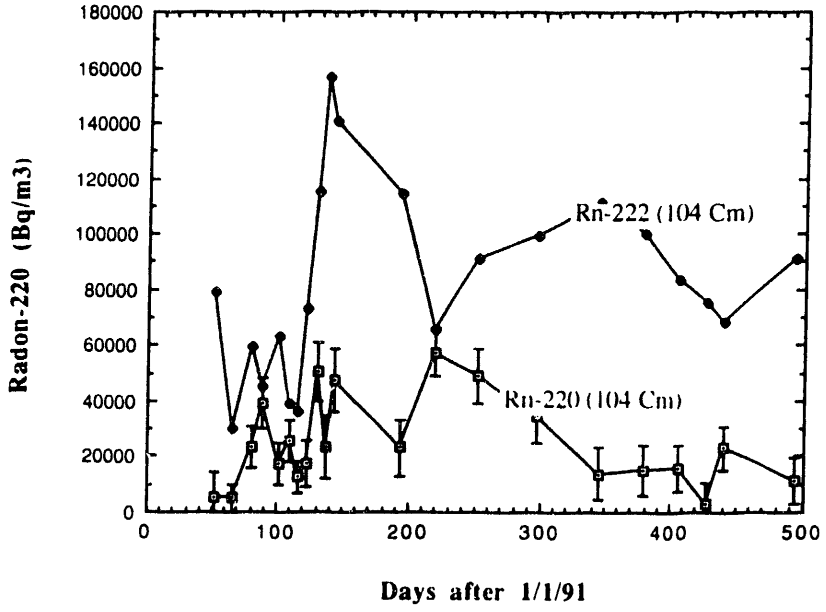


Figure 6. Variation of Rn and Tn (^{220}Rn) at site 14-80 for 1991 and 1992. Note the poor correlation in contrast to some previous data at the site, and the relatively large error for Tn.

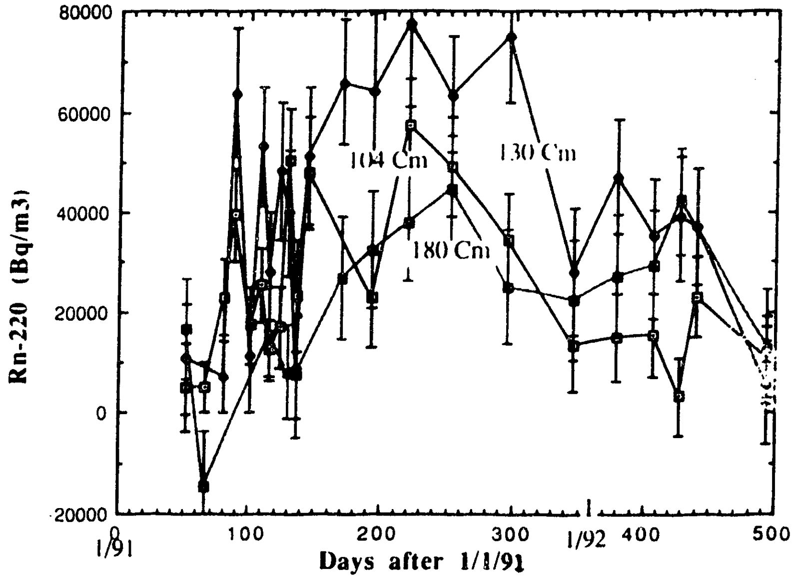


Figure 7. Variation of Tn at site 14-80 for 3 depths. The data suggest a high in summer, but other variations probably represent mainly analytical error.

