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GENERATION AND MOBILITY OF RADON IN SOILS
FINAL REPORT
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GENERATION AND MOBILITY OF RADON IN SOILS
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This report emphasizes research on the above project (DOE Research Grant DE-FG02-87ER60577) since 1993, but includes some description of previous work which has been discussed in prior reports and publications.

Objectives of the Research

The research has the objectives of answering the following questions:

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

Research Accomplishments

Objective 1. Radon Emanation Coefficients

The emanation coefficient is the proportion of radon, produced by radium decay, that escapes a solid grain or material into air-filled pore space. In a paper by Greeman and Rose (1996), the average emanation coefficients for 68 disaggregated soil samples were shown to be 0.20 for ^{222}Rn and 0.16 for ^{220}Rn . Correlations with the content of radium in the exchangeable, organic, Fe-oxide, clay, silt and sand fractions of the soil show that the organic-exchangeable fraction has a ^{222}Rn emanation coefficient of 0.46, whereas the coefficient for the silt-clay fraction is only 0.22, and for the sand it is lower. This difference appears to result from the open structure of the organic material, and its location on the surface of grains, thereby allowing easier escape of the newly generated Rn atom. The study also showed that Ra is enriched in soil organic matter by plant uptake. Thus, well developed soil profiles with abundant vegetation are expected to have higher radon in their soil gas than young soils or those lacking vegetation.

Objective 2. Form and behavior of ^{234}U , ^{230}Th and ^{226}Ra

An intensive study of these U-series decay products in two soil profiles has recently been completed by Graduate Assistant John Hogue (1996). The accomplishments and conclusions of this investigation are summarized as follows, and a condensed version of his thesis is attached as Appendix B:

1. The analytical methods for accurately determining these nuclides in soil and in soil fractions have been improved in order to obtain reliable results on soils.

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2. The two soil profiles studied are NY-1, a typical alfisol developed on Pleistocene till in the Adirondak Mts. of New York, and IL-1, a typical mollisol developed on till and loess at Champaign, IL. Earlier research by Greeman (1992) on radium in these soil profiles had indicated considerable disequilibrium between ^{238}U and ^{226}Ra .

3. Disequilibrium is found to be widespread in the decay series ^{238}U - ^{234}U - ^{230}Th - ^{226}Ra in both soils. For 9 total samples, the $^{230}\text{Th}/^{234}\text{U}$ activity ratio ranges from 0.93 to 1.68, the $^{226}\text{Ra}/^{230}\text{Th}$ ratio from 0.63 to 2.1, and the $^{238}\text{U}/^{234}\text{U}$ ratio from 0.88 to 1.12.

4. Analyses of the nuclides in the selective fractions exchangeable, organic, Fe-oxides, clay, silt and sand showed even greater disequilibrium. In the pedogenic fractions (exch., organic, Fe-oxide, B-horizon clays), the $^{230}\text{Th}/^{234}\text{U}$ ratio ranges from 0.09 to 0.52 in organic matter, and 0.45 to 4.2 in Fe-oxides. The $^{226}\text{Ra}/^{230}\text{Th}$ ratio is as high as 23 in organic matter. These data indicate that the different nuclides are being separated according to their chemical properties and in response to lattice damage from alpha decay.

5. In both profiles, ^{234}U is leached preferentially to ^{238}U from the residual sand, silt and clay fractions, as a result of radiation damage. In contrast, pedogenic phases formed from soil solutions are enriched in ^{234}U .

6. In both profiles, ^{230}Th activity exceeds ^{234}U activity, as a result of preferential leaching of uranium. This disequilibrium is much more pronounced in IL-1 because of its higher percentage of Fe-oxide and clay, combined with a higher affinity of these phases for Th.

7. At NY-1, preferential uptake and cycling of ^{226}Ra by trees and plants has enriched Ra in surface horizons and depleted it from deeper horizons. In contrast, the grasses at IL-1 have extracted and enriched Ra from the same organic-rich horizons, and little enrichment/depletion is found. Because the deeper soil is not depleted in Ra, and because a considerable fraction of Ra is bound in organic matter, the Rn concentration is enhanced.

8. Iron oxides are a very important host for Th and important for U, but relatively unimportant for Ra.

9. Because of the extensive and variable disequilibrium in the U-decay series in soils, the abundance of Ra is a much better indicator of Rn hazard than is the abundance of U.

Objective 3. Thermal Convection of Soil Air

Since 1993, various experiments and computer modeling have been done to study thermally driven air convection. The field experiments include long-term temperature measurements, tracer experiments and other measurements, such as soil air flow direction and velocity. In addition, computer modeling and lab experiments have investigated the process.

Field experiments

Most of the field experiments were conducted at the Shadow Lane site which is located on a 15 degree slope about

15 m long next to a small valley (Figure 1). The soil is about 1 to 1.5 m thick, and very sandy, developed by weathering of sandy dolomite. The results are discussed as follows.

1. Long-term temperature and radon measurements

The temperature was measured at 15-minute intervals by thermistors and thermocouples at 3 depths (0.3 to 1.4 m) at locations at the base, mid-slope and top of slope, using a datalogger. Radon was measured continuously using an Alpha-Nuclear radon detector at the top of the slope.

Figures 2 to 5 illustrate some of the variations in temperature and radon observed at this site. Figure 2 shows temperature at the hilltop site for Dec. 6 to 10, 1993. Air temperature is -9 to +6C. Temperature at 0.3 m depth shows a relatively smooth pattern at about 5C. Temperature at 0.75 m and 1.25 m ranges from 7 to 9.5C (i.e., warmer than air temperature by 1 to 18C) and shows several abrupt perturbations coinciding with the coldest periods. This behavior is consistent with the type of variation expected for thermal convection. The convection process is apparently episodic, occurring during the periods of extreme temperature difference. Radon variations are illustrated in Figure 3; only very small perturbations coincide with the temperature spikes in this time period.

For contrast, figures 4 and 5 show temperatures and radon for July 20 to 24, 1993. Outdoor temperature is 12 to 27C vs. 14 to 17C in the soil, so that soil is commonly cooler than air. Slight diurnal temperature effects are seen in the soil. Figure 5 shows radon values for the same period. Radon counts range from about 80 to 115 cts/15 min., with a distinct peak in the late afternoon for July 20 to 22 and perhaps on the other days. The temperature patterns are distinctly different from the winter patterns, though appreciable effects are evident for radon.

Figure 6 shows temperature data (6 hr. averages) for the site. At a depth of 1 m at the middle of the slope, temperature shows a relatively simple yearly cycle, from a maximum of about 15C in summer to about 2C in winter. Outdoor air temperature ranges from maximums of about 25C to minimums of about -15C. Temperature differences of 10 to 20°C between outdoor air and soil at 1 m are not uncommon in winter and in summer. The median difference in January is 14C, and the maximum is 26C. Based on our modeling discussed below, these temperature differences are adequate to drive appreciable convection of soil air.

Temperatures at 1.3 m depth at the top of the slope are more complex, showing common episodic deviations of a degree or so from a steady value slightly higher than that beneath the slope. A thermocouple sensor emplaced in January 1994 about 1 m from the thermistor shows much smaller temperature fluctuations might be a measurement artifact, but it seems possible that these differences result from circulation of warmer air during thermal convection episodes.

2. Tracer experiments

In the spring of 1994, two tracer experiments were conducted at the Shadow Lane site to search for convective transport. In these experiments, about 1 liter of methane (CH_4) was injected into a probe in the mid-slope position. Double sampling probes of the type designed by Reimer (xxxx) were emplaced at about 20, 50 and 100 cm up and down slope from the injection site, and about 20 cm parallel to the slope. Soil air was pumped out of one probe of a pair, through a methane detector, and back into the other probe, so as to cause minimal disturbance of the soil air.

Figure 7 shows the results of 2 experiments. Unfortunately, the presence of about 1 m of snow at the site during the winter of 1994 made the experiment impossible to conduct during the coldest weather in January and February. This snow was still present on 3/16/94 when the first experiment was conducted, with a temperature difference of about 4.5C between air and soil. Based on the approximately symmetrical nature of the patterns, diffusion was the major transport process at this time. However, the data suggest the CH_4 values 5 hours after injection are slightly higher upslope from the injection point than downslope. Since the meter reads only to $\pm 0.1\%$, the result is poorly defined. Suggestive results were also obtained for shorter times. A similar test on 4/8/94 showed no evidence for an asymmetrical pattern. Comparison of the results with our computer models indicates that a difference of the magnitude suggested for 3/16 is in the range expected for thermally driven convective transport.

Two additional tracer experiments were conducted at the Shadow Lane site in 1995. For the tracer experiments, 16 pairs of soil gas probes were inserted in the soil as shown in Figure 8. In each pair, an inlet and outlet port were spaced about 5 cm apart so that the soil air would be disturbed only very locally by the sampling process. An HNU VOC analyzer was used to measure gas compositions.

The second experiment produced more useful data so will be discussed here. In this second experiment, about 40 ml of benzene was injected at 8:30 AM on July 11, 1995 at the point indicated as the source. After 5 hours, the concentration of benzene vapor was approximately symmetrically distributed around the source, suggesting that diffusion was the main transport mechanism (Figure 9). During this period, air temperature was about 10C higher than soil temperature. The pattern was similar at 6:25 PM. however, at 9:25 AM the following day, higher concentrations were upslope from the source (Figure 10). This data suggests that up-slope convection of soil air occurred during the night. At later times, concentrations suggest that downslope transport also occurred (Figure 11). The downslope transport may result from downslope convection of soil air during periods of appreciable temperature gradient, and/or from downslope flow of dense benzene-bearing vapor. These results, in combination with our previous field observations, indicate that processes other than diffusion can cause transport in soil gas.

3. Other measurements

In 1994, the soil air flow direction and velocity was measured at the Colerain site near Spruce Creek, PA, a steep (45°) slope about 200 m long covered with at least several feet of sandstone talus blocks ranging from 10 cm to 2 m in size (Figure 12). The talus obviously has very high air permeability. In the middle of the slope, the talus is covered by trees and thin soil, but the base is largely barren. During summer and fall, air is observed to flow out of openings between blocks at the base of the slope. Velocities were measured to be about 0.5 m/s on October 6 when outdoor air temperature was 13C and air flowing out of talus was 5C. Similar results were obtained on an earlier visit in September and later visit in winter. Air flow using the direction of smoke flow was observed to be out of most of the lower section of the talus slope.

Numerical simulation

The numerical simulation is conducted by using the STOMP simulator (Lenhard et al., 1995; White et al., 1992) with some modifications. Water, air flow and transport as well as energy and radon transports are considered. Up to now, several cases have been explored.

In the following cases, the simulation domain is a sloping layer (grade = 1) whose porosity is 0.3 and permeability 10^{-6} cm² (coarse sand). The radon generation rate in each cell (1 m by 1 m) is 2.78×10^{-2} atoms/cm³-s (0.75 pCi/cm³), equivalent to 50 mBq/g Ra content of the soil and a 0.3 emanation coefficient. In the figures, a boundary open to diffusion and flow is represented by dotted or dashed lines and a closed boundary by solid lines.

Case 1: The upper boundary is open over part of the base and top of the slope. All other boundaries are closed. The temperature is 5C on the upper boundary and 10C on the lower boundary (Figures 13 and 14).

In this case, soil air flow driven by the thermal gradient is up the slope. Cold air flows into the soil at the foot of the slope, is warmed, and flows out at the top of the slope. The highest soil air velocity is in the lower part of the slope and close to the hotter boundary. Close to the upper boundary, the soil velocity is very low. The maximum velocity is 5.27 cm/day.

Figure 14 shows the contours of soil gas radon concentration for this case. The highest radon concentration is located mainly in the slope. Toward the open boundaries, the radon concentration decreases because of diffusive loss. Compared with figure 15, with no temperature gradient and no soil air convection, the radon contours move upslope due to thermally driven soil air convection by a maximum of about 1.5 m.

Case 2: With all other conditions the same as case 1, the temperature is 10C on the upper boundary and 5C on the lower boundary (i.e., the temperature gradient is reversed). See figures 16 and 17.

In this case, the warm air flows into the soil at the top of the slope. The air becomes colder and flows out at the

foot of the slope. The highest soil air velocity is in the lower part of the slope and close to the colder boundary. Close to the upper boundary, the soil air velocity is very low. The maximum velocity is 5.80 cm/day.

Figure 18 shows the contours of soil gas radon concentration for this case. Similarly to Case 1, the highest radon concentration is located mainly on the slope. Toward the open boundaries, the radon concentration decreases. Compared with figure 15 (no temperature gradient and no air convection), the radon contours move downslope due to thermally driven soil air convection by a maximum of about 1.5 m.

From the above two cases, the existence of thermally driven air convection is clear, as is the fact that soil air may flow either up or down the slope depending on the direction of the thermal gradient. The velocities are low, but additional calculations show that the velocity increases approximately proportional to temperature gradient and to permeability.

Case 3: The upper boundary is completely open and all other boundaries are closed. The temperature is 5C on the upper boundary and 10C at the lower boundary. See figures 18 and 19.

In this case, the cold air flows into the soil in the lower half of the slope and flows out from the upper half of the slope. The soil air velocity close to the upper boundary is much higher than in either case 1 or case 2. The maximum soil air velocity is 4.97 cm/day.

The pattern of radon contours in this case is much different from those in case 1 and case 2 because of the different boundary condition. The radon concentration becomes higher with increasing depth. Along the slope, high radon values are found only at depth.

Compared with figure 20 (no temperature gradient and no soil air convection), the longest distance the radon contours move upslope due to thermally driven soil air convection is about 1.5 m.

Case 4: With all the other boundary conditions the same as case 3, except that the temperature is 10C on the upper boundary and 5C on the lower boundary (i.e., reversed gradient). See figures 21 and 22.

The soil air flow direction is opposite to that in case 3. The maximum soil air velocity is 5.25 cm/day. The pattern of the radon contours is similar to that in case 3. However, the high radon concentrations are displaced downward along the lower boundary by about 3 meters in this case compared with case 3. Compared with figure 20 (no temperature gradient and no soil air convection), the longest distance the radon contours move downslope due to thermally driven soil air convection is about 1.5 m.

This simulation result demonstrates one possible explanation for the observation of outward flow from the base of the slope at the Colerain site near Spruce Creek, PA (figure 12).

Case 5: With all the other boundaries the same as case 3, a house is put at the position: $x = 27 - 35$ m, $y = 17$ m. The room temperature is assumed to be 20C. See figures 23 and 24.

Compared with case 3, it can be seen that much higher soil air velocities are located near the house. The maximum soil air velocity is 14.11 cm/day. The pattern of the radon contours near the house is much different from that in case 3. This is caused by the higher house temperature. The radon transport to the house is markedly increased by the thermally driven soil air convection near the house. The radon flux entering the house is increased by the interactions of flow up the slope and flow because of the warm house.

Another interesting effect that has been modeled in a preliminary way is the effect of soil moisture on air flow, which was not considered in the above calculations. Moist air has lower density than dry air. Therefore, if outside air is dry and soil air is moist (as it usually is), then air will tend to flow up-slope. This effect will be added or subtracted from the simple temperature effect..

Conclusions

Based on the results to date, in combination with the modeling results, thermal convection clearly occurs in highly permeable geologic materials. Radon can be moved a measurable distance by the thermally driven soil air convection (up to 1.5m when thermal gradient is 5C), and much greater distances with the temperature differences of up to 25C found in the field measurements. A higher permeability, such as that in the talus at Colerain Park, also results in much higher flow rates. Radon flux into a house can be considerably increased due to local variation of temperature.

The soil air flow direction can be changed due to the variation of the direction of thermal gradient. Houses at the top of hills are expected to experience increased radon flux in winter, and those at the bottom of slopes would have higher flux in summer. Houses on slopes would experience increased flux from convection whenever the soil and air temperature differ appreciably. However, a high permeability soil is required for the effect to be important.

Future Plans

Graduate student Yingjie Guo is continuing his thesis project of computer modeling the behavior of soil gas on slopes, field experiments, and lab experiments. We expect him to finish his thesis in late 1997 or early 1998. This work should lead to one or more publications.

By the end of 1997 we expect to analyse another soil profile for ^{234}U and ^{230}Th . This data will be added to the manuscript in appendix B and submitted to a journal.

A manuscript is nearly ready for submission on radium in the soil profiles, by Greeman et al. (See Appendix A).

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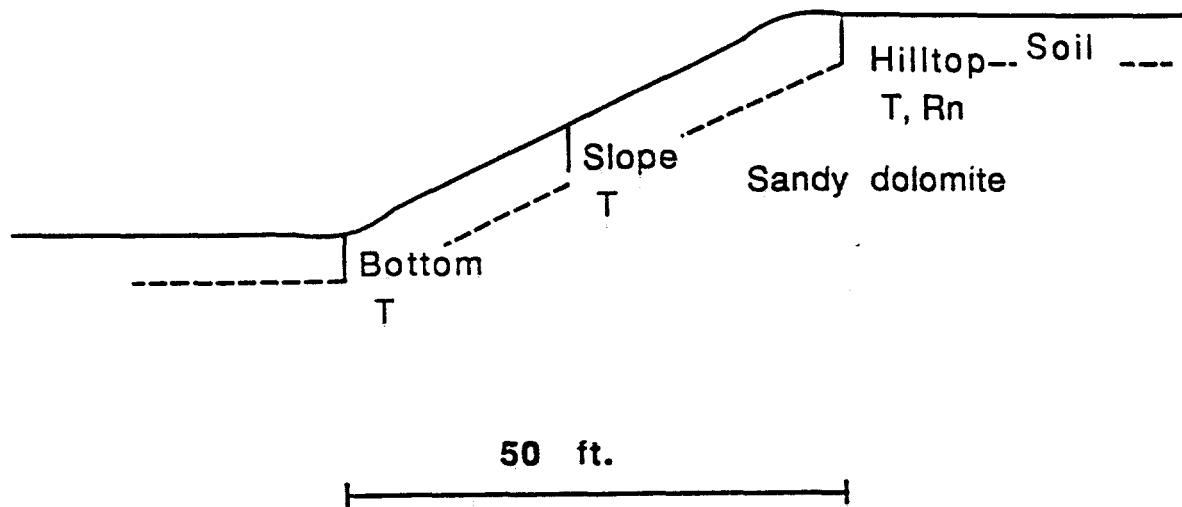
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Shadow Lane Site - Cross-section

Soil: very sandy



Experiments (Shadow Lane and Talus sites):

Temperature

Radon

Moisture

Tracer

Permeability/Diffusion

Figure 1. Sketch of the Shadow Lane site, showing location of measurements and types of measurements.

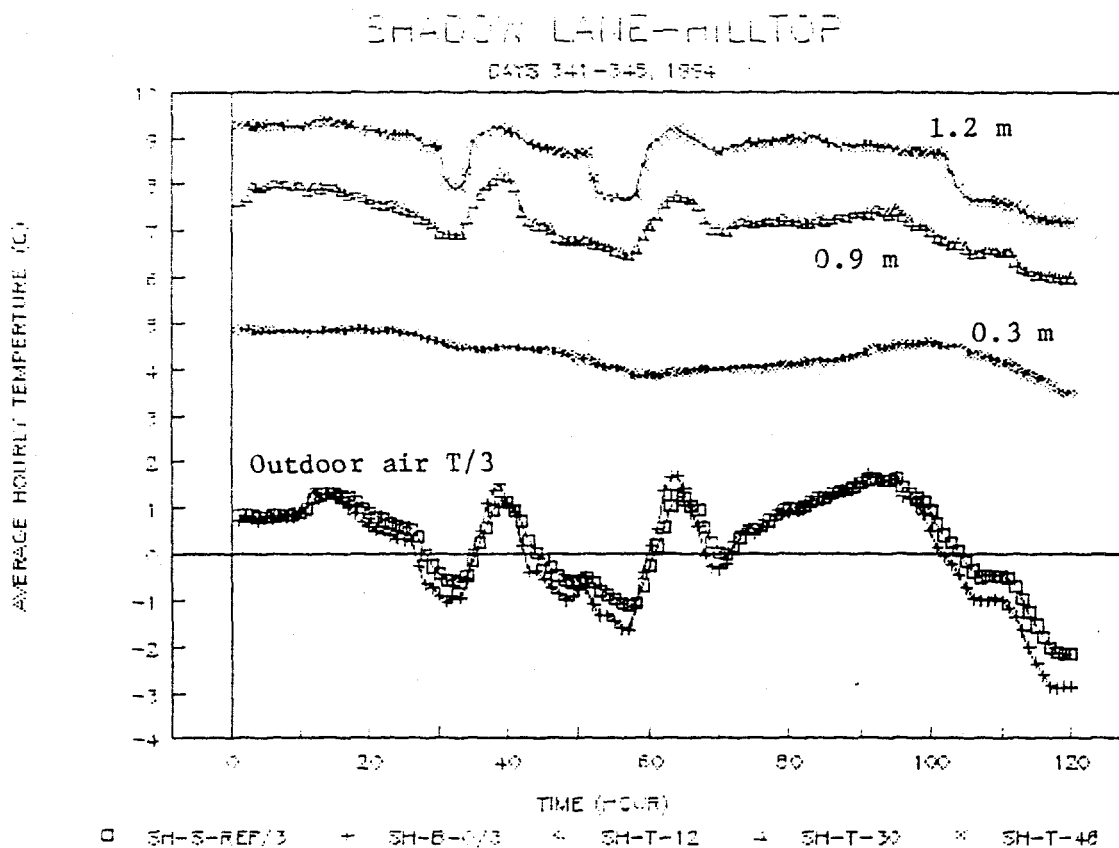


Figure 2. Soil and air temperature at the Shadow Lane site for the period Dec. 6 to 10, 1993. The air temperature value is divided by 3.

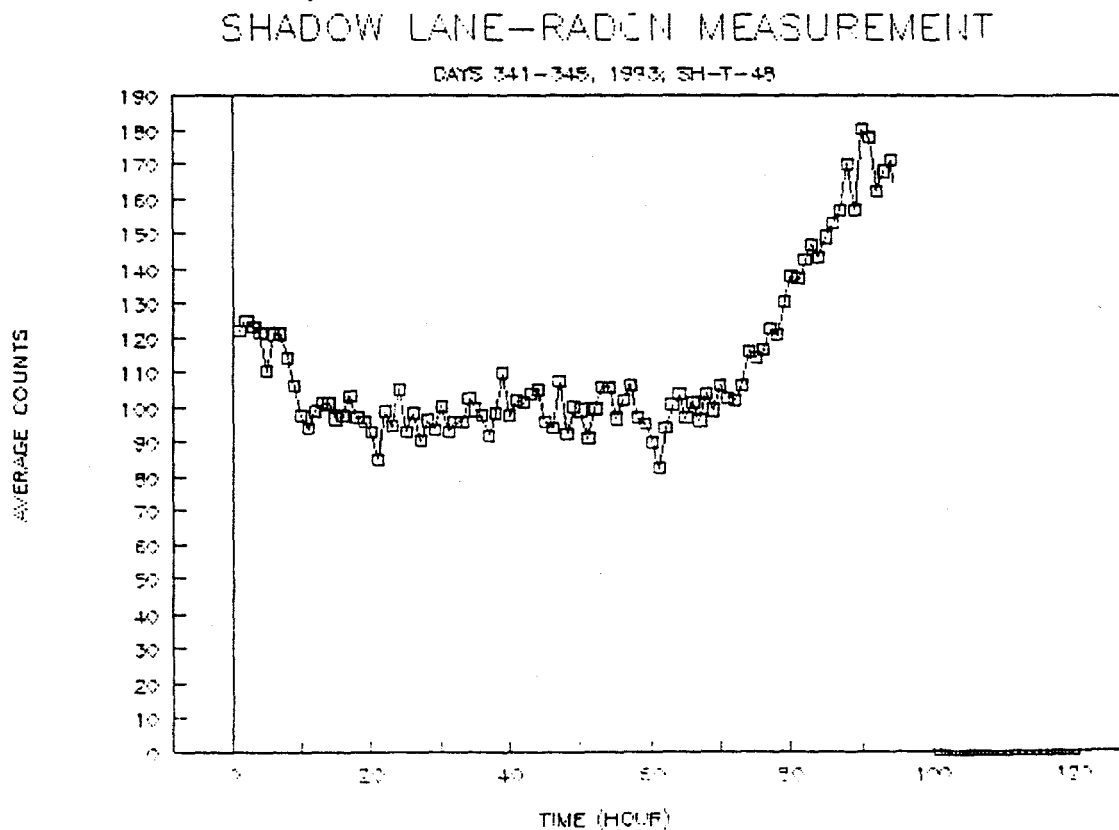


Figure 3. Radon at the Shadow Lane site, 1 m depth at top of hill, for Dec. 6 to 10, 1993.