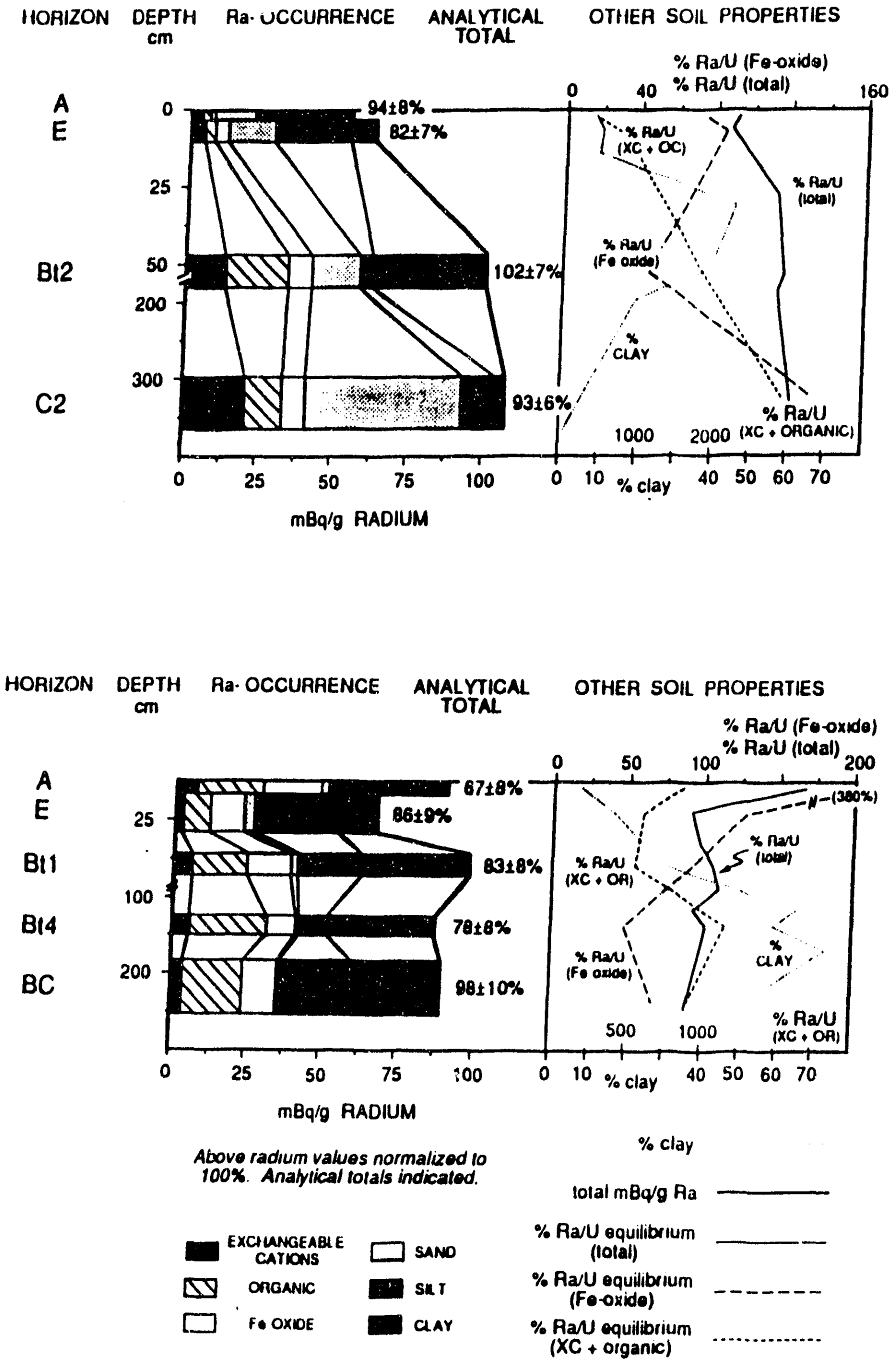
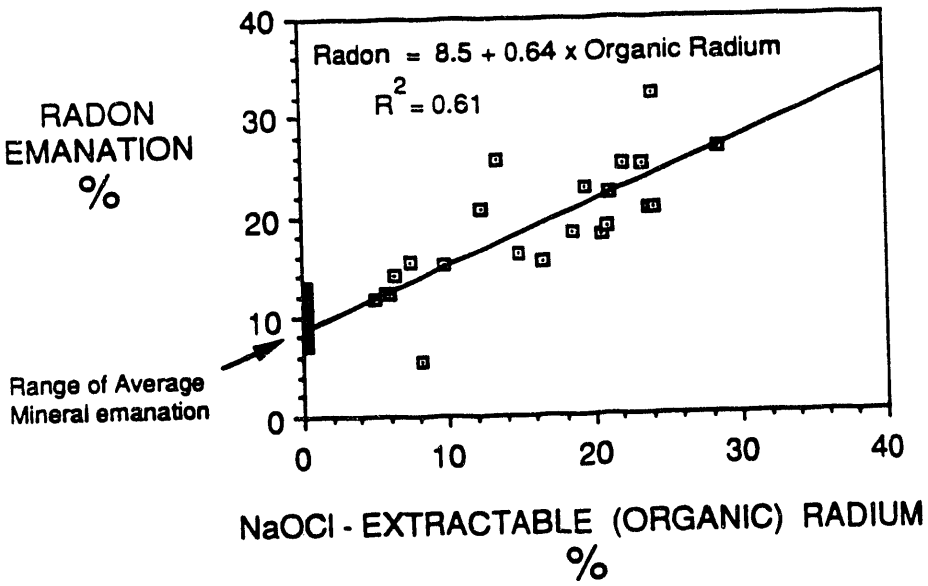