SHADOW LANE - HILLTOP

DAYS 201-205, 1993

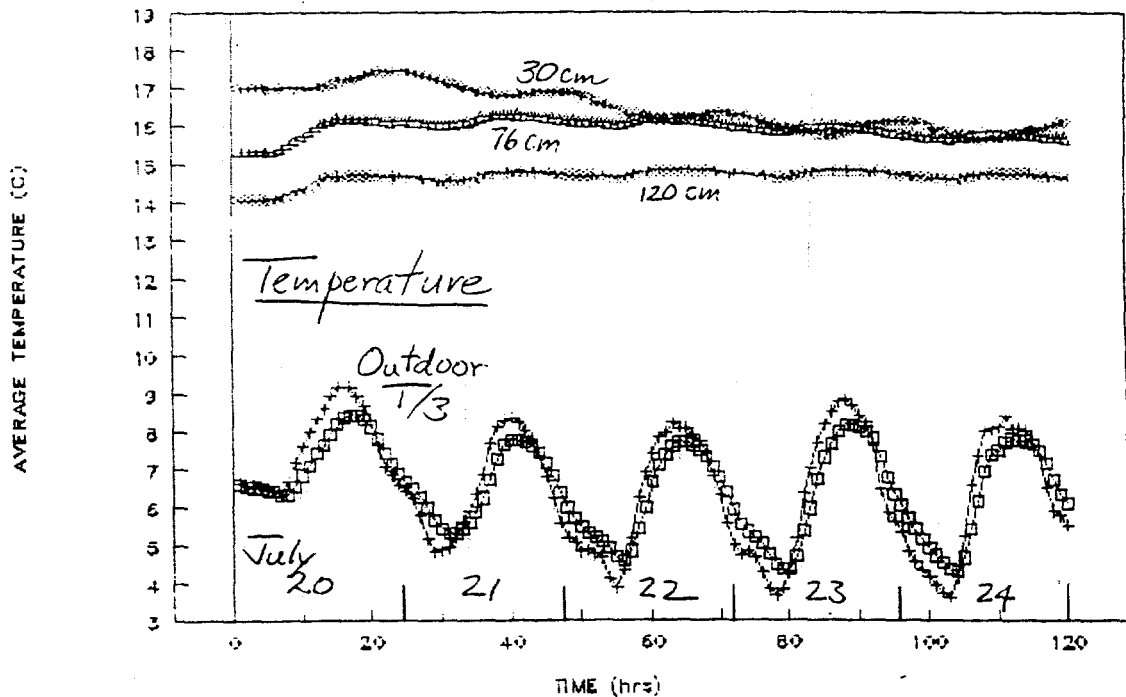


Figure 4. Soil and air temperature at the Shadow Lane site for the period July 20 to 24, 1993. Air temperature is divided by 3.

SHADOW LANE - RADON MEASUREMENTS

DAYS 201-205, 1993; SH-T-48

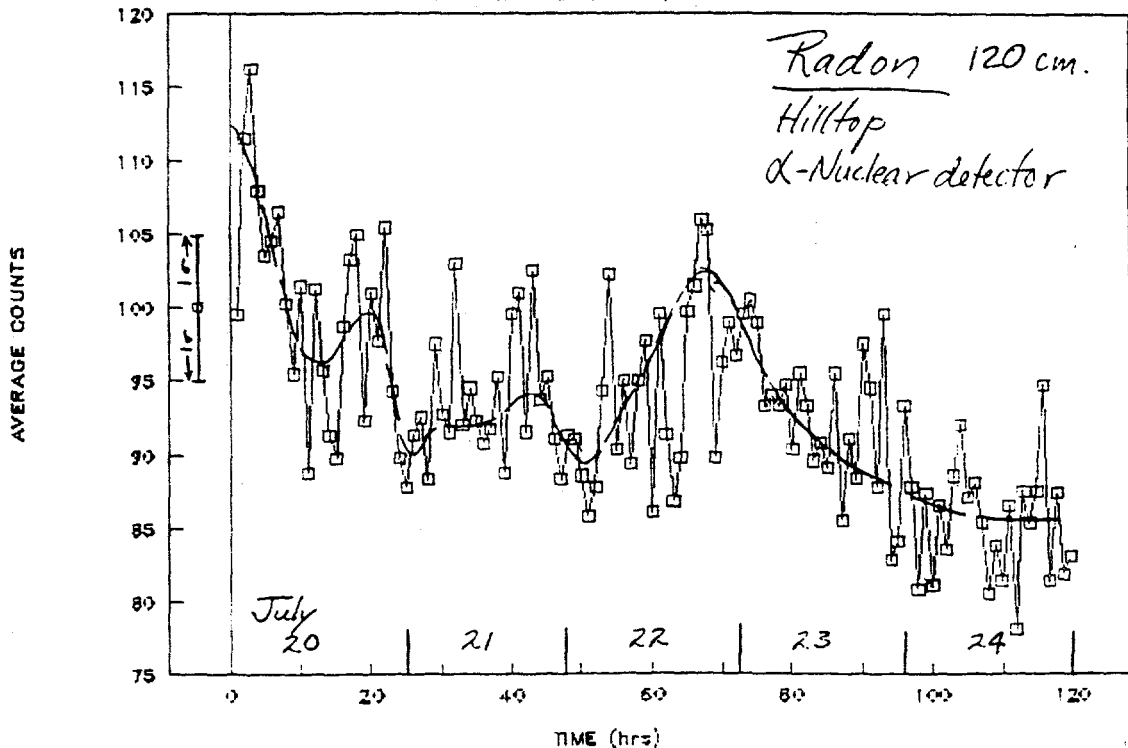


Figure 5. Radon values (counts/15 min.) for the Shadow Lane site,

Annual Variation of Temperature in Shadowlane

Days 84-462

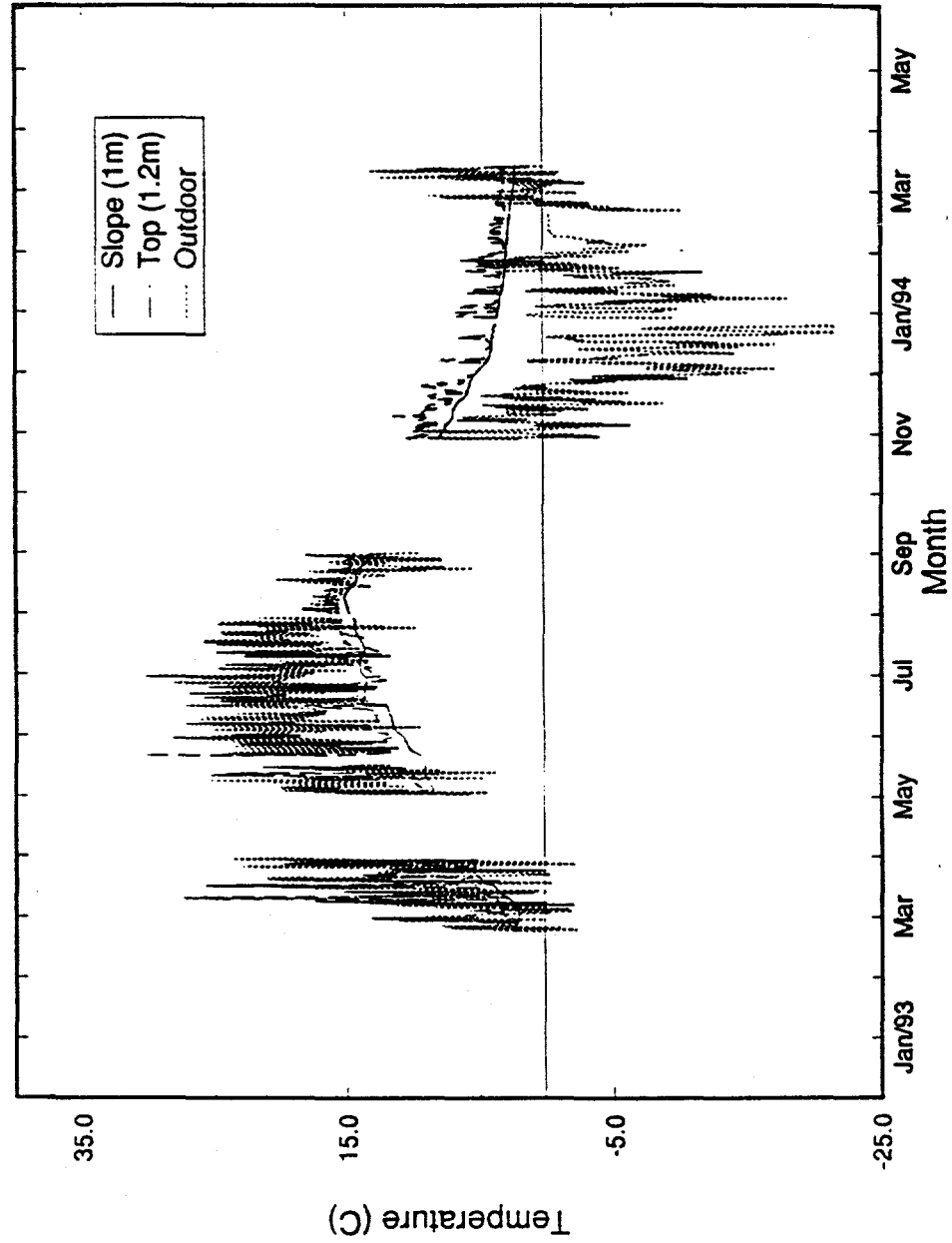


Figure 6. Temperatures at the Shadow Lane site, as recorded by thermistors at 1 m depth on the slope position, and 1.2 m depth in the top position, and at the surface, for the period March 1993 to March 1994. Values are 6-hour averages of 15 minute readings.

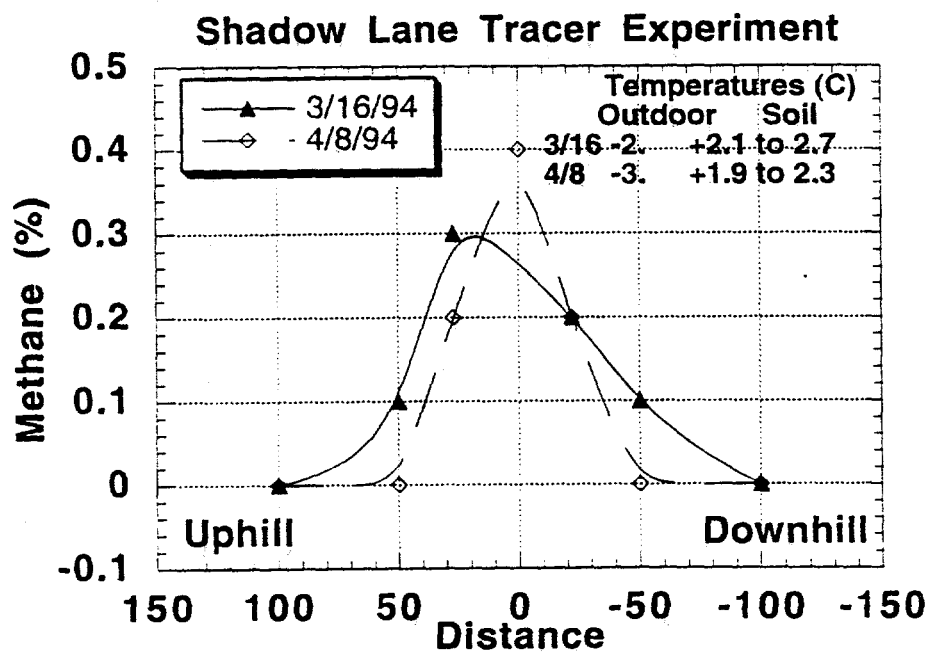
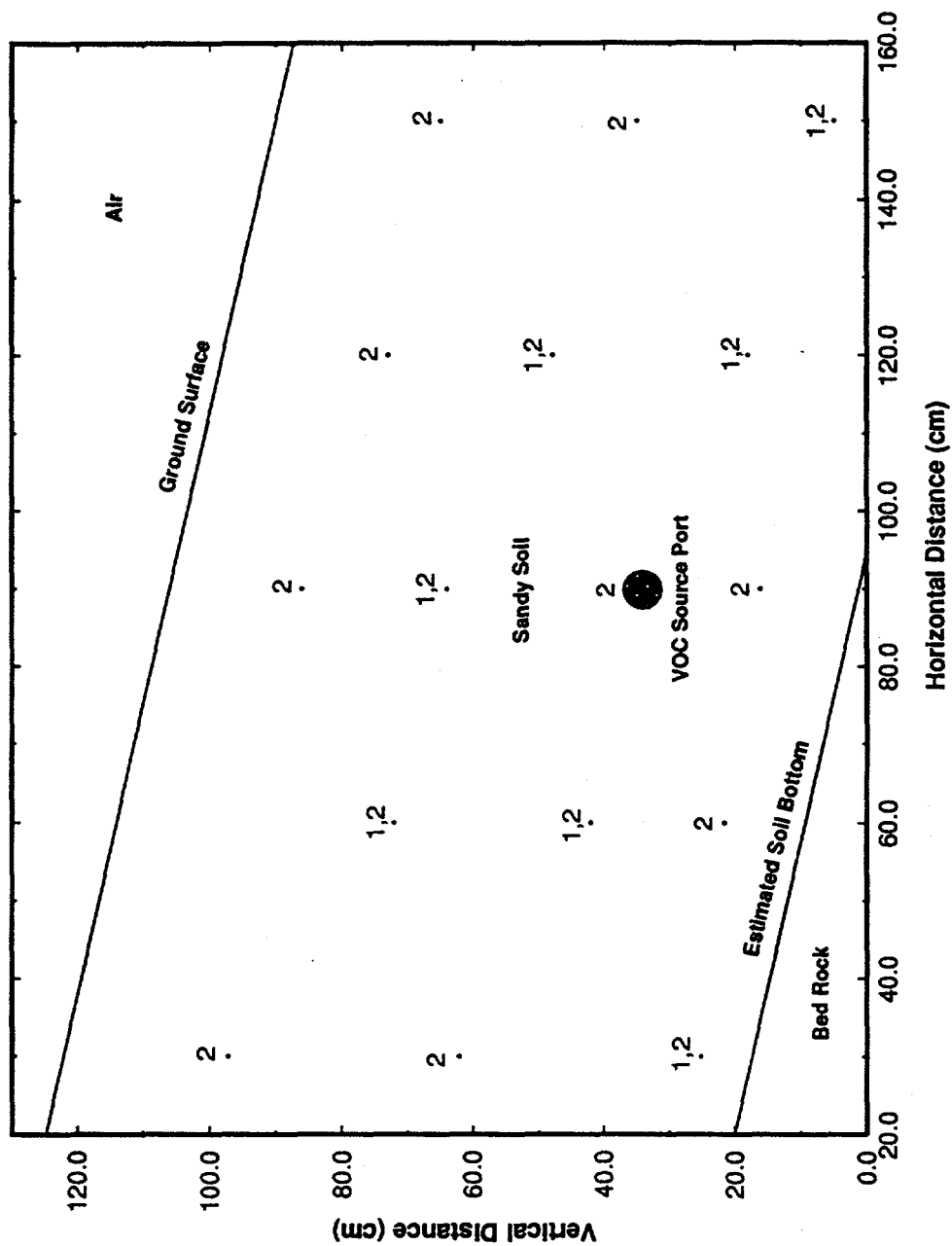


Figure 7. Results of tracer experiments on 3/16/94 and 4/8/94 at the slope location at Shadow Lane, using methane. The data were taken 5 hours after injection at a depth of 1 m at the distances shown from the injection point. Prevailing open air and soil temperatures are shown on the diagram.

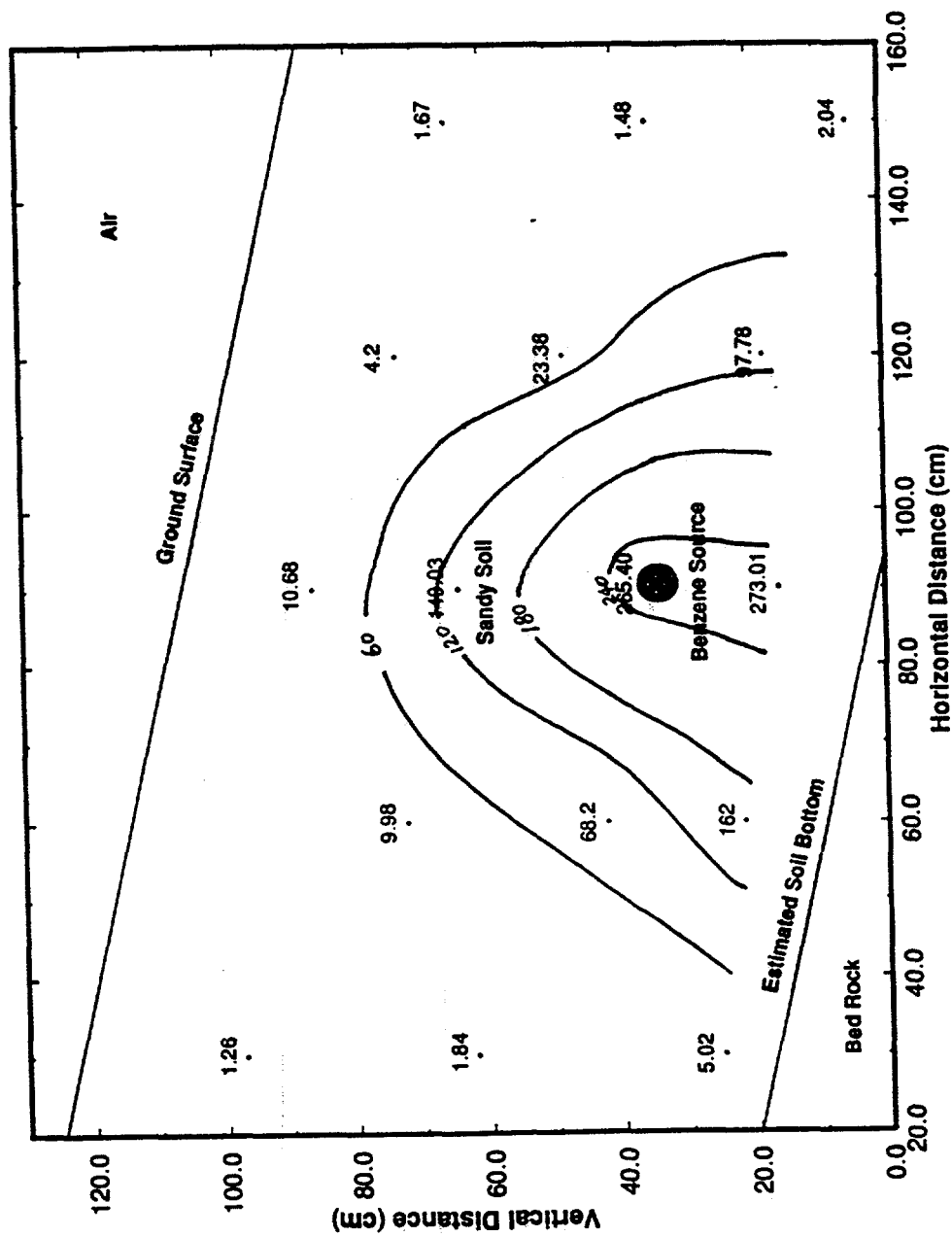
Figure 8. Arrangement of Measurement Ports



1,2 ---- ports for both experiments 1 and 2. 2 ---- ports for experiment 2 only.