Figure 8. Form of radium and soil properties at sites NC-1 (above) and 14-80 (below). Note 2x scale expansion for top part of depth scale. Abbreviations: XC= exchangeable cations, OR= organics. Data obtained by selective chemical extraction. Note that XC+OR fraction amounts to the majority of pedogenic Ra (OR+XC+Fe-oxide) and that the Ra/U activity ratio is hundreds to thousands.

Radon Emanation vs. Organic Radium



Organic radium with an emanation coefficient of about 64% for seventeen samples of soil from NY, PA, NC, and IL. Intercept of 8.5% Rn emanation (non-organic emanation) is within one standard-deviation of average "mineral" emanation. One-sigma analytical error ±15%.

Figure 9. Radon emanation coefficient vs. "organic radium"

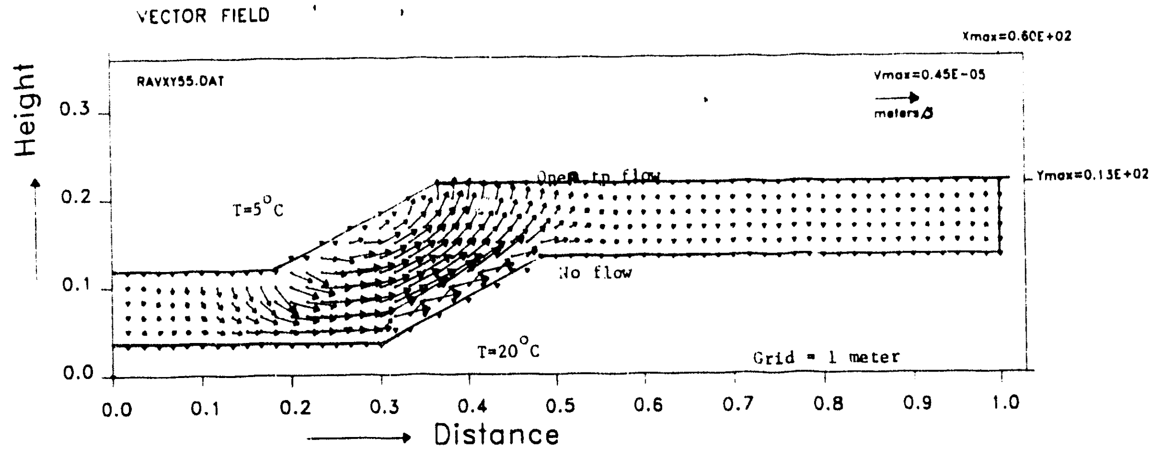


Figure 10. Calculated air convection driven by temperature difference between air and deep soil, for steep slope, with top surface open to flow. Permeability 10^{-7} m^2. Computer model by Weixing Guo.