Figure 9. Contours of Soil Gas Benzene Content (ppm) in Shadowlane

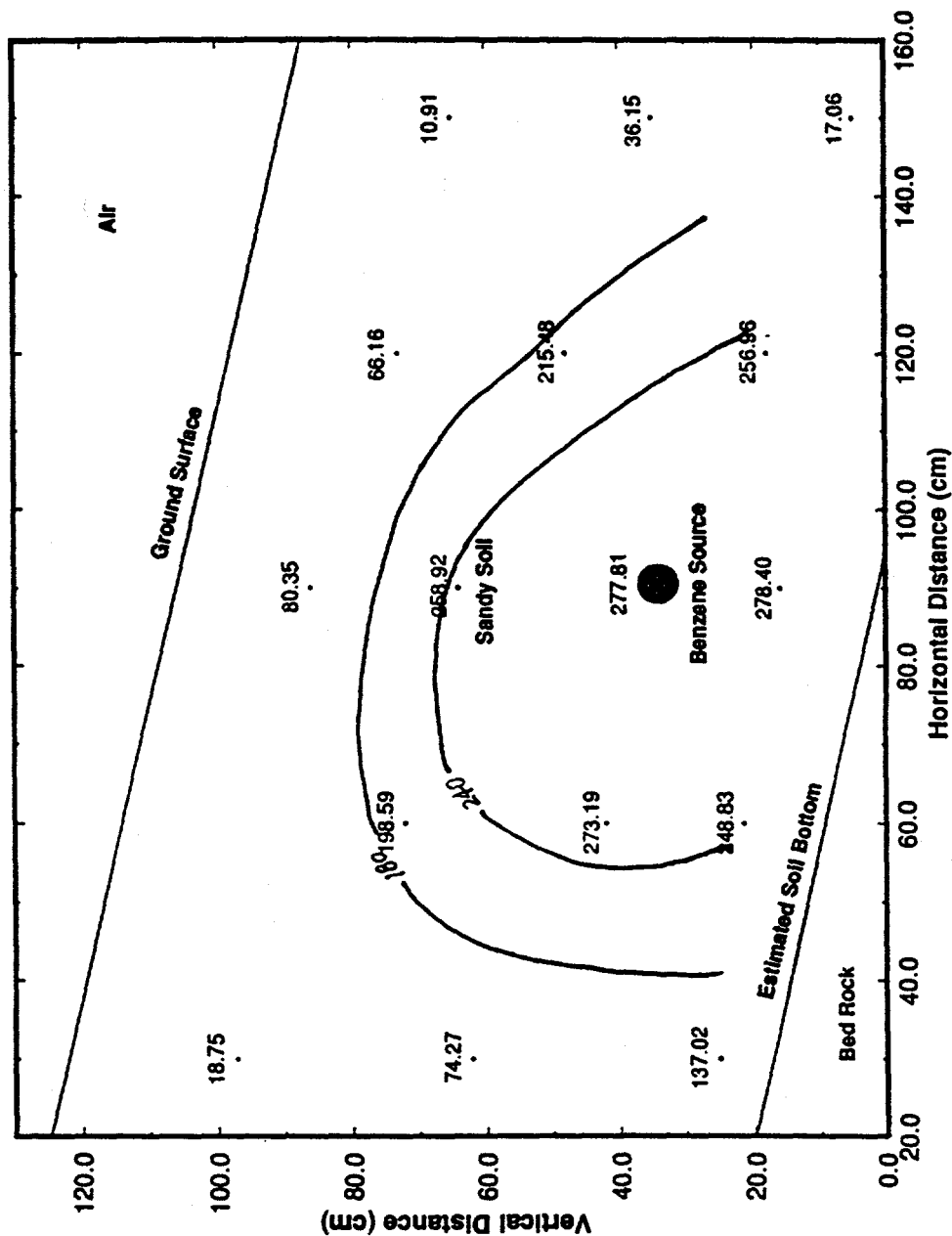
5 hours after injection / 1:25 pm, 07/11/95 / $T_s = 23.42\text{ C}$, $T_b = 13.83\text{ C}$



T_s , T_b — Temperature on Ground Surface and Estimated Soil Bottom, Respectively

Figure 10. Contours of Soil Gas Benzene Content (ppm) in Shadowlane

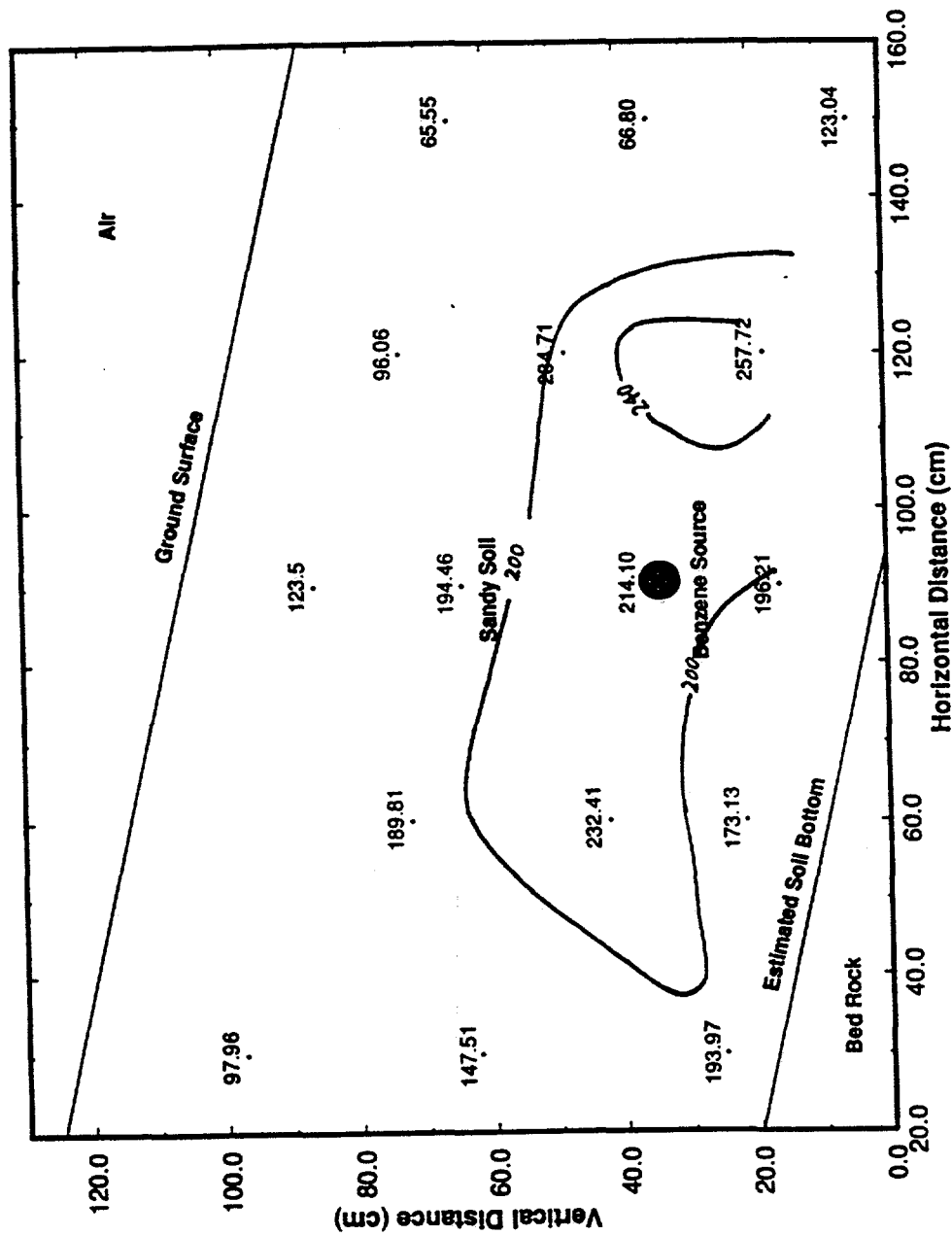
25 hours after injection / 9:25 am, 07/12/95 / $T_s = 20.78^\circ\text{C}$, $T_b = 13.85^\circ\text{C}$



T_s , T_b — Temperature on Ground Surface and Estimated Soil Bottom, Respectively

Figure 11. Contours of Soil Gas Benzene Content (ppm) in Shadowlane

80 hours after Injection / 4:25 pm, 07/14/95 / Ts = 29.33 C, Tb = 13.96 C



Ts, Tb — Temperature on Ground Surface and Estimated Soil Bottom, Respectively

AIR FLOW DATA
Colerain Talus Site
Huntingdon Co. PA

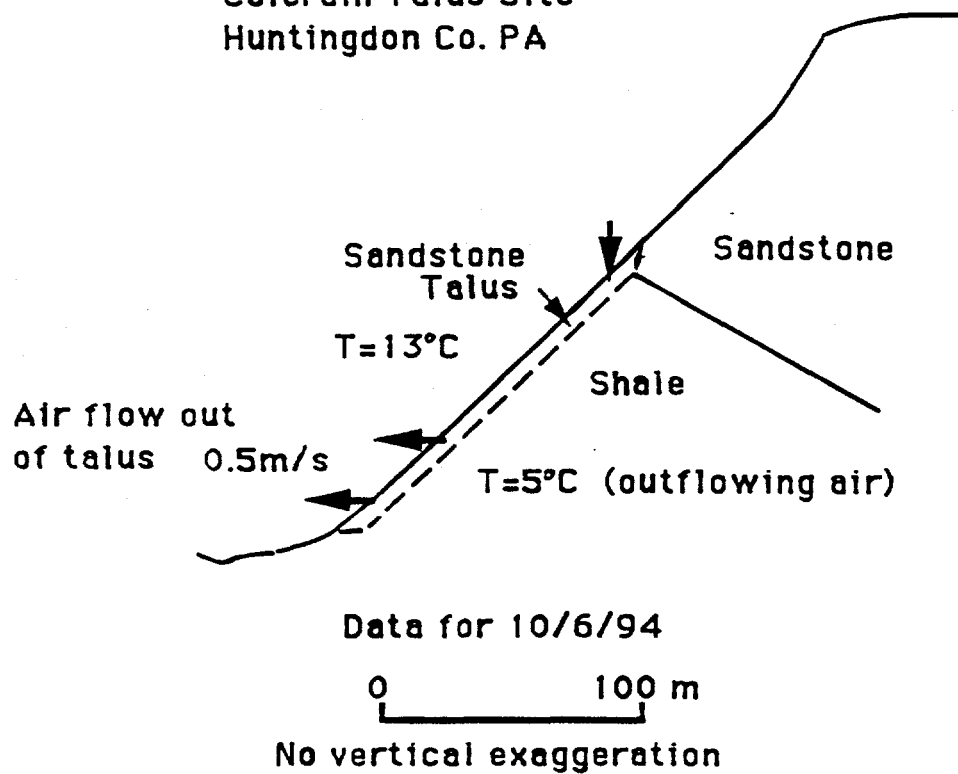


Figure 12. Sketch of the Colerain talus site, showing flow of air and air temperature on October 10, 1994.

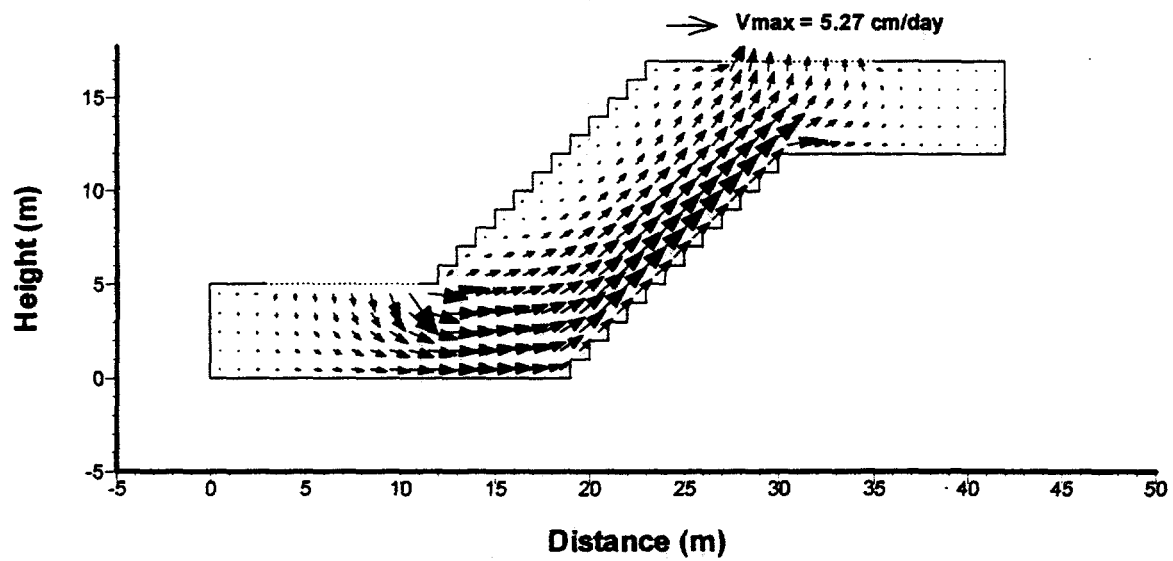


Figure 13 Air convection simulation in a sloping layer (grade=1) with the upper boundary partially open (5 C at top and 10 C at bottom; porosity=0.3; permeability = 10^{-6} cm^2)

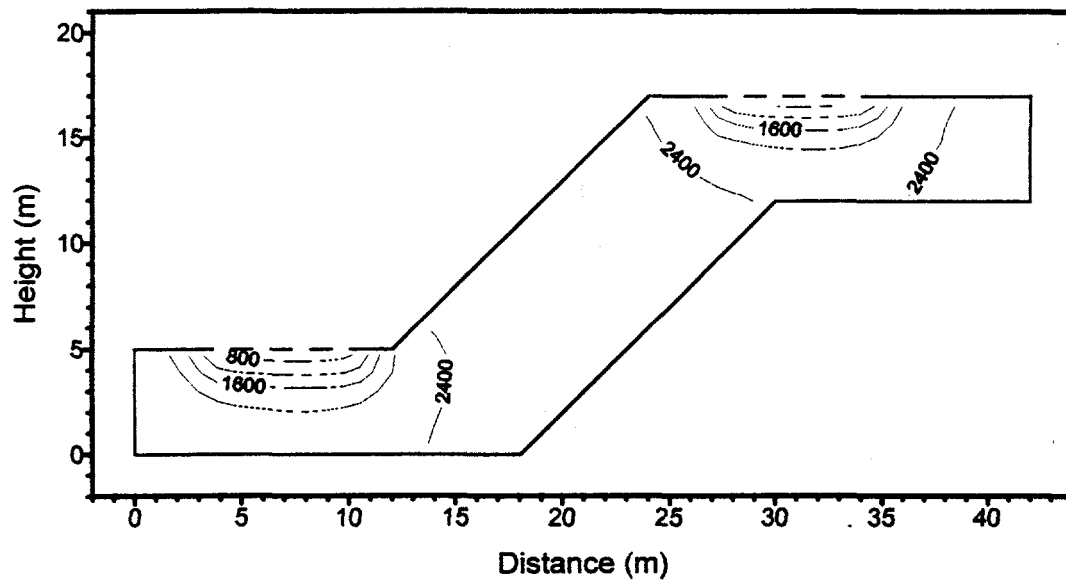


Figure 14 Contours of gas radon concentration (pCi/L) in a sloping layer(grade=1) with the upper boundary partially open and all the other boundaries closed (5 C at top and 10 C at bottom; porosity = 0.3; permeability = 10^{-6} cm²; radon generation rate=752.027 pCi/L)

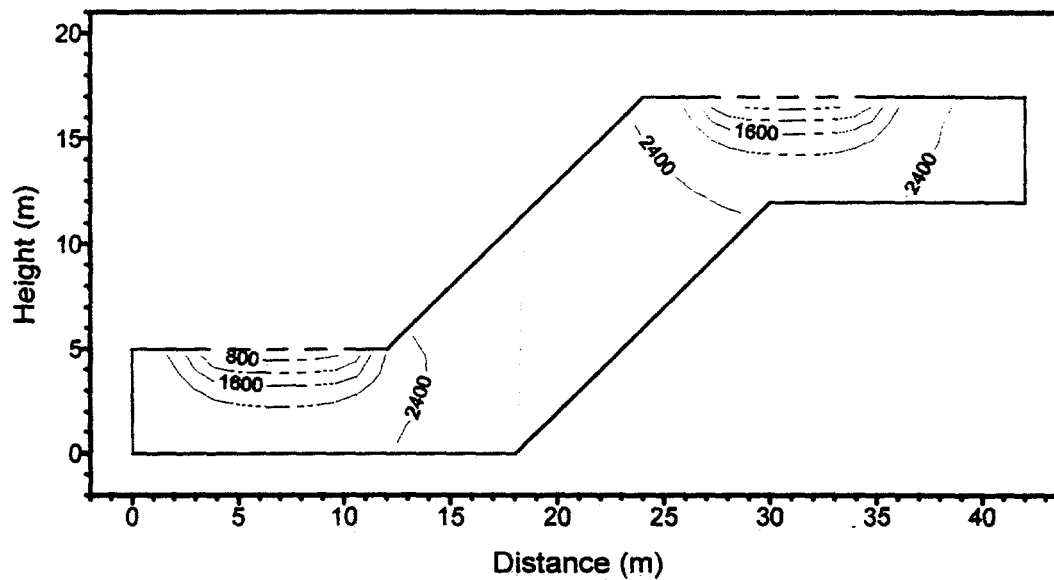


Figure 15 Contours of gas radon concentration (pCi/L) in a sloping layer(grade=1) with the upper boundary partially open and all the other boundaries closed (5 C at top and 5 C at bottom; porosity = 0.3; permeability = 10^{-6} cm²; radon generation rate=752.027 pCi/L)

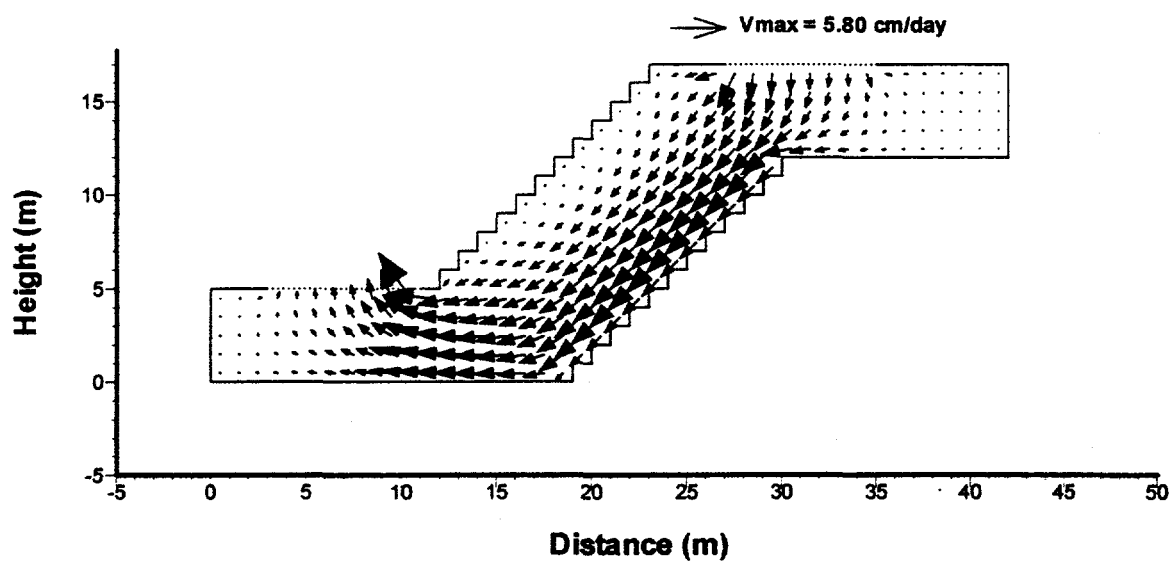


Figure 16 Air convection simulation in a sloping layer (grade=1) with the upper boundary partially open (10 C at top and 5 C at bottom; porosity=0.3; permeability = 10^{-6} cm^2)

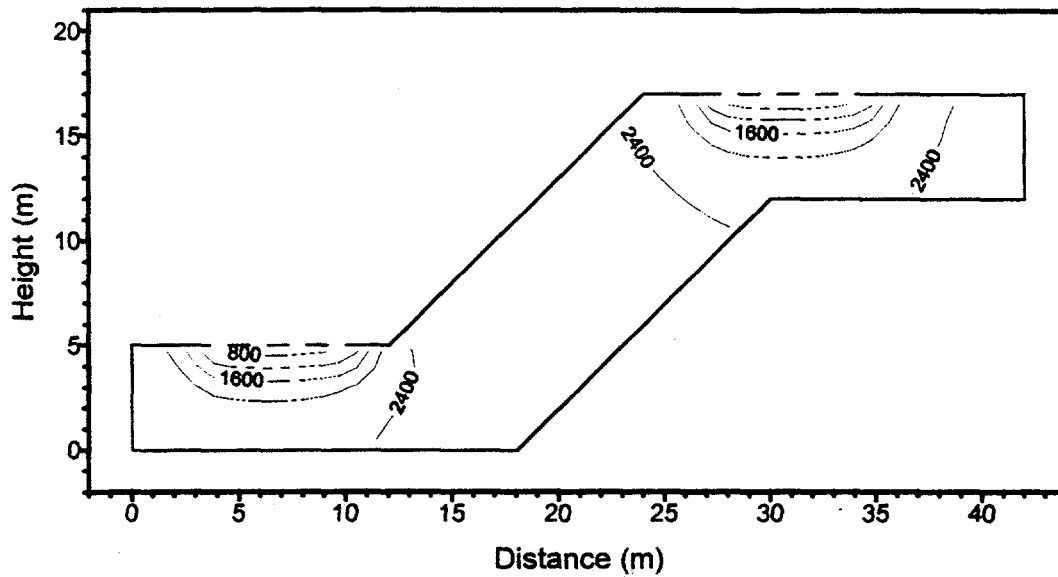


Figure 17 Contours of gas radon concentration (pCi/L) in a sloping layer(grade=1) with the upper boundary partially open and all the other boundaries closed (10 C at top and 5 C at bottom; porosity = 0.3; permeability = 10^{-6} cm²; radon generation rate=752.027 pCi/L)

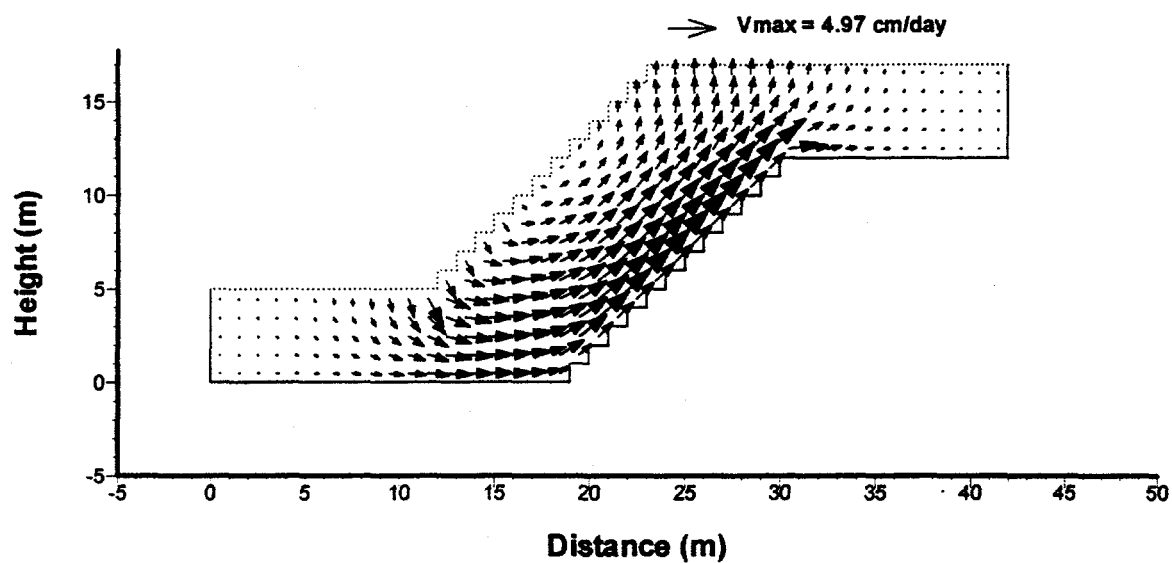


Figure 18 Air convection simulation in a sloping layer (grade=1) with the upper boundary open (5 C at top and 10 C at bottom; porosity=0.3; permeability = 10^{-6} cm^2)

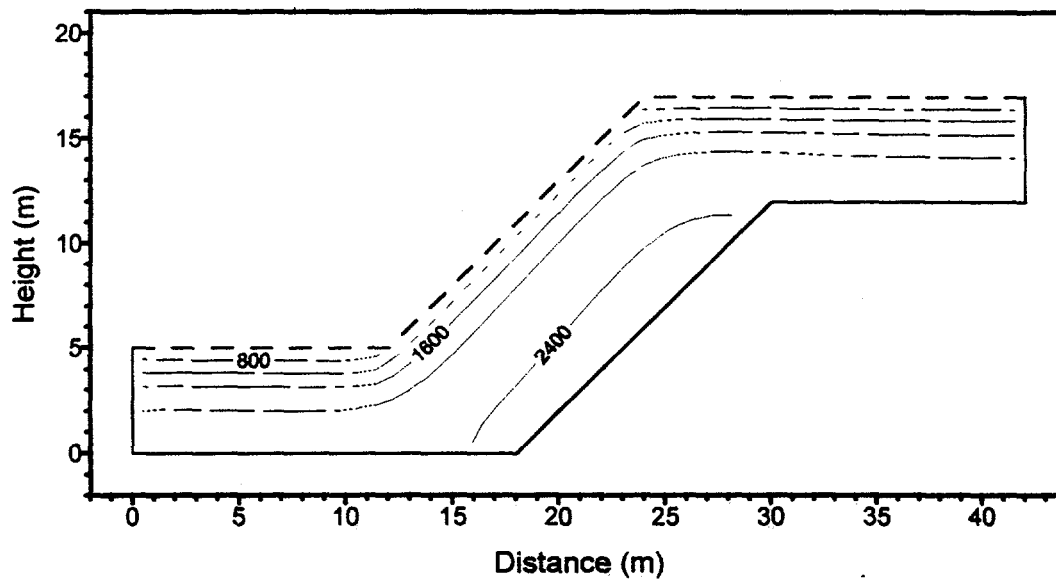


Figure 19 Contours of gas radon concentration (pCi/L) in a sloping layer(grade=1) with the upper boundary open and all the other boundaries closed (5 C at top and 10 C at bottom; porosity = 0.3; permeability = 10^{-6} cm²; radon generation rate=752.027 pCi/L)

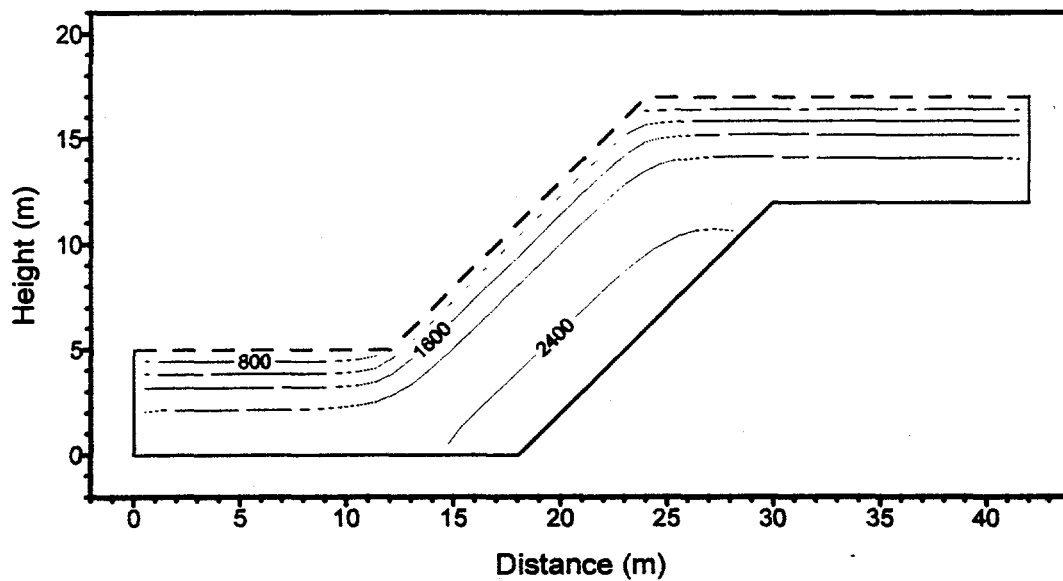


Figure 20 Contours of gas radon concentration (pCi/L) in a sloping layer(grade=1) with the upper boundary open and all the other boundaries closed (5 C at top and 5 C at bottom; porosity = 0.3; permeability = 10^{-6} cm²; radon generation rate=752.027 pCi/L)

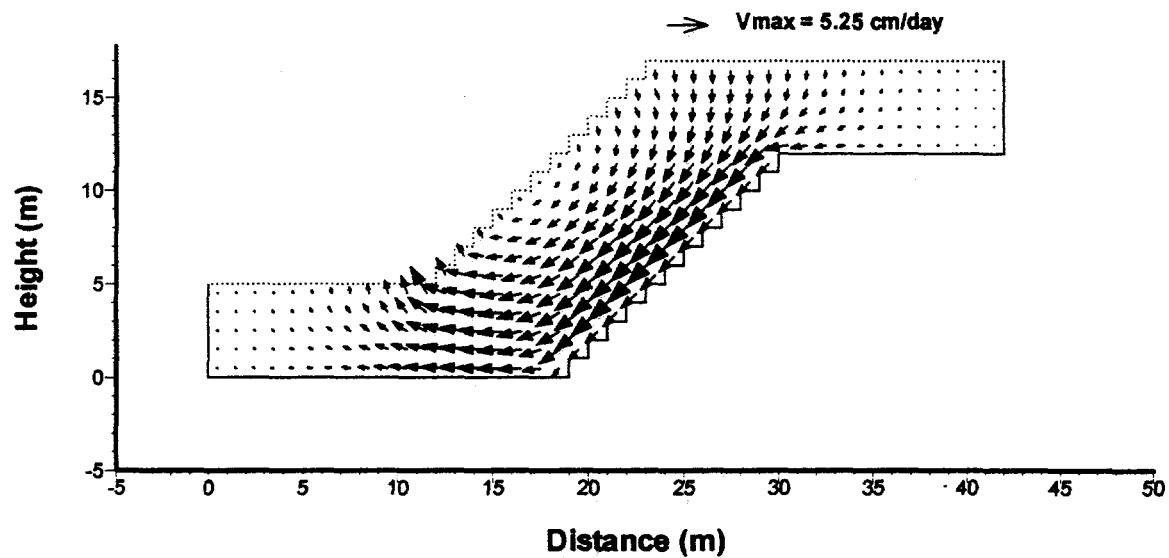


Figure 21 Air convection simulation in a sloping layer (grade=1) with the upper boundary open (10 C at top, 5 C at bottom porosity=0.3; permeability = 10^{-6} cm^2)

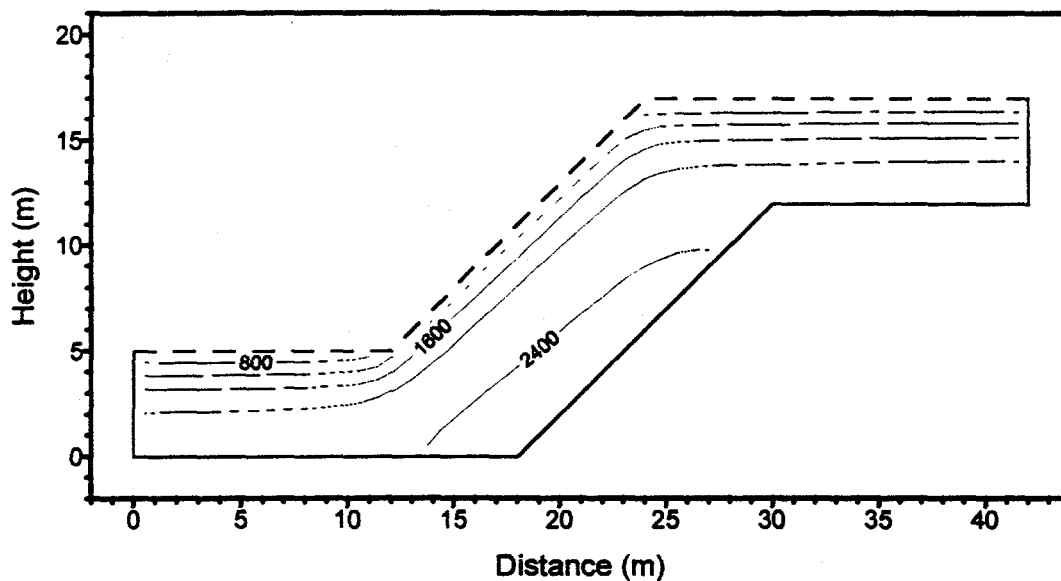


Figure 22 Contours of gas radon concentration (pCi/L) in a sloping layer(grade=1) with the upper boundary open and all the other boundaries closed (10 C at top and 5 C at bottom; porosity = 0.3; permeability = 10^{-6} cm²; radon generation rate=752.027 pCi/L)

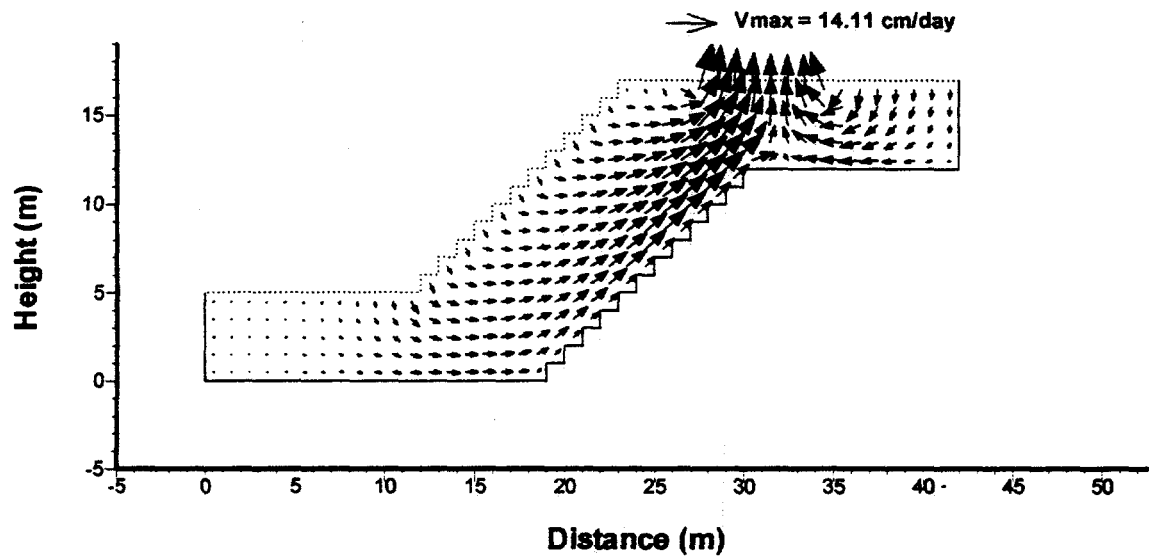


Figure 23 Air convection simulation in a sloping layer (grade=1) with the upper boundary open (5 C at top, 10 C at bottom and 20 C in the room ($x = 27 - 35$, $y = 17$); porosity=0.3; permeability = 10^{-6} cm^2)

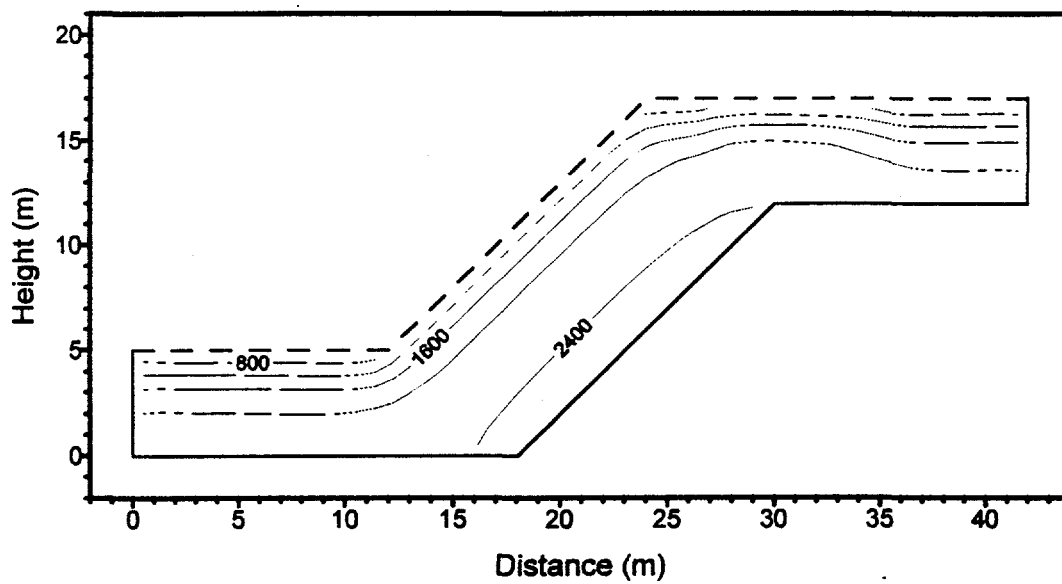


Figure 24 Contours of gas radon concentration (pCi/L) in a sloping layer (grade=1) with the upper boundary open and all the other boundaries closed (5 C at top, 10 C at bottom and 20 C in the room ($x = 27 - 35$, $y = 17$); porosity = 0.3; permeability = 10^{-6} cm^2 ; radon generation rate=752.027 pCi/L)

APPENDIX A

Manuscript of
GEOCHEMISTRY OF RADIUM IN SOILS OF THE EASTERN UNITED STATES

Daniel J. Greeman, Arthur W. Rose, John W. Washington, Robert
R. Dobos and Edward J. Ciolkosz

GEOCHEMISTRY OF RADIUM IN SOILS OF THE EASTERN UNITED STATES

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ABSTRACT

The abundance and form of ^{226}Ra , ^{238}U and ^{232}Th have been determined on samples of soil, rock, and vegetation at 12 sites in the eastern United States. Progressive, selective chemical extraction determined abundance and radiometric equilibrium condition of these nuclides in 6 soil fractions: exchangeable cations, organic matter, "free" Fe-oxides, sand, silt, and clay.

In soils, average profile $^{226}\text{Ra}/^{238}\text{U}$ activity ratios are within 16% of unity for all 12 sites, implying little relative loss between daughter products. However, ^{226}Ra greatly exceeds U activity in vegetation ($^{226}\text{Ra}/^{238}\text{U}$ up to 65), in soil organic matter ($^{226}\text{Ra}/^{238}\text{U}$ up to 30), in soil Fe oxides ($^{226}\text{Ra}/^{238}\text{U}$ up to 1.8) and in the C-horizons of deeply weathered soils ($^{226}\text{Ra}/^{238}\text{U}$ up to 1.5).

A major factor in Ra behavior is cycling by vegetation, which concentrates $\text{Ra} > \text{U}$, creating the observed Ra excess in typical surface soil horizons (up to $\text{Ra}/\text{U} = 1.8$) in 1000 years. Of the total Ra in average the A-horizon, 42% occurs as exchangeable ions and in organic matter, but only 6-8% of the parent U and Th

occur in these soil forms. A larger occurrence of U and Th in the Fe-oxides of this horizon, 17-19% of the total, and low U and Th values relative to those at depth, imply rapid chemical partition of cycled U and Th.

Transfer by vegetation and/or direct chemical partitioning of Ra into organic and exchangeable forms leaves all soil minerals Ra-poor ($Ra/U = 0.73$) and compensates for loss by radioactive decay of unsupported ^{226}Ra in Ra-rich organic matter.

INTRODUCTION

The behavior of radium in soils is important because of the hazard of its daughter products, especially radon, which furnishes an estimated 55% of the average human dose of radioactivity (NCRP, 1987). Radium behavior in soils is also crucial to understanding the uptake of Ra, which next to ^{40}K is the greatest source of radioactivity in food (Hewamanna, 1987).

Previous research documents significant fractionation of Ra from other U-series nuclides during weathering (Rosholt, 1982), but data on complete sets of soil horizons and associated vegetation are limited or lack quantitative assessment of pedogenic controls on radium occurrence.

In order to determine the effect of weathering and vegetation on the behavior of ^{226}Ra in soil, Ra was determined in soil, rock, and vegetation on samples from of twelve widely varied soil sites in the eastern U. S. (Figure 1). Progressive, selective chemical extraction was used to determine speciation of

^{238}U , ^{232}Th , and ^{226}Ra in the different soils. Uranium is measured to examine relations of Ra to its ultimate parent, and ^{232}Th content as a proxy for ^{230}Th , the immediate parent of ^{226}Ra .

The action of the biosphere is taken into account by analysis of representative specimens of plants at the sites, and by study of the organic fraction of the soil. Models consistent with spatial and chemical radionuclide speciation, constrained by U-series equilibrium condition, are developed to help explain the form and geochemical evolution of Ra in soils.

Since the major U-series nuclides parental to Rn (Figure 2) have greatly divergent half-lives (4.5 Ga to 1.6 ka) and chemical properties, short-lived nuclides may not be affected by relatively slow pedogenic processes, while longer-lived nuclides may be profoundly affected, especially where chemical, physical, and biological soil processes are intense, as in A-horizons.

For convenience in the remainder of this paper, Ra refers to ^{226}Ra , U refers to ^{238}U and Th to ^{232}Th , unless another isotope is specifically indicated.

Previous Research

Up to 75% of U and Th, occur in pedogenic phases (i.e., Fe-oxides, organic matter and other secondary phases formed during weathering), indicating that soil formation involves release of a portion of the radionuclides from primary hosts and some mobility (Hansen and Stout, 1969; Hansen, 1970; Megumi, 1979; Lowson et al., 1986; Gueniot et al., 1988). The effect of plants and

organic matter as significant agents in mobility of U-series nuclides was not explicitly evaluated by these researchers.

Profiles of Ra abundance with soil depth commonly show relatively constant values with depth, with more similarity to the behavior of U than to Th (Baranov et al., 1963; Hansen and Huntington, 1969; Hansen, 1970). Some soil studies indicate significant recent introduction of Ra to the A-horizon, based on Ra excess over ^{230}Th , but most do not provide a clear mechanism for this excess (Michel, 1984; Hansen and Huntington, 1969). Radium is strongly adsorbed by soils (Nathwani and Phillips, 1979a), and tends to be enriched in the fine clay fraction, but not particularly in the Fe-oxide fraction (Megumi, 1979). Nathwani and Phillips (1979a) inferred that soil organics have 10 times the affinity for Ra as soil clays. Taskeyev et al. (1978) showed that a significant part of the Ra in an organic-rich soil inundated by Ra-rich groundwater was held in organic form.

Vegetation has a significant affinity for Ra (Rusanova, 1963; Popova et al., 1964; Taskayev et al., 1977). Activities of Ra in vegetation commonly exceed those of U, and U commonly exceeds Th (Rosholt et al., 1966; D'Souza and Mistry, 1970; Titayeva et al., 1977; Mahon and Mathewes, 1983; Drichko and Lisachenko, 1984).

SAMPLING AND ANALYTICAL METHODS

This investigation is based on data from 12 sites of varying soil type, parent material and location (Table 1, Figure 1). Except for site IL-1 which is farmed, all the soil sites have

been relatively undisturbed for at least 100 years as evidenced by mature tree stands. All the soil profiles show well developed horizons.

The 12 soil sites were chosen to represent major soil groups (alfisols, ultisols, mollisols, spodosols, and inceptisols) developed on several parent materials (shales, sandstones, alluvium, glacial tills, crystalline rocks, and carbonates) spanning a large proportion of the eastern U. S. Ages of soils range from millions of years to as little as 500-2000 years with several soils developed on Pleistocene till with ages of about 10,000 years. At each site, 9 to 12 soil horizons were described in 1.5-3.5 meter deep backhoe pits using USDA nomenclature. Soil and rock fragment samples of 2-35 kg were collected from every soil horizon. Vegetation was collected not farther than 10 meters from the pit. Rock samples of 2-10 kg were collected as close as possible to the pit or from outcrops on strike with the pit.

Soil samples were disaggregated and sieved through a 2 mm mesh. The data reported here are for this "soil" fraction. Analyses of particle size distribution, organic carbon, "free iron oxides" (ferric oxides and hydroxides) and exchangeable cations were determined by the procedures of Ciolkosz et al. (1988). Vegetation samples were thoroughly rinsed in distilled water, dried, and identified by Profs. R. A. Purcell and C. S. Keener (Biology Department, Penn State Univ.). The vegetation was then oven dried at 105°C, and ashed at 500°C. The ash was

digested twice in 40 ml of 50% HNO_3 at 50°C for 2 hrs, the solution centrifuged and decanted from any minor mineral residue and evaporated to 30 ml.

Progressive, selective chemical extractions were performed on field moist soil samples of 6 to 16 g representing the major horizons. The procedure differed slightly for different sets of samples (Figure 3).

Exchangeable cations were extracted with 1M MgCl_2 adjusted to pH 5 to minimize adsorption of cations. Organic matter was decomposed with 5% NaOCl adjusted to pH 5 with HNO_3 . This treatment primarily oxidizes the electron-rich aromatic and carboxylic functional groups of large organic molecules in geologic samples (Kogel-Knabner and Hatcher, 1989) so any trace element thus liberated is probably associated with those functional groups and groups adjacent to them. The "free" oxides of Fe, Mn, and Al were extracted with Na-dithionite in conjunction with the complexing agents citrate and bicarbonate at pH 8, $75\text{--}80^\circ\text{C}$ (Jackson, 1956).

Experiments confirmed that no significant re-adsorption of extractant-liberated radionuclides to substrate or vessel occurred. Rocks and soils were completely digested in covered Teflon beakers by refluxing with 25 ml conc. HF , 20 ml conc. HNO_3 , and 5 ml distilled water for 48 hrs. The solution was then taken to dryness, dissolved with 5 ml conc. HNO_3 , and diluted to 30 ml.

The solutions from the digestions described above, the vegetation digestions, and the supernatants from selective extraction steps were loaded into 50 ml radon-bubbler tubes, purged with He gas and sealed. Ra was then determined using a modified Rn-bubbler technique (Greeman et al., 1990). Uranium, Th, and for a few samples Ba were determined by neutron activation at Activation Labs Inc. (Ancaster, Ontario) on bubbler solutions taken to dryness, and on 1-10 g of ground rock, soil, and plant ash sealed in polyvials.

In general, selective extractions were reproducible and extractions yielded similar trends in occurrence of each radionuclide in various phases over the full depth range at each site. Analytical totals, comprised of six to eight separate analyses, averaged 85% for Ra; 94% for U; and 97% for Th. The extractions are therefore considered to be relatively accurate, at least for the dominant forms. All speciations are normalized to 100% totals, and calculations for profiles have been normalized to 100% depth coverage from an actual average of 51% coverage, using thickness-weighted values. Additional details of methods are discussed by Greeman (1992).

RESULTS

Radium Enrichment in Soil Relative to Bedrock

The gross concentration of Ra in the soils is related to the U content, which is enriched by up to 2-fold in soils over non-carbonate rocks and up to 15-fold in soils over carbonate rocks (Table 2). The large enrichment of Ra in limestone soils is

doubtless responsible for the high levels of Rn reported in homes on these soils (Tanner, 1986).

Averaged over all depths at a given site, Ra is within 10% of $^{226}\text{Ra}/^{238}\text{U}$ equilibrium at nine of the twelve sites, indicating minimal disequilibrium for these profiles as a whole. At site 6-11 Ra is only 88% of equilibrium, and at site IL-1 and NC-1 Ra is 111% and 116% of equilibrium, respectively (Table 2). All 11 samples of bedrock and float rock show Ra/U equilibrium within the error of analysis, $\text{av. } ^{226}\text{Ra}/^{238}\text{U} = 0.95 \pm 0.09$.

Thorium and U are depleted in the surface horizons at nearly all sites (Table 3); values increase with depth up to 2-fold and 6-fold, respectively. However, radium trends are more variable (Figure 4). At the most intensely weathered sites (TN-1 and NC-1), Ra is markedly depleted in the surface horizons in a manner similar to U. At most of the other sites, Ra varies relatively little through the profile, excepting distinct enrichment in most surface A-horizons (Figure 4).

At 7 of the 12 sites (14-80, 14-81, 14-82, 6-10, NY-1, IL-1, and TN-1), the Ra/U activity ratio exceeds unity in the A-horizon compared to deeper horizons (Figure 5). Site 14-83 has elevated Ra/U in the upper B horizon, and site 8-99 has high values in the E horizon. Evidently Ra is being separated from U during the soil-forming process at these sites.

Radium is also enriched (up to 150% Ra/U equilibrium) in the C-horizons of some soils (14-84, NY-1, and TN-1) in contrast to overlying horizons where the Ra/U values are 20-50% lower.

Radionuclides in Vegetation

The representative plants contain abundant U, Th, and Ra in their ash (average 1.2 ppm U, 2.2 ppm Th, and 41 mBq/g Ra), with radionuclide content varying considerably, 100-fold, 40-fold and 30-fold, respectively (Table 4). Concentration factors (CF, the value in plant ash divided by the average soil value) for U, Th, and Ra average 0.54, 0.33, and 0.91, respectively, for 16 samples of live plants, indicating clearly that fractionation into plants is in the order $Ra > U > Th$.

An obvious feature of the live plants is the extremely large range in Ra/U activity ratios (1600-fold) and high Ra/U, up to 65. With the exception of grasses from site IL-1 (Ra/U = 0.04), all Ra/U activity ratios for plant ash are either close to or greatly exceed unity.

Selective Extractions

Sequential selective extractions show that, on average, 35% of the total Ra is dispersed amongst the pedogenic phases (exchangeable + organic + poorly-crystalline soil-oxide) in soils, compared to only 16% of the U (Table 5). In 3 of 4 profiles, the amount of pedogenically bound Ra increases with depth, up to 5-fold (Figures 6, 7).

Uranium and Th on average have <2% of their total occurrence in soils as exchangeable cations, generally restricted to the upper 30 cm (Table 5; Figure 6). In contrast, Ra has up to 35% of its total occurrence as an exchangeable cation, when the extractant is NH_4 -acetate, and 18% using the $MgCl_2$ extractant.

In two of the four sites, maximum exchangeable-Ra values occur at depth (Figure 7).