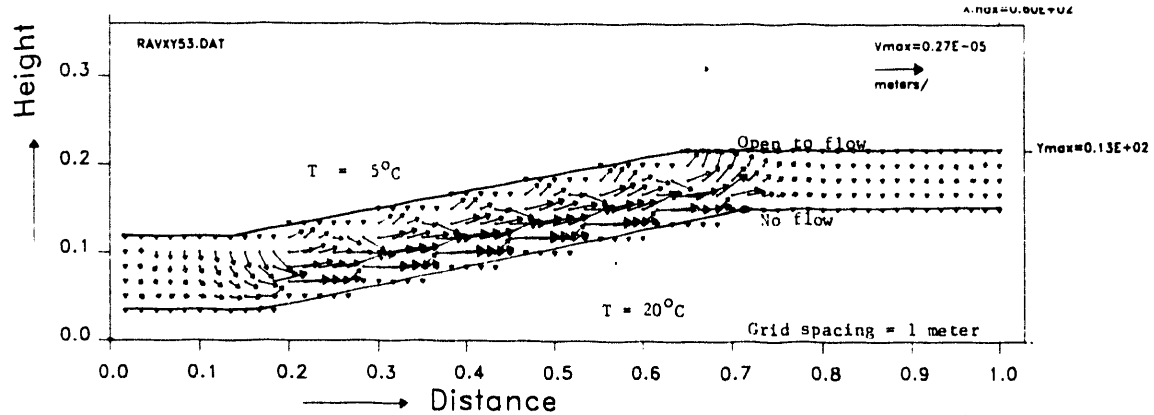


Figure 11. As for Figure 10, except shallowerslope. Steps on the open surface boundary create calculation dispersion, but general flow is up the slope.

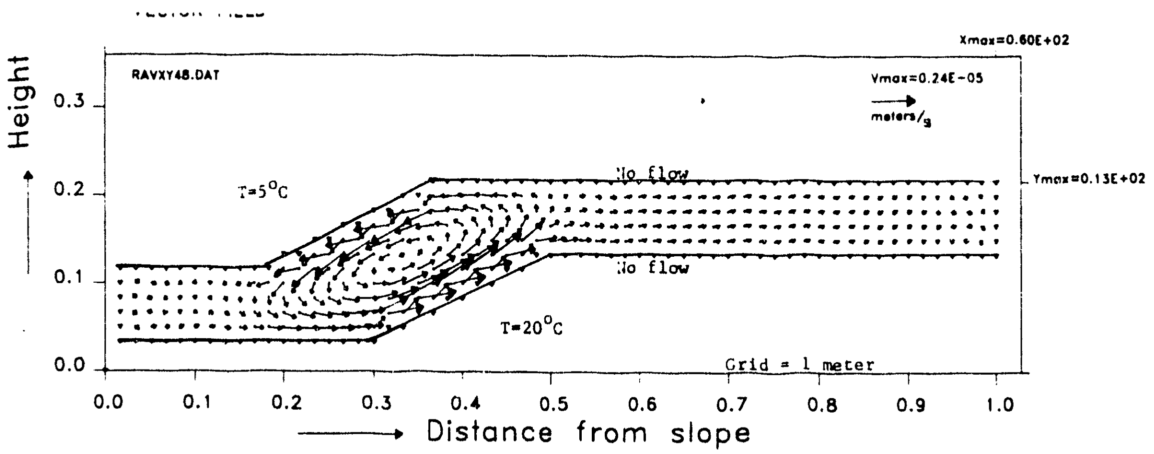


Figure 12. As for Figure 10, except top surface is closed to flow.