Exchangeable Ra values (av.- 10% of total occurrence) are intermediate between exchangeable Mg and Ca values (av.- 3.5% and 33%, respectively) and much higher than exchangeable K (0.26% of the total K). Correlations of exchangeable U, Th and Ra with other soil properties, including cation exchange capacity (C.E.C.), base saturation, pH, or exchangeable Mg^{2+} , Ca^{2+} , K^{+} , and H^{+} , are not statistically significant.

In contrast to the behavior of its precursors ^{238}U and probably ^{230}Th by analogy with ^{232}Th , Ra is strongly associated with the humic fraction (sum of exchangeable cation + organic extractions) in all horizons: the maximum percentage of the total Ra in this form is 42%, minimum 9.4%, and average 26% (Table 5, Figure 7). Radium in the humic fraction typically exceeds equilibrium with U by 1000% (as high as 3000%); the Ra/U ratio in this form generally increases with depth (Figure 7).

The vast majority of pedogenic, nonmineral U and Th is associated with "free" (dithionite-extractable) Fe-oxides: maximums are 38% and 67%, and averages are 15% and 31% of the total U and Th, respectively (Table 5, Figure 6). In general the occurrence of U and Th in Fe-oxides increases with depth (up to 3-fold and 5-fold, respectively).

A smaller proportion of Ra is in the Fe-oxides than its precursors U or Th: maximum- 21%, and average 10% of the total Ra. Unlike U or Th, the abundance of Ra in Fe-oxides does not

show a clear pattern with depth. Radium-uranium equilibrium in Fe-oxides ranges up to 10-fold with depth, from 38% of equilibrium to 380% of equilibrium, (Figure 7) but site-averaged Ra/U in Fe-oxides is close to 100% (Table 5).

Radionuclides in Residual Sand, Silt and Clay

Mineral phases show a net loss of Ra relative to U; profile-averaged $^{226}\text{Ra}/^{238}\text{U}$ values range from 0.72-0.91, indicating consistent leaching of Ra from the residual phases (Table 5).

DISCUSSION

Depletion of U and Th from Surface Horizons

A relatively consistent feature of the twelve soils is the apparent depletion of Th (all 12 sites), U (9 of 12 sites), and Ra (7 of 12 sites) from the surface 30 cm, and an even more marked depletion of U and Th if only the A- and E- horizons are considered (Table 3).

Similar patterns are reported by others. A profile developed in a warm, humid climate in Mississippi showed surface depletions of U and Th (Rosholt et al., 1966). Hansen and Stout (1968) and Hansen and Huntington (1969) found surface depletion of Th in most profiles and of U in two profiles of California soils in cool areas. In three of four brown acid soils (spodosols, alfisols) studied by Gueniot (1983), U and Th are strongly depleted from the soil surface. In warm, humid climates U and Th are commonly mobilized out of the soil surface.

The depletion of U and Th from the A- and E-horizons results primarily from eluviation of radionuclide-bearing Fe-oxides from

the surface as colloidal particles and in part by solution. A secondary reason for depletion of U and Th from surface horizons is leaching of these elements from the sand and silt fraction of some soils (e.g., NC-1 sand, NY-1).

Surface Enrichment of Radium

In spite of its origin from decay of U and Th isotopes, Ra generally shows less depletion from the A-horizon than U, or in some cases even absolute enrichment in the A horizon (Figures 4 and 7), indicating a different mechanism for Ra emplacement toward the surfaces of soils.

In the organic-rich A-horizons at five sites (14-80, 14-81, 14-82, 6-10, IL-1) Ra activities exceed U activities to give $^{228}\text{Ra}/^{238}\text{U}$ values up to 1.8 (Figure 5). At three other sites, excess Ra occurs in the E-horizon (8-99) or in the humus-rich Bhs-horizon (14-83, NY-1) immediately below the E-horizon. The majority of the Ra excess, accounting for over 40% of the total Ra in these A-horizons, occurs in the "humic fraction" (exchangeable cations + organic extract) of these soils. For example, $^{226}\text{Ra}/^{238}\text{U} = 8.5$ in the exchangeable cations + organic extract for site 14-80. A smaller proportion of excess Ra in Fe-oxides in the soil surface, with $^{226}\text{Ra}/^{238}\text{U}$ up to 3.8, also contributes to excess Ra in the A-horizons of these soils.

Sands and silts of the A-horizon at the Ra-rich sites show equilibrium $^{226}\text{Ra}/^{238}\text{U}$ values, 1.04 ± 0.32 , 0.90 ± 0.09 and 1.17 ± 0.14 (Table 6) and cannot contribute to Ra excess.

On average, <2% of the total ^{238}U is associated with the humic fraction. Ingrowth of daughter isotopes, such as ^{234}U or ^{226}Ra is expected to be minimal there. Similarly, the humic fraction contains essentially no ^{232}Th (av. <2% of the total) and by analogy, little ^{230}Th . Since a large proportion, up to 42%, of the total Ra is associated with the humic fraction, and no apparent Ra-ancestor exists there, a significant proportion of the Ra occurrence, up to 90%, is unsupported.

The occurrence of unsupported Ra in the humic fraction in the biologically active A-horizons and in the organic-rich upper portion of the B-horizon, in combination with the distinct selectivity of plants for Ra over U, strongly suggests that plants are responsible for the observed Ra enrichments. At the sites of the current study, $^{226}\text{Ra}/^{238}\text{U}$ averages 2.8 for 16 samples of live plants (Table 4). Radium-uranium values averaging 19 were reported by Mahon and Matthewes (1983).

In significantly drier soils, Rosholt et al. (1966) and Ghaleb et al. (1990) show ^{230}Th excess relative to ^{238}U in some near-surface horizons as a result of higher U mobility in those soils. Excess ^{226}Ra , relative to ^{238}U , can eventually result from decay of excess ^{230}Th in the soil surface. However, in more humid eastern U.S. soils, ^{232}Th is mobilized downward from the surface (Figure 6); by analogy ^{230}Th is probably also similarly mobilized. Therefore, Ra enrichment most probably results from cycling by vegetation.

Radium Supply by Vegetation

The change with time in radium content in any given soil horizon is given by:

$$dC_{Ra}/dt = F_{in} - D - F_{out} + I \quad (1)$$

where C_{Ra} is the Ra content in the horizon per unit area, t is time, F_{in} is the rate of any Ra flux into the system (e.g., plant-incorporated Ra), D is radioactive decay ($-l_{Ra}C_{Ra}$), F_{out} is the Ra flux rate out of the system (e.g., solution loss, plant uptake), and I is radioactive ingrowth from ^{230}Th ($l_{Th}^{230}C_{Th}^{230}$).

As a first step, the time period for buildup of Ra within pedogenic phases in the A-horizon is estimated for a soil initially in U-series equilibrium through Ra but with no pedogenic Ra. F_{in} is the Ra flux contributed from above by vegetation, with at least some of this Ra flux being derived originally via plant roots from beneath the A-horizon. F_{out} is loss of Ra from the A-horizon, by a combination of downward flow in solution and suspension, and uptake from the A-horizon into vegetation. The loss is assumed to be proportional to the pedogenic Ra in the horizon, i.e., $F_{out} = aC_{Ra}$. Plants are assumed not to change in Ra content over time so that the flux of Ra from plants (F_{in}) remains constant. Ingrowth from radioactive parent, ^{230}Th , is assumed insignificantly small because $^{226}Ra \gg ^{230}Th$ in the exchangeable-organic-Feoxide phases, as discussed previously.

Substituting $-l_{Ra}C_{Ra}$ for D and aC_{Ra} for f_{out} , eliminating I for the reasons stated above, and expressing F_{in} as k (the flux of Ra from vegetation), the general form of the equation is:

$$dC_{Ra}/dt = k_{in} - l_{Ra}C_{Ra} - aC_{Ra} \quad (2)$$

If C_{Ra} is taken as pedogenic Ra (i.e., Ra in resistant minerals is immobile and in equilibrium with its precursors), so that $C_{Ra} = 0$ at $t = 0$, then the time to furnish the observed Ra can be estimated. First integrating:

$$t = (\ln k - \ln {}^0k - (l_{Ra} + a)C_{Ra}^0)/l_{Ra}+a \quad (3)$$

For site 14-80, the amount of pedogenic Ra per unit area of A-horizon soil, C_{Ra-p} , is given by:

$$C_{Ra-p} = T A_{Ra} r F_s \quad (4)$$

where T is the horizon thickness (A-horizon, 8 cm), A_{Ra} is the activity of pedogenic radium per unit mass of fine soil (50 mBq/g, Figure 6), r is the bulk density of the soil (1.3 g/cm³, Washington, 1991), and F_s is the proportion of fine fraction (<2 mm) in the soil (0.787, Greeman, 1992). From equation (4) the calculated pedogenic Ra, C_{Ra} , is 0.41 Bq/cm².

The flux of radium per unit area by litterfall, k_{in} , to the A-horizon from vegetation is estimated by:

$$k = R_a V P_a C_t / F_c \quad (5)$$

where $R_a V$ is the average Ra in plant ash (41 mBq/g, Table 4), P_a is the average proportion of ash in dried plants (0.062, Table 4), C_t is the annual turnover of carbon from vegetation to a forested soil (0.06 g C/cm²-yr for temperate deciduous forests, Olsen et al., 1983), and F_c is the average proportion of carbon in dried terrestrial plants (0.45, Mason and Moore, 1982). From equation (5), the calculated plant flux value is 0.34 mBq/cm²-yr. Additional fluxes are from leaf wash (Ra dissolved directly from

green leaves and branches) and stem flow (water flowing down the trunk). For 12 deciduous forests, Cole and Rapp (1981) show that these two fluxes average 48% and 107% of the flux in litterfall for Ca and K, respectively. Assuming that for Ra this flux amounts to 50% of the litterfall, then $k = 0.51 \text{ mBq/cm}^2\text{-yr}$.

If $a=0$ (no loss of Ra from the A-horizon), the time for accumulation of the observed excess of Ra in the A horizon is 990 years, calculated from equation (3). Since the soil at site 14-80 has an age of 350,000 to 1,200,000 years (Cronce, 1988), the time available is more than adequate for accumulation of the required excess Ra in the A-horizon. Clearly, vegetation cycling can be extremely important in determining the form and behavior of Ra in this and other soils with age exceeding a few thousand years.

At steady state conditions, the requirement is:

$$dC_{\text{Ra}}/dt = k - l_{\text{Ra}}C_{\text{Ra}} - aC_{\text{Ra}} = 0 \quad (6)$$

For this condition, $a = k/C_{\text{Ra}} - l_{\text{Ra}}$. Given that $k = 0.51 \text{ mBq/cm}^2\text{-yr}$, $C_{\text{Ra}} = 0.41 \text{ Bq/cm}^2$, and $l_{\text{Ra}} = 4.33 \times 10^{-4} \text{ yr}^{-1}$, then $a = 8.1 \times 10^{-4} \text{ yr}^{-1}$. This value is very similar to the analogous value calculated for Ba at site 14-80. The Ba content of two ashed vegetation samples averages 300 ppm and the A-horizon contains 400 ppm Ba. Since Ba is not lost or gained by radioactive decay, then at steady state $F_{\text{in}} = F_{\text{out}} = aC_{\text{Ba}}$ or $a = 1.16 \times 10^{-3} \text{ yr}^{-1}$. The agreement of this value within 20% of the steady state value suggests that cycling of Ra by vegetation is the dominant process maintaining unsupported Ra in the A-horizon.

For the steady state condition calculated above, the maximum loss rate of Ra from the A-horizon is $8.1 \times 10^{-4} \text{ yr}^{-1}$ or or $0.33 \text{ mBq/cm}^2\text{-yr}$, and the loss due to radioactive decay is $0.17 \text{ mBq/cm}^2\text{-yr}$. These figures indicate that about 33% of the vegetation flux into the A-horizon is lost by radioactive decay, and 67% is lost to deeper horizons or recycled into new vegetation. Although much of the outflux of Ra at steady state must consist of Ra recycled from the A-horizon into new vegetation, at least 33% of the Ra flux into vegetation must be derived from beneath the A-horizon in order to make up for radioactive decay.

Uranium and Th are also cycled through vegetation. However, since $Ra > U$ in the vegetation samples of this study, the effect is less significant for U than for Ra, at least in the soils of the region studied. In addition, the low amount of U and Th in the organic fraction of the A-horizon (Figure 6) indicates that these elements are lost from organic matter more rapidly than Ra. Nevertheless, the slight upward flux of U previously discussed seems likely to result from cycling by vegetation rather than upward movement in capillary moisture or other effects proposed by previous researchers.

Radium at Depth

Excess Ra in the C-horizon is common; at sites NC-1, 14-84, and TN-1, Ra activity in the C-horizon exceeds U activity, to a maximum of 150% Ra/U equilibrium for site TN-1 (Figure 5). In

the C-horizon of NC-1, exchangeable Ra values are high, up to 35%, depending on the extractant, and $Ra/U = 1.25$ (Figure 7).

One possible cause of this excess is the adsorption of dissolved Ra from infiltrating soil water or groundwater onto exchange sites in clays or organic matter in the lowest soil horizons. Many soils in the northeastern U.S. have enhanced levels of exchangeable bases in their deeper horizons possibly resulting from influx of dissolved constituents (including Ra?) in groundwater (Ciolkosz et al., 1990).

The addition of Ra to lower soil horizons by downward percolating soil water probably results from the net loss from the upper soil horizons. In both sites where Ra excess is best expressed in C horizons; upper horizons show Ra loss. In the upper 18 cm of soil at site NC-1 where weathering is most intense, $^{226}Ra/^{238}U$ averages 0.88, and at site TN-1, $^{226}Ra/^{238}U$ in the B horizon is 0.85 ± 0.06 (Figure 5).

The excess Ra derived in the above manner (1.5 Bq/g) is about 20% of the total Ra in the C-horizon, in reasonable agreement with the observed 18% exchangeable Ra there. Thus, the action of percolating soil water could significantly affect the equilibrium condition of the lower soil horizons.

Unsupported "Humic" Radium

An important feature in all soils extracted for Ra is the ubiquitous occurrence of unsupported Ra in the humic (exchangeable + organic) fraction throughout the entire profile, not just in the A-horizons (Table 6, Figure 7). Using the

conservative Ra cycling value of $0.51 \text{ mBq/cm}^2\text{-yr}$, Ra from plants at sites 14-80 and NC-1 can only account for a fraction of the amount (19% and 11%, respectively) of the Ra necessary to maintain unsupported humic Ra in the total profile (Figure 7). Radium in Fe-oxides is close to secular equilibrium with U, $^{226}\text{Ra}/^{238}\text{U} = 0.94$ (Figure 7) and these soils show no evidence of Fe reduction that might release Ra from this source.

The amount of unsupported Ra in the organic matter (av. 26% of the total Ra) can be estimated by subtracting the activity of ^{238}U in the organic matter (generally <2% of the total U) which by radioactive decay would eventually yield some Ra. The proportion of U and Th in organic matter is representative of the occurrence of ^{230}Th and ^{234}U , since both of the latter have long half-lives relative to soil processes.

The amount of unsupported Ra, Ra_u , in the humic fraction per unit area over the entire soil profile is given by:

$$\text{Ra}_u = (\text{Ra}_h - \text{U}_h)\text{TrF}_a \quad (8)$$

where $(\text{Ra}_h - \text{U}_h)$ is the difference in Ra and U activity in the humic fraction (Table 6), T is the thickness of the soil column, and other symbols are as before. From equation (8) the amount of unsupported Ra is 6.5, 11 and 0.76 Bq/cm^2 for sites 14-80, NC-1 and NY-1, respectively (Table 6). Assuming steady state, decay of this unsupported Ra must be countered by transfer of Ra from elsewhere if the amount of organic Ra is to remain constant.

Almost all residual mineral fractions show some Ra depletion relative to U (Tables 5 and 6). The weighted, profile-averaged

$^{226}\text{Ra}/^{238}\text{U}$ in minerals for sites 14-80, NC-1 and NY-1 is 72%, 87% and 91% of secular equilibrium, respectively. Assuming Ra ingrowth from parent Ra over time, minerals at sites 14-80, NC-1 and NY-1 will produce 2.3, 1.3 and 0.23 mBq Ra/cm²-yr, respectively. This ingrowth approximates the excess Ra in humic matter for all three sites, and except for NY-1, is significantly larger than the radium from plant flux (0.51 mBq/cm²-yr). Thus, a significant source of Ra in humic materials, especially at depth, is decay-recoil transfer and solution partitioning of Ra from mineral to organic matter.

It is also reasonable to assume that some of the Ra in the organic matter results from decay of deep plant roots, which are known to have up to 100 times the Ra as the plant shoots (Popova et al., 1964; D'Souza and Mistry, 1970; Gunn and Mistry, 1970). The most conservative conclusion is that much of the Ra in the organic matter results from the action of plants but a significant source could be from minerals shown to have $\text{Ra} < \text{U}$.

CONCLUSIONS

1. Uranium, Th and Ra are enriched in soils relative to parent materials, especially in carbonate soils where a 10-fold enrichment is common. This enrichment is a major factor in the high Ra contents and results in high Rn values in soil gas and homes in carbonate terranes (Tanner, 1986).

2. Radium is strongly enriched in the humic fraction of soils as exchangeable ions and in organic matter, in which it is largely unsupported by U. In particular, it tends to be enriched

relative to U in the A-horizon. This enrichment in the A-horizon is largely caused by cycling through vegetation, which preferentially incorporates Ra relative to U, followed by strong retention of Ra in the resulting humic material, but loss of U and Th from this fraction.

3. Uranium in the A horizon also appears to be affected by cycling through vegetation in humid climates, to leave the A-horizon at most sites with a higher U/Th ratio than deeper horizons. The cycling of U by vegetation probably accounts for $^{234}\text{U} > ^{230}\text{Th}$ and a less pronounced depletion of U than Th from the A-horizon found in previous studies of humid temperate soils.

4. Because of the depleted nature of the surfaces of soils and frequent disequilibrium of Ra from U values, the surface soils (<30 cm depth) should not be considered fully representative of values deeper in the profile. Thus, surveys of U in soils could contain inherent biases not previously recognized.

5. Deeper in the soil profile, decay loss of unsupported Ra in humic materials appears to be accounted for by incorporation of Ra leached from minerals, and perhaps in part by Ra uptake and fixation in roots.

6. In some profiles, $\text{Ra} > \text{U}$ in the C-horizon. This pattern appears to result from fixation of Ra leached from the overlying A- and B-horizons, and from Ra in groundwater.

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CAPTIONS FOR FIGURES

Figure 1. Location of soil sites.

Figure 2. The uranium-238 decay series (after Rose et al., 1979).

Figure 3. Methods used for selective extractions.

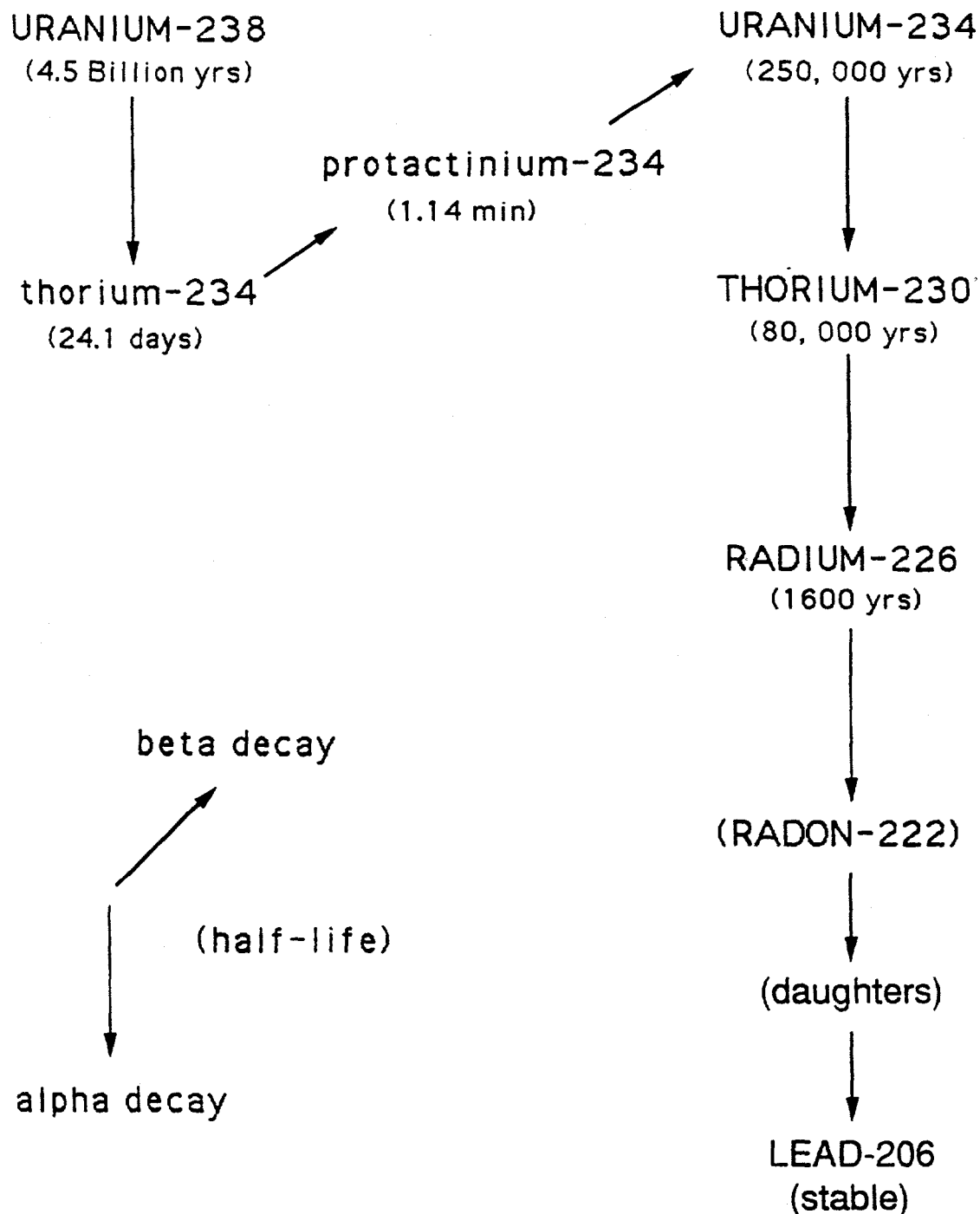
Figure 4. Vertical distribution of nuclides and activity ratios through soil profile. a. Uranium. b. Thorium. c. Radium. d. U/Ra activity ratio. e. U/Ra activity ratio.

Figure 5. Results of selective extractions for U and Th, and some soil properties through soil profiles. a) Site 6-11; b) Site NY-1; c) Site IL-1; d) Site 14-80; e) Site TN-1; f) Site NC-1.

Figure 6. Results of selective extractions of radium through soil profiles. a) Site NC-1; b) Site NY-1; c) Site 14-80; d) Site 14-81.

Figure 7. Estimated fluxes of radium from plants, minerals and organic matter per cm² at site 14-80. Note that flux of Ra from vegetation is too small to compensate for unsupported Ra decay in organics, but flux of Ra from minerals is similar to Ra decay in organics. Model assumes steady state.

URANIUM DECAY SERIES



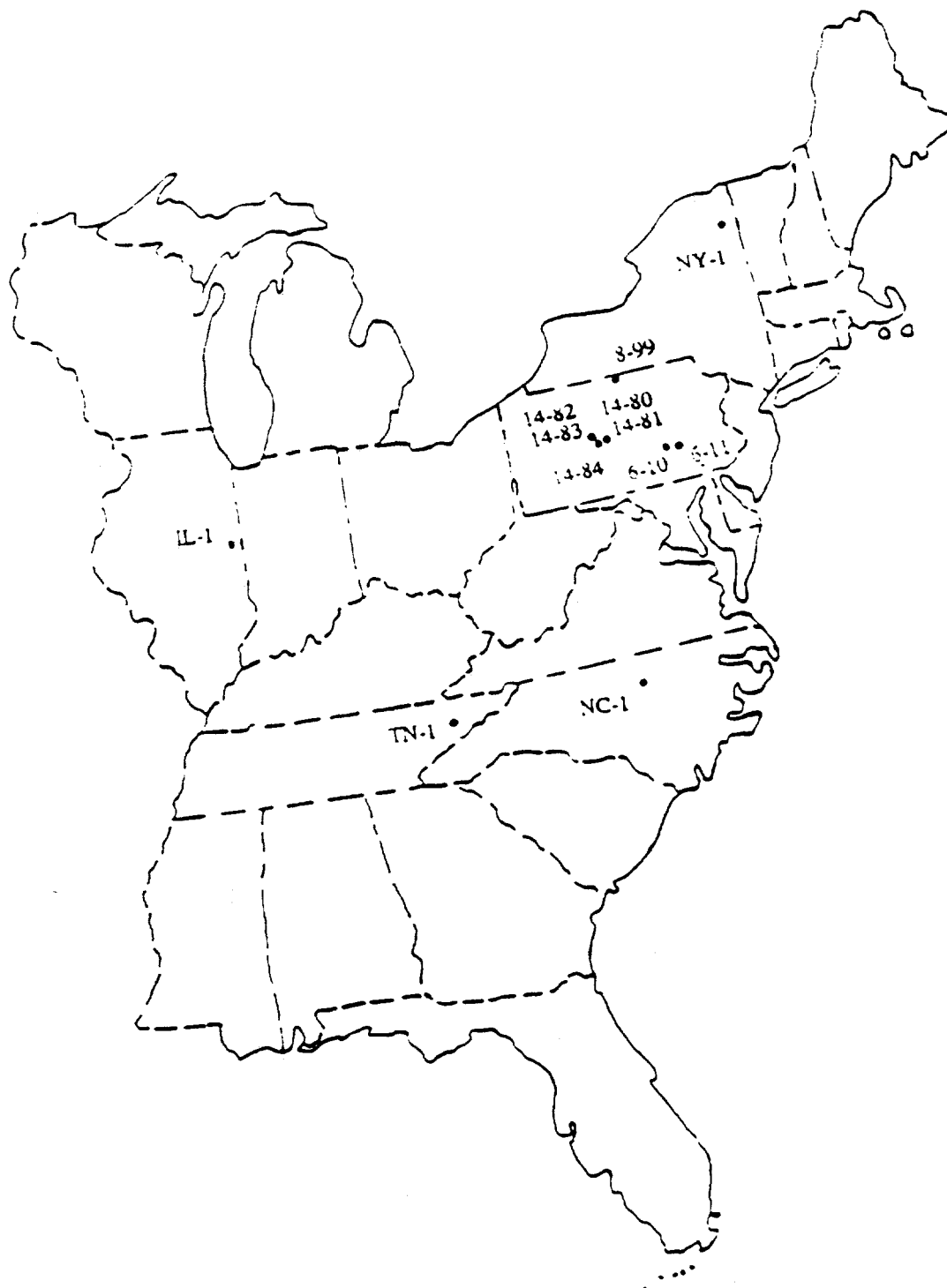
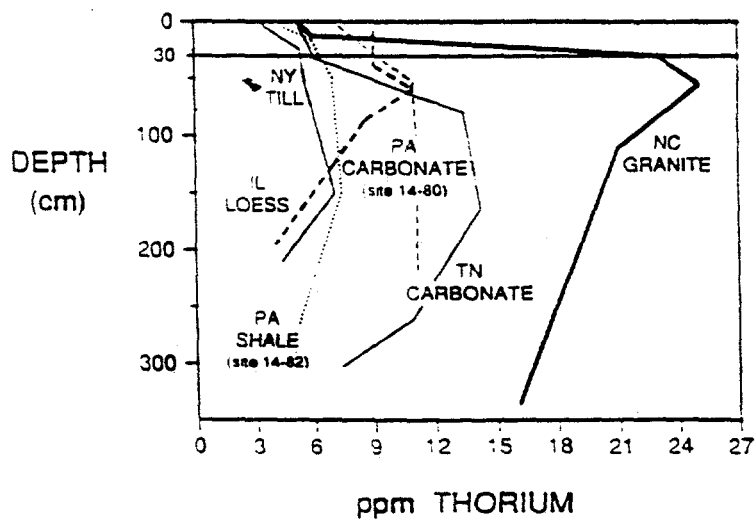
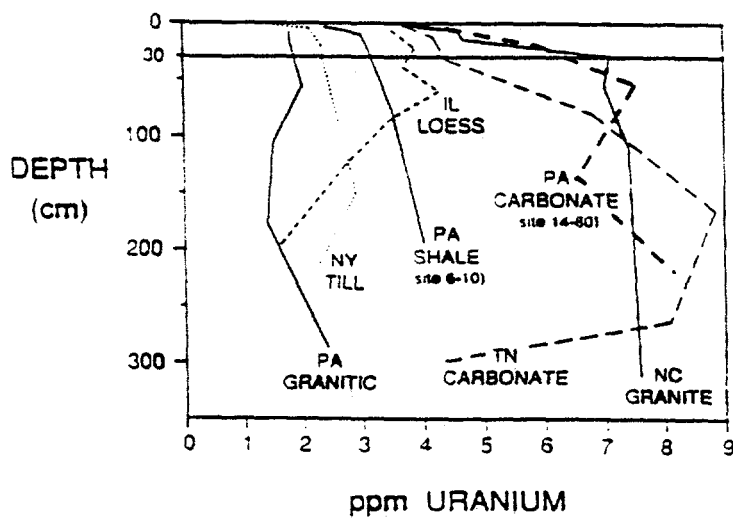


FIG. 2

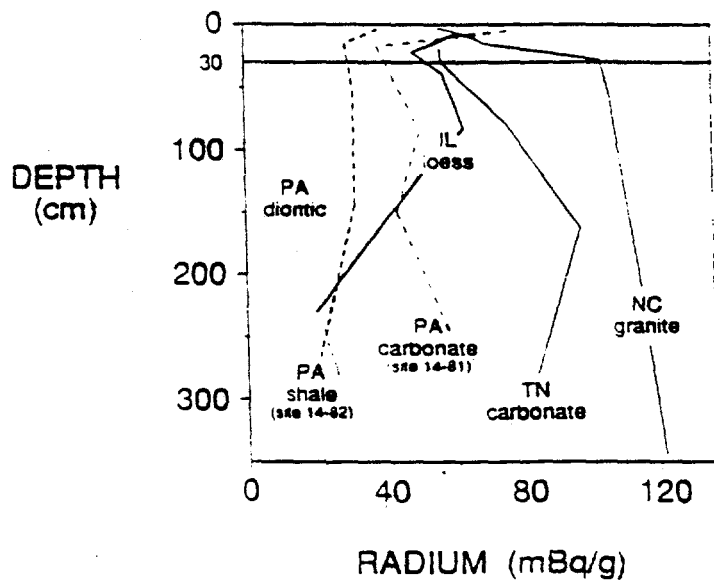
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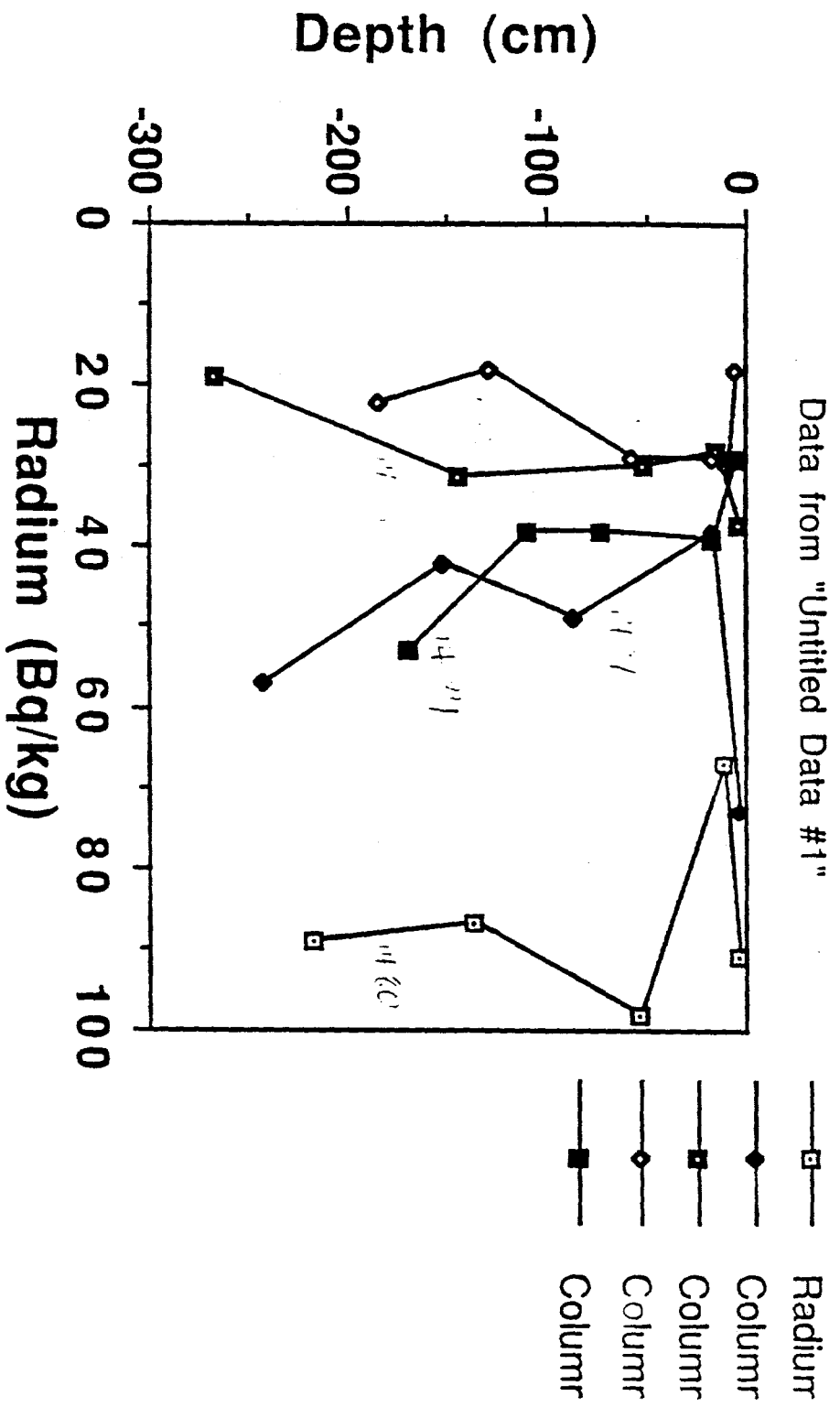


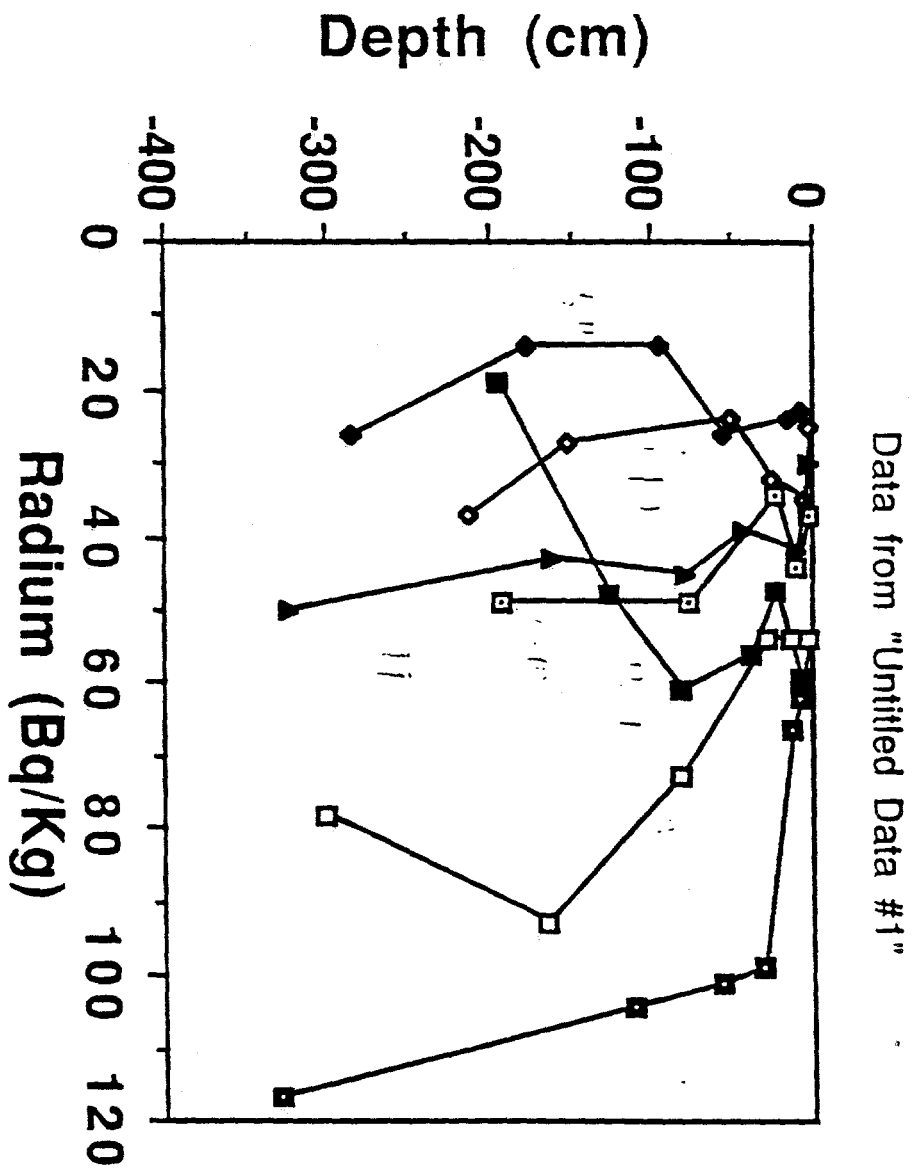
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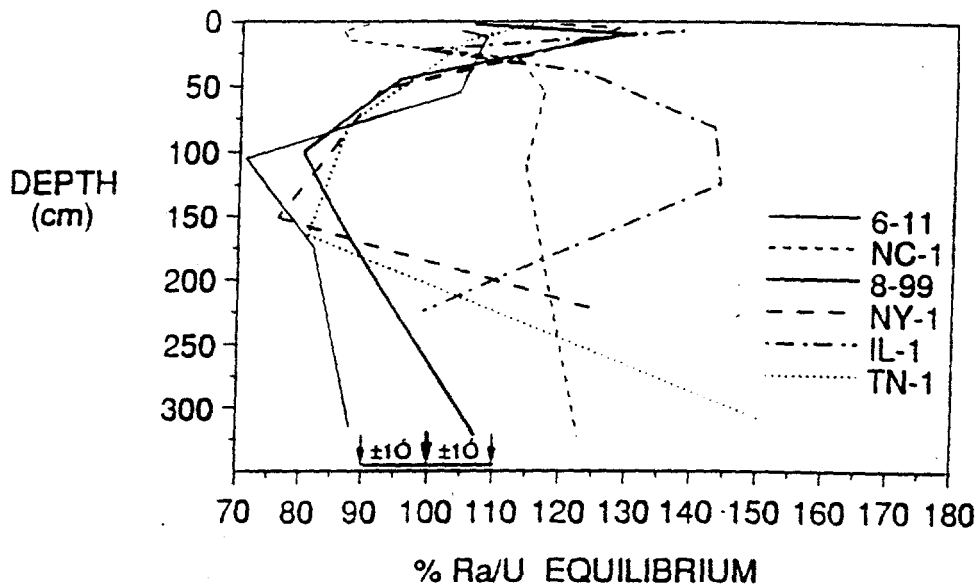




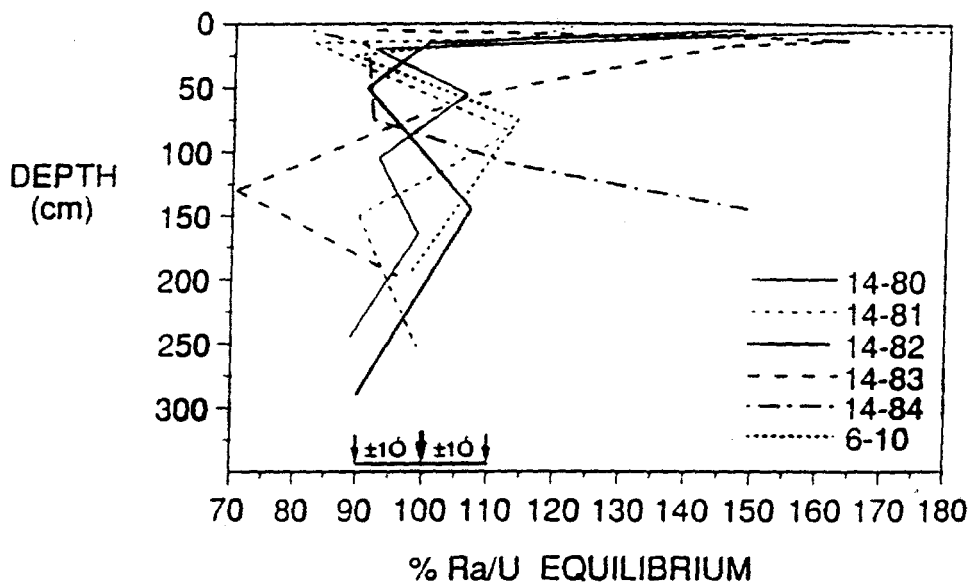


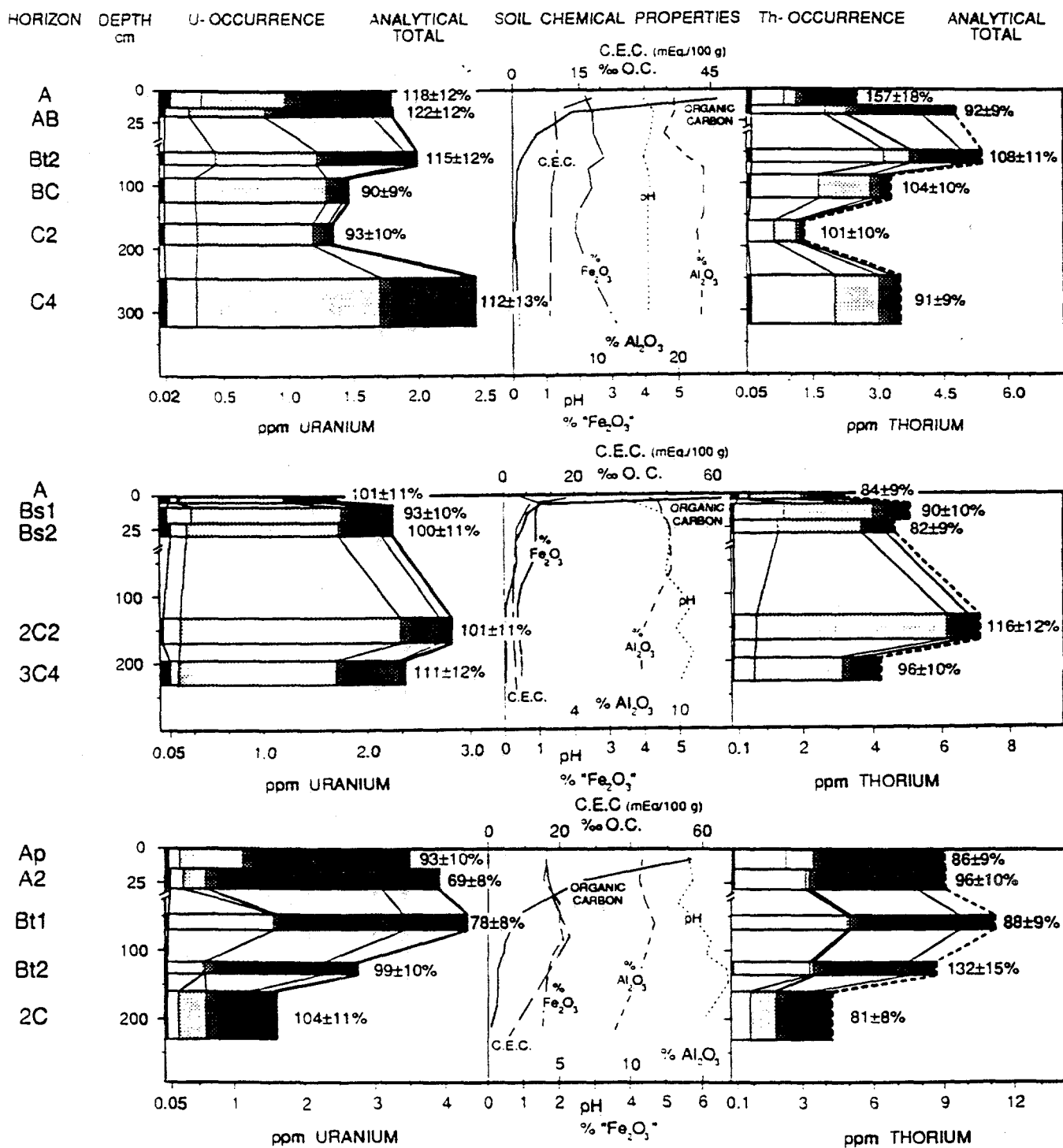
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8-99 R_e

D



E





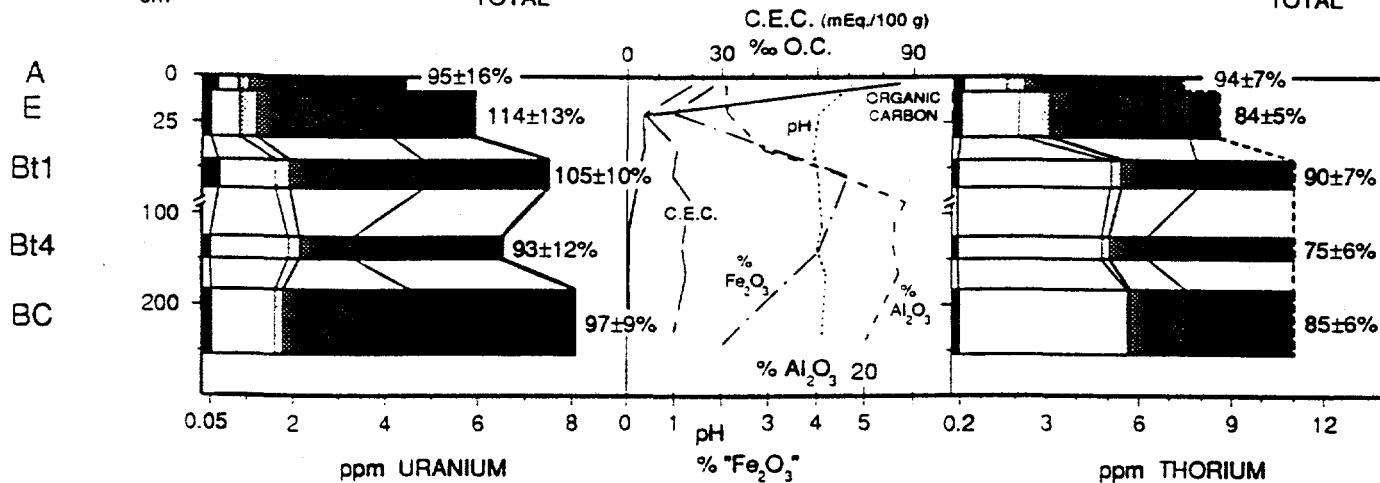
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6-11