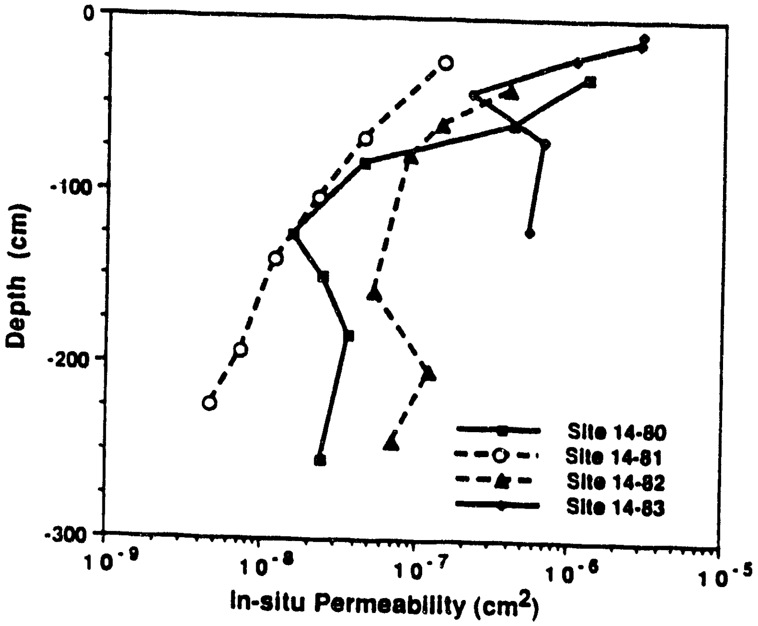


Figure 13. Permeability of soil profiles at four sites in central Pennsylvania, after Washington (1991).