B
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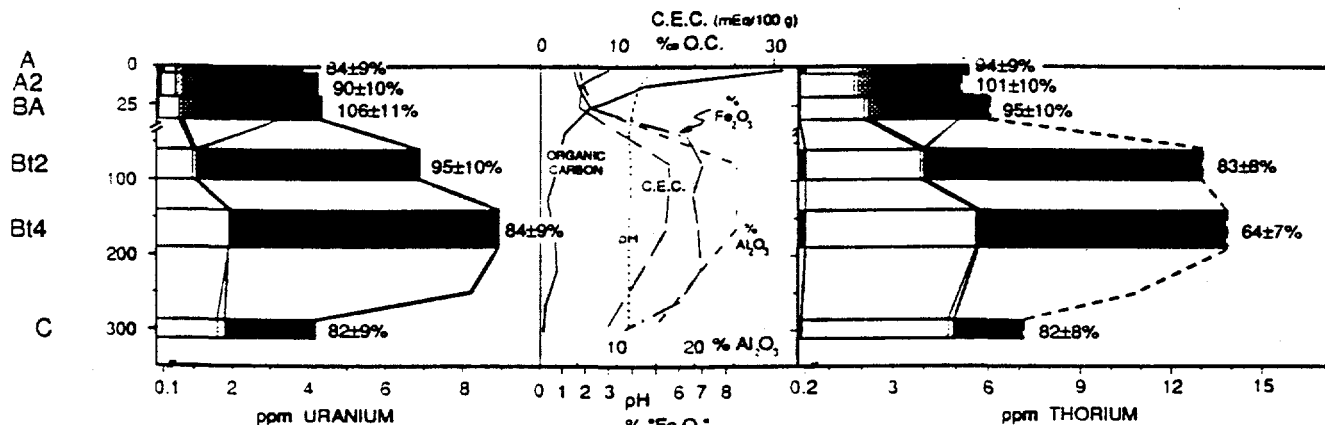
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12-

Fig 56

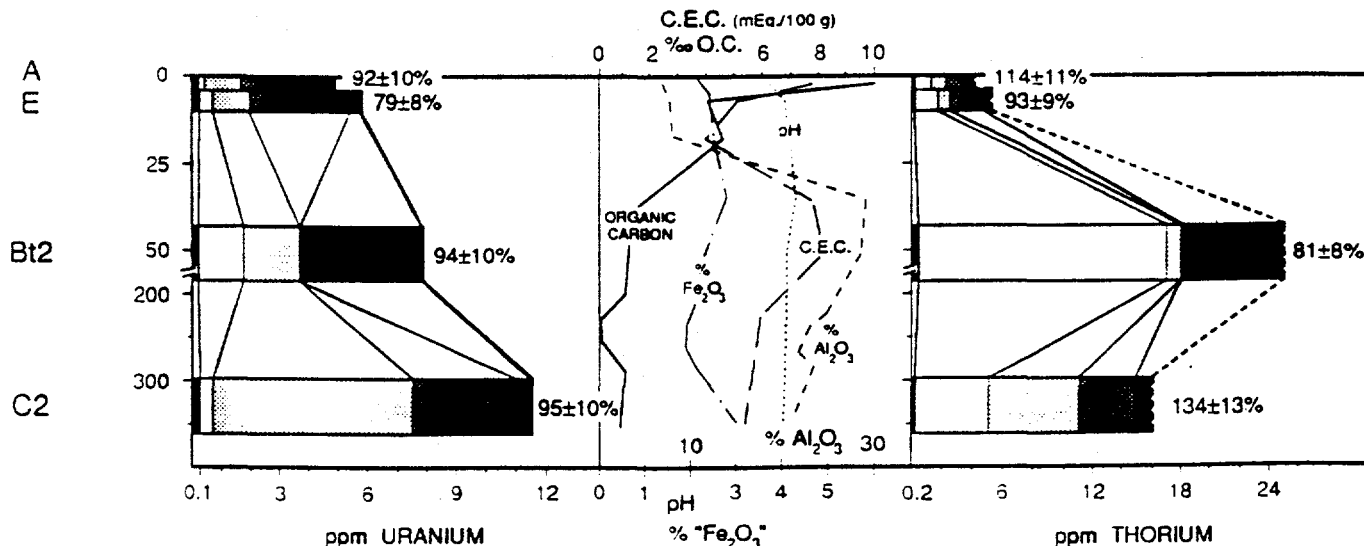
HORIZON DEPTH U-OCCURRENCE ANALYTICAL SOIL CHEMICAL PROPERTIES Th-OCCURRENCE ANALYTICAL



14-8



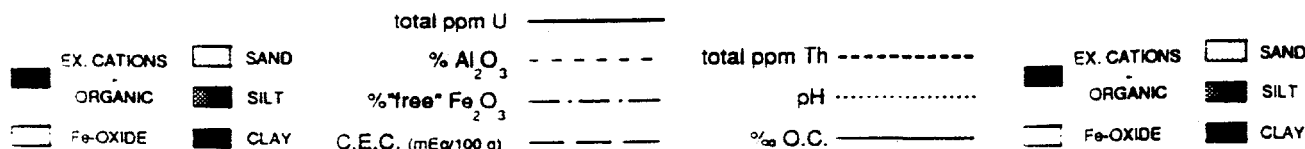
TN-



NC-1

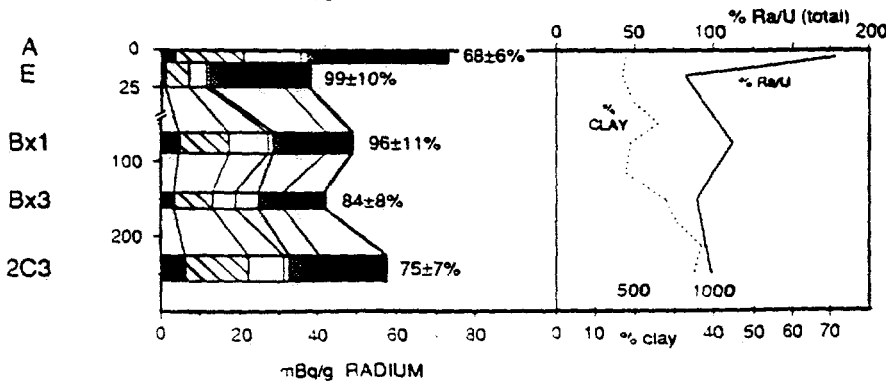
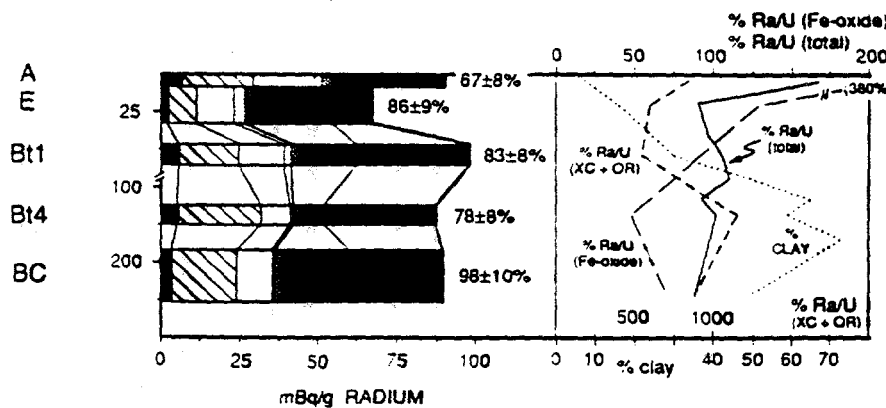
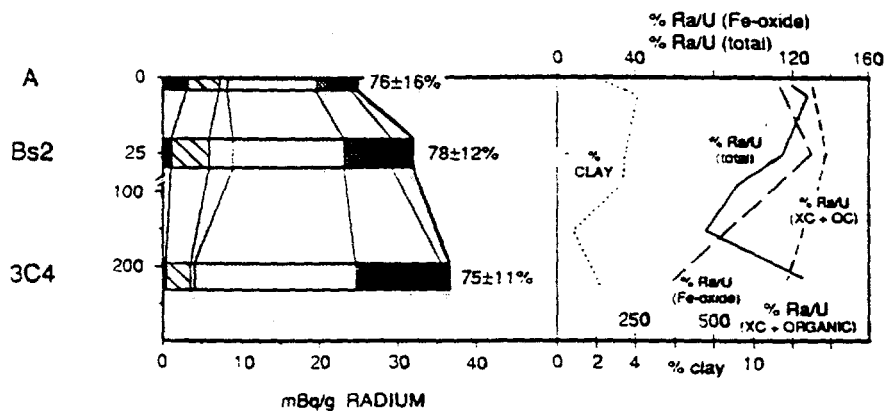
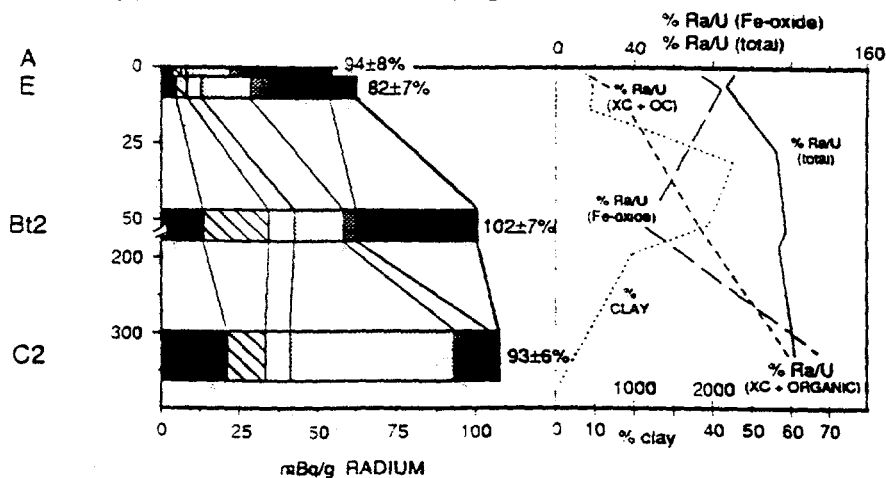
Above uranium values normalized to 100%. Analytical totals indicated.

Above thorium values normalized to 100%. Analytical totals indicated.



FLZ 5

HORIZON DEPTH Ra- OCCURRENCE ANALYTICAL TOTAL OTHER SOIL PROPERTIES



Above radium values normalized to 100%. Analytical totals indicated.

EXCHANGEABLE CATIONS SAND
ORGANIC SILT
Fe-OXIDE CLAY
% clay
total mBq/g Ra
% Ra/U equilibrium (total)

Table 1. Soil site location, parent material, classification, and type.

SITE	LOCATION	LATITUDE	LONGITUDE	PARENT	CLASSIFICATION ¹	SOIL TYPE ²
14-80	Old Fort, PA	40-50-04 N	77-39-20 W	dolostone	Hapludalf	Hagerstown sil
14-81	Old Fort, PA	40-49-55 N	77-39-18 W	dolostone	Fragiudalf	Clarksburg sil
14-82	Philipsburg, PA	40-51-06 N	78-12-43 W	lithic sandstone	Fragiudult	Cookport sl
14-83	Philipsburg, PA	40-51-06 N	78-12-35 W	lithic sandstone	Hapludult	Clymer sl
14-84	Julian, PA	40-52-04 N	77-55-51 W	shaley alluvium	Dystrochrept	Pope sil
6-10	Krumsville, PA	40-35-36 N	75-49-18 W	shale	Hapludult	Bedington sil
6-11	Pricetown, PA	40-25-20 N	75-48-02 W	dioritic gneiss	Hapludult	Glenelg sl
NC-1	Justice, NC	36-02-57 N	78-10-24 W	granite	Hapludult	Appling sil
8-99	E. Athens, PA	41-56-46 N	76-29-30 W	shale till	Fragiochrept	Bath sil
NY-1	Ausable Forks, NY	44-28-21 N	73-43-31 W	granitic till	Haplorthod	Adams sl
IL-1	Champ.-Urbana, IL	40-05-11 N	88-13-52 W	loess, till	Argiudoll	Flanigan sil
TN-1	Jefferson City, TN	36-14-07 N	83-30-06 W	dolostone	Paleudult	Dunsmore sil

¹ USDA nomenclature.

² Soil Survey Staff, 1975. Abbreviations: sl = sandy loam; sil = silt loam.

Table 2. Thickness-weighted average U, Th, Ra, Ra/U, enrichment factors and clay content for soil profiles.

Site	U	Th	Ra	Ra/U	Enrichment factors					Clay	Rock
	ppm	ppm	mBq/g	% eq.	Th	U	Al	Fe	Si	%	
14-80	7.14	10.4	88	99	21	13	16	14	8.3	52	Ls
14-81	3.93	8.0	51	105	16	7	10	11	10	26	Ls
14-82	2.05	5.8	25	99						11	Ss
14-83	1.94	6.9	24	100						11	Ss
14-84	3.14	9.32	37	96	2.3	2.0	1.8	4.0	0.9	19	Sh
6-10	3.48	12.2	46	107	1.3	1.4	1.2	1.1	0.9	27	Sh
6-11	1.93	3.3	21	88	1.6	1.3	1.2	1.2	0.9	17	Gn
8-99	3.61	11.3	45	101	1.1	1.1	0.9	0.9	1.0	18	Sh
NC-1	7.07	19.8	101	116						28	Gr
TN-1	6.06	10.5	75	100	26	15	10	13	5.7	63	Ls
NY-1	2.41	5.36	30	101						3	Gr
IL-1	2.84	7.19	39	111						28	T

Enrichment factor: average soil/bedrock concentrations, using float where bedrock was unobtainable. Rock abbreviations: Ls, limestone; Ss, sandstone; Sh, shale; Gn, dioritic gneiss; Gr, granite; T, till.

Table 3. U, Th, Ra, Th/U, Ra/U, Fe-oxide, organic C, and clay in soil.

SAMPLE	HORIZ.	DEPTH cm	CLAY wt. %	Org. C wt. %	Fe-oxide ¹ wt. %	U ppm	Th ppm	Th/U	Ra mBq/g	Ra/U % Eq.
14-80-1	A	0-8	7.3	8.42	1.8	4.4	7.3	1.7	91	168±4
14-80-2	E	8-33	16	0.40	0.89	5.9	8.5	1.5	67	92±5
14-80-4	Bt1	46-61	51	0.14	4.6	7.5	11	1.5	98	106±5
14-80-7	Bt4	124-150	59	0.07	4.1	6.5	11	1.7	87	102±4
14-80-9	BC	183-254	50	0.05	2.1	8.1	11	1.4	89	89±5
14-81-1	A	0-8	18	6.86	-	3.3	6.3	1.9	73	179±10
14-81-2	E	8-25	17	0.45	-	3.8	6.8	1.8	38	83±5
14-81-5	Bx1	69-104	19	0.01	-	3.6	8.0	2.2	49	113±7
14-81-7	BC	140-163	28	0.09	-	4.0	7.7	2.0	42	90±6
14-81-10	2C3	224-260	35	0.06	-	4.6	9.1	2.0	57	99±5
14-82-1	A	0-8	9.5	9.30	-	2.0	4.6	2.3	37	148±15
14-82-2	E	8-23	14	0.56	-	2.3	6.1	2.7	28	100±10
14-82-4	Bw2	41-61	15	0.14	-	2.7	7.0	2.6	30	91±8
14-82-8	Bx4	132-157	15	0.08	-	2.4	7.4	3.1	31	107±10
14-82-11	2C	229-304	9.5	0.09	-	1.7	5.1	2.9	19	90±11
14-83-3	E	3-8	1.6	0.33	-	1.6	4.6	2.9	18	92±14
14-83-4	Bhs	8-13	7.0	1.18	-	1.5	4.7	3.1	30	164±17
14-83-5	Bs	13-23	8.6	0.65	-	1.7	5.2	3.2	29	145±15
14-83-7	Bt2	43-71	10	0.01	-	2.2	7.2	3.2	29	106±11
14-83-10	BC	122-137	14	0.01	-	2.0	8.1	4.0	18	70±8
14-83-12	C2	168-203	9.2	0.03	-	1.9	7.0	3.7	22	96±12
14-84-1	A	0-10	19	2.49	2.0	2.9	9.2	3.2	29	82±6
14-84-2	Ab	10-25	17	0.92	2.2	3.5	9.9	2.8	39	91±5
14-84-5	Bw3	61-86	22	0.34	2.4	3.4	10	2.9	38	92±6
14-84-7	BC	104-114	16	0.25	1.8	2.7	8.2	3.0	38	113±7
14-84-9	2C2	142+	17	0.41	4.5	2.9	8.6	3.0	53	149±7
6-10-2	A	0-5	20	7.83	2.0	2.4	9.8	4.0	37	122±10
6-10-3	E	5-15	8.2	1.39	2.2	3.0	10	3.3	44	116±8
6-10-4	Bt1	15-30	20	0.77	2.3	3.1	11	3.5	34	89±10
6-10-6	Bt3	58-94	33	0.18	4.3	3.5	13	3.7	49	114±6
6-10-9	BCt	178-208	33	0.14	4.3	4.0	13	3.2	49	98±7
6-11-2	A	0-13	19	4.61	2.3	1.8	2.5	1.4	23	104±15
6-11-3	AB	13-20	14	1.31	2.5	1.8	4.8	2.7	24	108±15
6-11-5	Bt2	46-66	22	0.16	2.8	2.0	5.4	2.7	26	104±9
6-11-7	BC	86-124	16	0.10	2.3	1.5	3.3	2.1	14	71±11
6-11-9	C2	160-193	10	0.05	1.9	1.4	1.3	0.9	14	82±12
6-11-11	C4	246-323	8.1	0.08	3.2	2.4	3.5	1.5	26	88±6

¹ Fe-oxide extractable in Na-dithionite ("free" Fe-oxide- Fe_2O_3).

Abbreviations: HORIZ. = horizon; Eq. = equilibrium; wt. = weight; Org. = organic. Analytical error: clay = ±5%; Fe-oxide = ±5%; U = ±4-9%; Th = ±3-8%; Ra = ±6-12%. Analytical errors quoted at ±1σ.

Table 3 (cont.). U, Th, Ra, Th/U, Ra/U, Fe-oxide, organic C, and clay in soil.

SAMPLE	HORIZ.	DEPTH cm	CLAY wt. %	Org. C wt. %	Fe-oxide ¹ wt. %	U ppm	Th ppm	Th/U	Ra mBq/g	Ra/U % Eq.
NC-1-2	A	0-3	7.7	1.00	0.58	3.8	4.1	1.1	54	90±5
NC-1-3	E	3-10	9.2	0.41	0.76	4.6	5.2	1.1	62	86±4
NC-1-4	EB	10-18	8.8	0.46	0.72	4.7	5.9	1.3	66	87±10
NC-1-5	Bt1	18-43	45	0.19	3.4	7.1	23	3.2	99	113±3
NC-1-6	Bt2	43-69	38	0.13	3.0	7.0	25	3.6	101	117±4
NC-1-8	BC1	89-130	20	0.01	1.8	7.4	21	2.8	104	114±5
NC-1-11	C2	297-361+	3.2	0.12	0.53	7.6	16	2.1	117	125±7
8-99-2	A	0-5	11	4.48	1.2	2.3	5.5	2.4	30	106±10
8-99-3	E	5-15	11	0.98	1.2	2.6	6.9	2.6	42	129±11
8-99-5	Bw2	36-51	14	0.26	1.6	3.4	8.8	2.6	39	95±10
8-99-7	Bx2	61-99	26	0.10	2.3	3.6	11	3.1	45	101±10
8-99-9	Bx4	135-188	21	0.08	2.0	4.0	13	3.3	43	87±9
8-99-12	Bx7	284-361	15	0.09	1.6	3.7	12	3.1	50	110±10
NY-1-2	A	0-3	2.3	6.26	0.6	1.7	3.2	1.9	25	119±19
NY-1-3	Bhs	3-8	3.7	1.03	1.0	2.2	3.4	1.5	35	128±13
NY-1-4	Bs1	8-20	4.1	0.72	0.9	2.2	5.1	2.3	-	-
NY-1-5	Bs2	20-30	3.6	0.48	0.9	2.2	4.6	2.1	32	115±12
NY-1-7	CB	41-64	3.2	0.35	0.7	2.1	5.4	2.6	24	92±15
NY-1-10	2C2	132-170	0.8	0.02	0.4	2.8	7.1	2.5	27	76±10
NY-1-12	3C4	196-231	2.4	0.02	0.5	2.4	4.2	1.8	37	126±13
IL-1-1	Ap	0-15	23	5.61	1.6	3.5	9.1	2.6	59	138±12
IL-1-2	A2	15-30	29	2.55	1.5	3.9	9.1	2.3	47	98±10
IL-1-3	A3	30-48	31	1.49	1.6	3.7	9.1	2.4	56	123±11
IL-1-4	Bt1	48-71	36	0.76	2.0	4.3	11	2.6	-	-
IL-1-5	Bt2	71-94	35	0.58	2.0	3.5	8.6	2.5	61	143±12
IL-1-7	Bt4	117-135	34	0.33	1.8	2.7	7.1	2.6	48	144±12
IL-1-9	2C	160-231	15	0.17	1.5	1.6	4.3	2.7	19	99±20
TN-1-2	A1	0-5	20	3.10	1.5	3.8	5.3	1.4	54	115±11
TN-1-3	A2	5-20	21	1.36	1.7	4.2	5.2	1.3	54	105±9
TN-1-4	BA	20-35	28	0.66	2.2	4.3	6.0	1.4	54	102±9
TN-1-6	Bt2	62-100	76	0.26	7.0	6.8	13	1.9	73	87±7
TN-1-8	Bt4	138-188	77	0.17	6.8	8.9	14	1.5	93	84±5
TN-1-10	CB	230-288	74	0.08	6.0	8.2	11	1.3	-	-
TN-1-11	C	288-313	43	0.05	3.7	4.2	7.1	1.7	78	151±15

¹ Fe-oxide extractable in Na-dithionite ("free" Fe-oxide- Fe_2O_3).

Abbreviations: HORIZ. = horizon; Eq. = equilibrium; Wt. = weight; Org. = organic. Analytical error: clay = ±5%; Fe-oxide = ±5%; U = ±4-9%; Th = ±3-8%; Ra = ±6-12%. Analytical errors quoted at ±1σ.

Table 4 • U, Th, Ra, and ash content in vegetation.