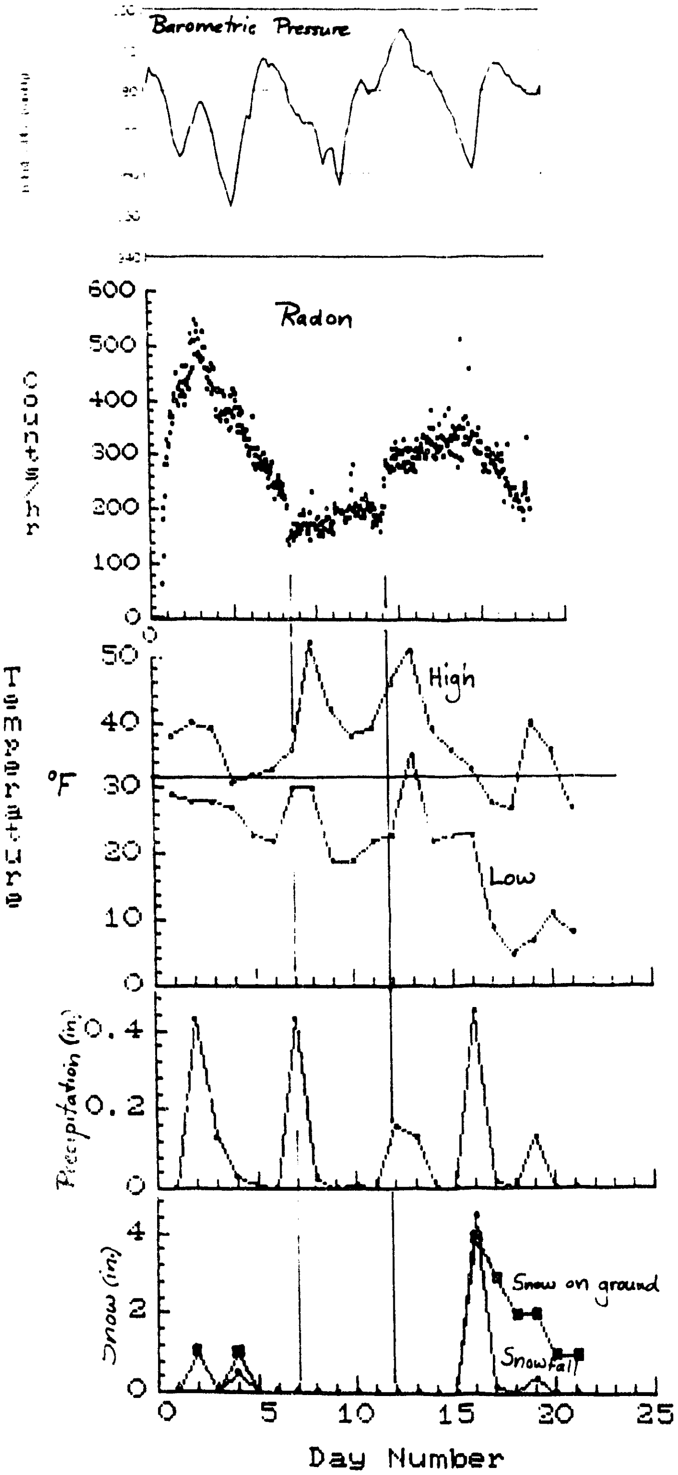


Figure 14. Hourly Rn by Alpha-Nuclear detector vs. time for the period Dec. 12, 1990 to Jan. 3, 1991 at site 14-80, 60 cm depth. The initial Rn increase represents growth of daughters, but other changes represent variation of soil-gas Rn. The vertical lines at 7 and 12 days mark abrupt changes in Rn values, which appear to correlate with precipitation and snow melting. Essentially no barometric or diurnal effect is evident.

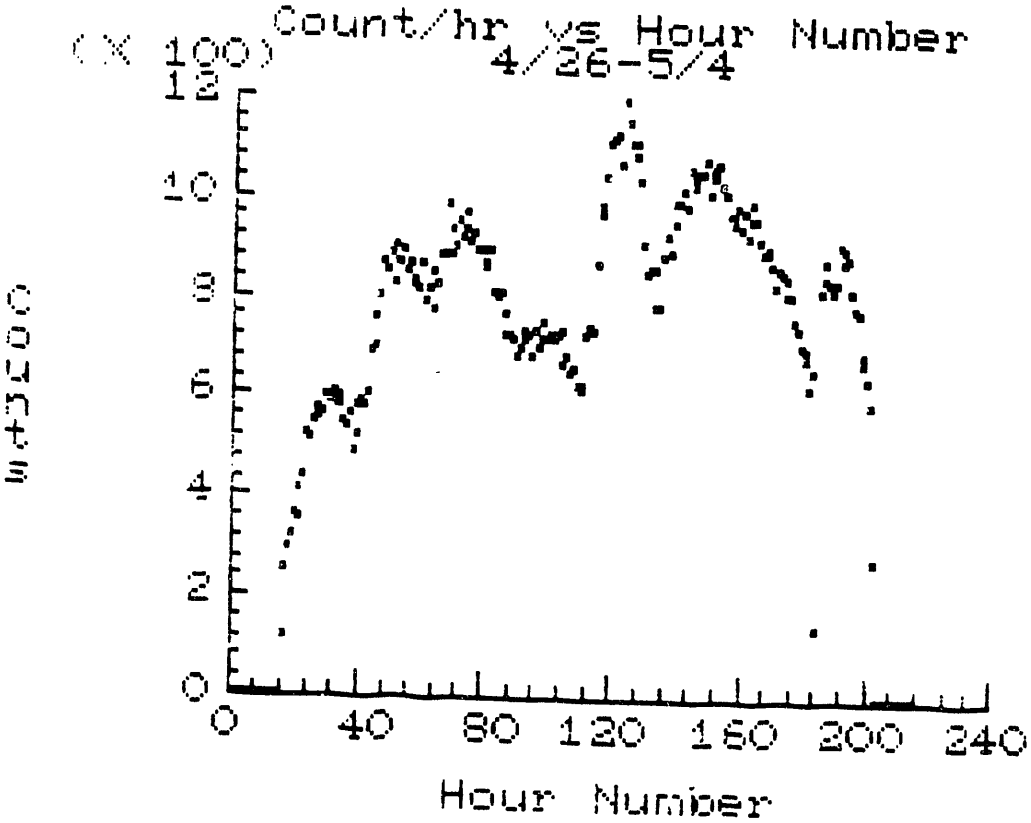


Figure 15. Rn values (cts/hr) for the period Apr. 26-May 5, 1991 at site 14-80, 60 cm depth, from Alpha Nuclear detector. The initialincrease in the first 3 days is due to Rn daughter equilibration, but other variations, including diurnal effects, represent changes in soil-gas Rn. The diurnal pattern shows peaks in early morning and lows near midday. The variation is believed to result from changes in moisture content and the resulting effects on diffusion coefficient and air-water partitioning.

END

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