SAMPLE	SITE	% ASH	U (CF) ppm	Th (CF) ppm	Ra (CF) mBq/g	Th/U W. R.	Ra/U A. R.
- LIVE PLANTS -							
<i>Ailanthus altissima</i>	TN-1	7.2	0.15 (0.03)	0.21 (0.02)	120 (1.6)	1.4	65
mixed shrubs #1	TN-1	4.3	0.07 (0.01)	0.33 (0.03)	54 (0.71)	4.5	62
<i>Viburnum acerifolium</i>	6-10	6.8	0.20 (0.07)	0.34 (0.03)	97 (2.1)	1.7	39
<i>Cornus</i> sp.	TN-1	7.8	0.43 (0.08)	0.39 (0.04)	43 (.57)	0.9	8.2
mixed shrubs #2	14-82.3	4.4	0.19 (0.10)	0.44 (0.06)	14 (.58)	2.3	6.0
<i>Solidago</i> sp.	14-84	7.8	0.80 (0.76)	0.55 (0.06)	48 (1.1)	0.7	4.9
<i>Betula lutea</i>	8-99	2.4	0.48 (0.22)	2.5 (0.26)	80 (1.8)	5.2	4.5
mixed shrubs #3	6-11	12*	0.30 (0.17)	0.89 (0.23)	13 (.58)	3.0	3.4
mixed shrubs #4	NY-1	4.3	0.13 (0.08)	0.63 (0.14)	52 (1.7)	5.0	3.4
<i>Pinus strobus</i>	8-99	1.6	0.44 (0.26)	2.8 (0.60)	15 (0.33)	6.3	2.7
<i>Cornus</i> sp.	14-80	5.1	0.75 (0.10)	0.24 (0.02)	9.3 (0.10)	0.3	2.5
<i>Pinus strobus</i>	NY-1	1.8	1.1 (0.67)	2.3 (0.49)	22 (0.73)	2.1	1.6
<i>Cladonia</i> sp.	NY-1	2.5	1.7 (1.0)	3.4 (0.72)	31 (1.0)	2.0	1.5
<i>Betula lutea</i> (root)	8-99	2.0	1.9 (0.85)	5.4 (0.57)	33 (0.75)	2.9	1.4
<i>Dicranum</i> sp.	NY-1	8.5	2.7 (1.6)	4.8 (1.0)	20 (0.68)	1.8	0.62
grasses	IL-1	17*	7.2 (3.1)	9.3 (1.1)	3.8 (0.10)	1.3	0.04
AVERAGE (n = 16)	-	6.2	1.2 (0.54)	2.2 (0.33)	41 (0.91)	2.6	2.8
- DEGRADED PLANT MATERIALS -							
hard wood fragment	TN-1	0.5	1.7 (0.15)	2.6 (0.24)	270 (4.1)	1.6	13
leaf fall (mixed)	14-80	6.7	1.1 (0.15)	1.0 (0.10)	110 (1.2)	1.0	8.5
O _e	TN-1	12*	3.4 (0.65)	4.9 (0.45)	90 (1.2)	1.5	2.1
litter	NY-1	3.5	6.6 (3.9)	24 (5.2)	140 (4.7)	3.6	1.7
O _i	NC-1	5.7	24 (3.4)	47 (2.4)	370 (3.9)	1.9	1.2
O _e -O _i	6-10	13*	1.9 (0.65)	9.7 (0.85)	90 (1.2)	5.2	1.0
O _e	8-99	8.4	1.9 (0.87)	4.3 (0.45)	18 (0.51)	2.3	0.75
leaf fall (<i>Quercum</i> sp.)	6-10	8.3	2.0 (0.69)	5.1 (0.45)	17 (0.72)	2.6	0.69
O _i	14-83	14*	6.4 (3.5)	13 (1.9)	34 (1.4)	2.0	0.43
O _e	NY-1	6.8	5.6 (3.3)	20 (4.3)	19 (0.66)	3.5	0.28
AVERAGE (n = 10)	-	~9	5.5 (1.7)	13 (1.6)	120 (2.0)	2.5	1.7

Abbreviations: CF = concentration factor (ash value/average soil value at that site); W. R. = weight ratio; A. R. = activity ratio; n = number of samples; O_e = hemic organic surface layer; O_i = fibric organic surface layer. Latin names for single plant species in italics. Asterisk indicates that mineral matter may not have been completely removed from sample (high ash totals >10%). All samples harvested mid- to late- summer. Analytical error (1σ): U = ±3-12%; Th = ±3-10%; Ra = ±5-9%.

Table 5. Profile-averaged occurrence of U, Th, and Ra in soil phases.

SITE	SOIL HUMIC MATERIALS ¹				"Free" Fe-OXIDES ²				PEDOGENIC ³				MINERAL MATTER ⁴											
	U (% of total occurrence)	Th	Ra	U (conc. ppm)	Th	U (% of total occur.)	Th	Ra	Ra/U (% Eq.)	U (conc. ppm)	Th	Ra	U	Th	Ra	Sand	Silt	Clay						
14-80	3.3	1.7	27	20	15	17	42	14	95	57	200	21	43	41		4.3	4.6	2.7	43	25	29	32	27	25
14-81	-	-	34	-	-	-	-	16	-	-	-	21	-	50		-	4.8	-	-	24	-	-	-	21
14-84	<1.4	<1.2	-	<3.0	<7.6	16	20	-	-	19	71	17	22	-		22	22	-	48	42	-	12	15	-
6-10	<1.1	<2.2	-	<2.4	<16	24	22	-	-	19	75	25	24	-		29	28	-	33	38	-	13	11	-
6-11	<1.0	<1.0	-	<1.4	<3.8	15	50	-	-	11	60	16	51	-		50	28	-	29	14	-	4.5	6.4	-
NC-1	1.5	1.2	28	15	21	9.1	40	5.9	109	62	780	11	41	34		35	19	36	36	27	19	18	12	11
8-99	<1.6	<0.4	-	<6.2	<8.7	9.0	19	-	-	18	130	11	19	-		25	22	-	52	42	-	12	17	-
IL-1	<1.5	0.6	-	<3.8	1.9	16	27	-	-	25	120	18	28	-		12	14	-	52	42	-	18	16	-
TN-1	<0.3	<0.7	-	<2.5	<11	20	41	-	-	26	90	20	41	-		3.1	1.7	-	8.9	6.7	-	68	50	-
NY-1	1.4	<0.7	13	5.0	<7.5	6.1	16	3.3	77	29	180	7.9	17	16		67	64	52	22	13	27	3.0	5.7	5.0
average	<1.5	<1.1	26	<6.6	<10	15	31	10	94	30	190	16	31	35		27	23	24	36	28	25	20	18	16

¹ Concentration of U, Th in soil humics = quantity of U, Th in exchangeable cation + organic fractions + 2 x (organic C).

² Concentration of U, Th in "free" Fe-oxides = quantity of U, Th in dithionite- + hydroxylamine extractable fractions + ("free" Fe-oxide).

³ Pedogenic U, Th = sum of exchangeable cation + organic + "free" Fe-oxides.

⁴ Abundance of U, Th, and Ra expressed as % of total occurrence.

Abbreviations: conc. = concentration; occur. = occurrence; Eq. = equilibrium; - = sample not analyzed. Analytical error U = $\pm 4-12\%$; Th = $\pm 4-10\%$; Ra = $\pm 6-18\%$. Errors quoted at $\pm 1\sigma$. All values are weighted averages of 3-7 horizons per site.

Table 6. Calculation of unsupported Ra and Ra fluxes within entire soil profiles.

SITE	DEPTH cm	BULK DENSITY g/cm ³	% SOIL (<2 mm)	Ra - U humics mBq/g	R Bq/cm ²	Excess U (minerals) Bq/cm ²	Ra loss L mBq/cm ² -yr	Ra excess E mBq/cm ² -yr	Plant Ra
14-80	250	1.42	87.8	20.9	6.5	5.5	2.8	2.3	0.51
NC-1	360	1.35	83.0	27.3	11	3.1	4.8	1.3	0.51
NY-1	230	1.77	53.6	3.48	0.76	0.53	0.32	0.23	0.51

R = amount of Ra unsupported by U or Th-230; L = loss of unsupported Ra by radioactive decay;
 E = radium produced by excess U (assuming secular equilibrium with Th-230); plant Ra = turnover of Ra
 in plant tissue (cycling); excess U assumes steady-state condition in minerals where Ra < U; values are
 72-92% of Ra/U equilibrium. Humics = cation-exchangeable + organic matter fractions.

APPENDIX B

Manuscript of
PATTERNS OF DISEQUILIBRIUM AMONG ^{238}U , ^{234}U , ^{230}Th AND ^{226}Ra
IN TOTAL SOIL AND SOIL PHASES IN TWO SOIL PROFILES

Condensed from the M.S. thesis of John R. Hogue

**PATTERNS OF DISEQUILIBRIUM AMONG ^{238}U , ^{234}U , ^{230}Th AND
 ^{226}Ra IN TOTAL SOIL AND SOIL PHASES IN TWO SOIL
PROFILES**

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ABSTRACT

Samples from two soil profiles developed on
Pleistocene till in New York (NY-1) and Illinois (IL-1)
were analyzed for ^{238}U , ^{234}U , and ^{230}Th using an alpha-
spectrometric analytical procedure modified to accomodate
soils. In addition, samples from selected horizons were
selectively leached and fractionated (i.e., exchangeable
cations, organic matter, free oxides, sand, silt and clay)
to determine the speciation of ^{238}U , ^{234}U and ^{230}Th , and
their variation with horizon. Radium had been previously
determined by Greeman on the same samples.

The daughter/parent activity ratios in NY-1, a typical
alfisol, vary widely between the horizons, indicating
independent geochemical behavior of ^{234}U , ^{230}Th and ^{226}Ra .
 $^{226}\text{Ra}/^{230}\text{Th}$ ratios of total soil samples range from 0.63 to
2.1, $^{230}\text{Th}/^{234}\text{U}$ ratios range from 0.94 to 1.34 and $^{234}\text{U}/^{238}\text{U}$
ratios from 0.97 to 1.12. (Correct for just NY-1) The
selective phase analyses show disequilibrium ratios up to
23 for $^{226}\text{Ra}/^{230}\text{Th}$ in organic matter and commonly 2 to 4 for

$^{230}\text{Th}/^{234}\text{U}$ in Fe-oxides and B-horizon clay. Selective uptake of ^{226}Ra by vegetation leads to depletion from the B-horizon and enrichment in the A-horizon relative to ^{230}Th .

In IL-1, a typical mollisol, the vegetation does not significantly redistribute these nuclides between horizons, and known uranium, thorium, and radium sorbers (e.g., organic matter, iron-oxides, and clays) are relatively abundant in all horizons. As a result, the daughter/parent ratios are more uniform with depth. $^{230}\text{Th}/^{226}\text{Ra}$ is within 14% of equilibrium for all measured horizons, and $^{230}\text{Th}/^{234}\text{U}$ ratios consistently exceed unity (between 1.17 and 1.68). The greater enrichment of ^{230}Th over ^{234}U in this profile relative to NY-1 is attributed to selective adsorption by the abundant fine-grained parent material which minimizes Th leaching.

INTRODUCTION

Radon in soil gas, generated by radioactive decay of ^{226}Ra in soil, is the major source of the radon hazard in homes. The abundance of radon in soil gas depends partly on the abundance of its parent, ^{226}Ra . In turn, the abundance of Ra depends on its precursors, ^{238}U , ^{234}U and ^{230}Th (Figure 1). Greeman (1992) showed disequilibrium between ^{226}Ra and ^{238}U in several soils from the eastern U.S., in particular relatively young soils developed on Pleistocene till in New York and Illinois. He postulated that this disequilibrium results from geochemical differences between the parent uranium and thorium nuclides. The half-lives of

^{234}U , ^{230}Th and ^{226}Ra (248,000, 80,000 and 1600 years, respectively) are long enough for significant chemical fractionation to occur in soil-forming processes.

Measurements of ^{238}U , ^{234}U and ^{230}Th in soil profiles, developed on a variety of parent materials, have been published by Hansen and Stout (1968), Lowson et al., (1986), Megumi (1979) and Rosholt et al. (1966). However, these studies lack data on ^{226}Ra , and most are on soils from semi-arid regions.

In this investigation, samples from well characterized soil profiles in the eastern U.S., previously analyzed for ^{226}Ra by Greeman (1992) have been analyzed for ^{238}U , ^{234}U and ^{230}Th in order to investigate the extent and major sources of disequilibrium between U and Ra. Selective extractions for exchangeable, organic and Fe-oxide fractions were also made to evaluate processes causing disequilibrium. The available methods for determining these nuclides required considerable modification in order to attain satisfactory results on these soils and selective extracts.

In a radioactive decay series with a very long-lived parent such as ^{238}U , radioactive equilibrium implies that the activities (A, disintegrations per unit time in a given mass) of parents and daughters are equal:

$$A^{238} = A^{234} = A^{230} = A^{226} \quad (1)$$

where the superscript indicates the nuclides ^{238}U , ^{234}U , ^{230}Th and ^{226}Ra . This is equivalent to saying that:

$$N^{238}\lambda^{238} = N^{234}\lambda^{234} = N^{230}\lambda^{230} = N^{226}\lambda^{226} \quad (2)$$

where N is the number of atoms of the superscripted nuclide and λ is the decay constant of the nuclide. In order for equilibrium to occur, the sample in question must have remained chemically undisturbed for a period of many half-lives of the daughter nuclide. Because of the long half-lives of ^{234}U , ^{230}Th and ^{226}Ra , it is entirely possible that the nuclides have been separated chemically during weathering and soil formation, and that re-equilibration has not had time to occur. It is also possible that although a given small volume of soil shows equilibrium, the U-series nuclides have been redistributed among the phases of the soil, so that some forms or their decay products have enhanced mobility, as indicated by Greeman and Rose (1996) for ^{226}Ra and ^{222}Rn . To examine this possibility, we have examined the distribution of the nuclides among exchangeable ions, organic matter, Fe-oxides, sand, silt and clay.

A number of chemical processes might cause disequilibrium between the U and Th isotopes. Uranium in oxidizing environments is generally much more soluble than Th, so may be leached from soil or transferred to a different phase more easily. Also, Th is generally more strongly adsorbed than U, and is expected to be much less mobile for this reason. By the time ^{234}U is formed, two alpha decays have occurred, and the recoil of the daughter

nuclide may have damaged the crystal lattice of the host mineral, allowing easier dissolution and mobility of ^{234}U than ^{238}U (Fleischer and Raabe, 1978). Vegetation may take up one element more than others and translocate the nuclides into the above-ground plant, later to be deposited at the soil surface, as found for Ra (Greeman, 1992; Greeman et al., manuscript). Circulating ground waters may bring U into the soil in question. Under some conditions, organic complexing may preferentially enhance solubility of Th. All of these processes can lead to disequilibrium.

Previous Studies of ^{238}U , ^{234}U and ^{230}Th Disequilibrium in Soils

Rosholt et al. (1966) found an excess of ^{230}Th in soils developed on sands, shales and till, with an average $^{230}\text{Th}/^{238}\text{U}$ of 1.3. Also, ^{234}U was noted to be depleted to an average $^{234}\text{U}/^{238}\text{U}$ value of 0.9, with most samples having ratios between 0.82 and 0.89. In a desert soil developed on a trachite, ^{234}U in the C horizon was depleted to 42% of equilibrium with ^{238}U , though the parent rock was close to equilibrium, and ^{230}Th was at 90% of equilibrium. This pattern indicates preferential leaching of ^{234}U relative to ^{238}U , but nearly complete retention of ^{230}Th . Rosholt et al. (1966) also proposed upward transfer of U by capillary movement of moisture.

Hansen and Stout (1968) observed near equilibrium for the U isotopes and an enrichment of ^{230}Th up to 30% in residual and alluvial soils in a semi-arid cool climate.

Their mechanism includes early selective leaching of ^{234}U but later addition of ^{234}U from solutions moving upward by capillary movement. Megumi (1979) found excesses of ^{234}U over ^{238}U and postulated addition of ^{234}U from groundwater enriched in this nuclide.

Lowson et al. (1986) found the activity of ^{230}Th to be approximately twice that of ^{238}U in a lateritic soil. They considered this a result of the greater mobility of uranium.

These previous studies indicate that disequilibrium is not uncommon in soils and probably arises by several processes. However, these authors did not tie the U and Th data to ^{226}Ra , and most did not study soils of humid temperate regions, as is done in the present study.

METHODOLOGY

Total Soil Analysis

Many procedures have been developed for decomposition of natural samples for U and/or Th analysis (Lally, 1992; Rosholt et al., 1966; Ku, 1970; Anderson and Fleer, 1982; Burnett and Hull, 1993; Greeman, 1992; McCabe et al., 1992). However, most involve decomposition of igneous rocks, ore bearing rocks or marine sediment, which have a chemistry distinctly different from soils. In our early attempts to analyze soils by $\text{HF-HNO}_3\text{-HClO}_4$ leaches, recovery of tracer was poor. Several new steps were investigated to obtain acceptable recoveries of these nuclides from soil

samples. After extensive trials, the method summarized in Figure 2 was developed.

Dissolution of Soil

The dry samples are ground into a fine powder. Samples are weighed in 2g portions into 50 ml Teflon beakers and spiked with 10 pCi of ^{232}U - ^{228}Th tracer (From: Isotope Products Labs, Burbank, CA). The sample is then refluxed overnight with 10 ml concentrated nitric acid (HNO_3), 20 ml concentrated hydrofluoric acid (HF), and 3 ml of deionized water in a covered Teflon beaker. This step oxidizes the organic matter and decomposes the Fe- and Mn-oxides, clay minerals, carbonates, and most silicates. In the morning, the beaker is uncovered and taken to dryness at about 250°C ; then 2 ml 70% HClO_4 and 5 ml 48% HF are added, and it is covered again for an overnight digestion.

Fluorides left from the HF treatments led to low recovery of Th in preliminary experiments, as Th is coprecipitated with insoluble complexes formed between fluorides and aluminum, along with the alkali and rare earth metals (Korkisch and Dimitriadis, 1973). The high Al content of soils relative to parent rocks accentuates this process in soils. Subsequent additions of HClO_4 taken to fumes failed to completely remove the fluoride. Treatments with hydrochloric acid (HCl) to dissolve green fluoride crystals of iron as described by McCabe et al. (1992) improved Th recoveries. However, the fluoride was not

completely removed until it was evaporated in the presence of excess H_3BO_3 , as described by Korkisch and Dimitriadis (1973). Therefore, the HClO_4 -HF step is followed by the addition of 5 ml 6N HCl , 25 ml H_2O , 2 ml 70% HClO_4 , and 5 g H_3BO_3 . The beaker is covered and heated for an hour, then evaporated to HClO_4 fumes. Two more additions and evaporations of 2 ml HClO_4 follow to ensure the complete removal of HF.

Subsequently, the sample is dissolved in 25 ml 1:1 HCl and the excess H_3BO_3 is removed by centrifuging and decanting the supernatant back into the teflon beaker. The H_3BO_3 settled at the bottom of the tube is rinsed and centrifuged 3 to 4 times with 5 ml 1:1 HCl , decanting into the above beaker, to ensure nearly complete removal of the radionuclides. The beaker is then taken to dryness, the residue is dissolved in 4 ml concentrated HCl , then diluted to 2M by adding 20 ml H_2O .

The actinides are then coprecipitated with $\text{Fe}(\text{OH})_3$ by the addition of NH_4OH to pH=7, using the iron from the sample as the carrier. The sample is centrifuged and the supernatant, which contains alkali and alkaline earth elements (Burnett and Hull, 1993), including radium, is decanted and discarded. The precipitate is washed with 50 ml H_2O , and re-centrifuged, again decanting and discarding the supernatant. The precipitate is dissolved with 1.5 ml 16M HNO_3 and brought to 2M HNO_3 by diluting to 12 ml with

H₂O. Centrifuging at 2000 rpm for 5 min. settles any residue present in the tube in order to eliminate plugging of the TRU-Spec column in the steps that follow. If residue is present, the washing is repeated and the supernatant is combined with that of the preceeding step. It was during this step that the Al-fluorides, which complexed Th prior to the incorporation of the H₃BO₃ step, became obvious.

Ion Exchange Column Separations

Ion exchange column separations are performed according to the procedure described by Burnett and Hull (1993) with only minor modifications. The solution from the previous step is passed through a TRU-Spec™ ion exchange column (EICHROM, Inc., Darien, IL), which is pre-rinsed with 10 ml 2M HNO₃. The eluant, containing iron, aluminum, polonium, bismuth, lead, etc., is discarded. The column is rinsed with 15 ml 2M HNO₃, again discarding the eluant. Uranium and thorium are eluted with 10 ml of 0.1M ammonium bioxalate (NH₄HC₂O₄), collecting the eluant in a 100 ml graphite-bottomed PFA teflon beaker.

The eluant is taken to dryness. The oxalate crystals are destroyed by four or more treatments with 2 ml concentrated HNO₃ and 2 drops 30% H₂O₂ followed by evaporation to dryness. The radionuclides are then dissolved in 10 ml 10 M HCl, in preparation for passage through a second cation exchange column for U and Th separation.

The second cation exchange column is a glass tube with an internal diameter about 7 mm and a length of 10 cm, filled with Bio-Rad™ AG1-X8 (100-200 mesh) exchange resin, and pre-rinsed with 10 ml 10M HCl. The radionuclides, dissolved in 10M HCl, are loaded into the column. The eluant, containing Th, is collected in a 100 ml PFA teflon beaker. The remaining Th is eluted by rinsing the column with 20 ml 10 M HCl. Subsequently, U is eluted with 20 ml 0.1 M HCl into a separate 100 ml PFA teflon beaker. The U and Th solutions are taken to dryness, and are ready for electrodeposition.

Electrodeposition

The electrodeposition techniques are based on the INS (Institute of Nuclear Science) procedure described by Burnett (1992) and McCabe et al. (1992). The source preparation utilizes a rotating disc electrodeposition unit with a temperature controlled electroplating cell (Burnett, 1992, p.9). The plating solutions are prepared in the 100 ml PFA teflon beakers by dissolving the radionuclides in 3 ml OPTIMA™ H₂SO₄, and diluting with 30 ml H₂O. The pH is adjusted to 3.2-3.7 for U and 3.1-3.3 for Th, by addition of SUPRAPURE™ NaOH solution. The temperature of the beaker is held constant at 60 °C in a water bath during electrodeposition. Plating is onto 24 mm stainless steel cathodes (From: Institute of Geological and Nuclear Sciences, Lower Hutt, New Zealand) rotating at 3000 rpm, in continuous contact with the surface of the plating solution

in the cell for 1.5 hr (U) to 2 hr (Th). The cell voltage is held at approximately 4 V with a current of 100 ma. These preparations result in thin, uniform deposits of U and Th onto 24 mm disks which are then counted by alpha spectrometry.

Counting by Alpha Spectrometry

The electroplated U and Th disks are counted in 600 mm² silicon surface barrier detectors (EG&G ORTEC, Oak Ridge, TN, Model # TU-024-600-AS). The detector is evacuated to 20 mm Hg air pressure during counting. The detectors were calibrated with a 2,350 Bq ²⁴¹Am source, yielding efficiencies consistently between 20 and 22 %. A multichannel pulse-height analyzer converts pulse energy outputs from the detector into count frequency versus energy histograms (Figure 3). Peak energy values are correlated with major radiation alpha energies for the members of the uranium and thorium decay series (Table 1), in order to determine which nuclide is represented in each peak.

The integral counts for each peak are obtained from these analyzers, and activity values and nuclide recoveries for the uranium series nuclides are calculated based on tracer yield, and an assumed condition of secular equilibrium between the ²³²Th and ²²⁸Th members of the thorium decay series in the soil sample. The thorium decay series, pictured in Figure 4, includes alpha- and beta-emitting radionuclides whose atomic mass numbers are

multiples of 4. The heaviest naturally occurring member of this series is ^{232}Th , which produces ^{228}Th via two short lived intermediate daughter decay products, ^{228}Ra ($t_{1/2} = 5.75$ yr.) and ^{228}Ac ($t_{1/2} = 6.13$ yr.).

Equilibrium in the soil between ^{232}Th and ^{228}Th is assumed in the calculations for ^{230}Th activity and thorium recovery from the samples. Although small deviations from equilibrium may occur due to alpha-recoil damage of mineral structures, making ^{228}Th more susceptible to leaching by ground and soil waters, the mobility of thorium is quite low so that appreciable disequilibrium from this cause is unlikely. Also, the greater mobility of the intermediate ^{228}Ra may allow for some migration before the production of ^{228}Th ; however the short half-life of ^{228}Ra relative to time scales for weathering and nuclide transport indicates that little disequilibrium is produced by this process. This approximation is made more reliable by adding the ^{228}Th tracer in activities five or more times greater than the natural ^{232}Th activity in the sample

Background counts are generated by low energy scatter from the higher energy peaks. The background counts are estimated from the number of counts detected in blocks of channels on either side of the peak. Peak interferences from ^{235}U may result from the branching ratios of some radionuclides (Vargas et al., 1994). However, in these natural samples, ^{235}U contributes less than 1% of the total activity of these peaks.

Another potentially important interference is from ^{224}Ra , the immediate daughter of ^{228}Th . Of its total emitted energy, only 4.9% is present under the ^{228}Th peak. However, because this Ra is generated from decay of ^{228}Th , the error associated with ignoring this contribution of ^{224}Ra is minimized by counting the sample a short time after plating. Even for samples counted more than one month after plating, the counts from the ^{224}Ra peak contribute less than 5% of the ^{228}Th peak.

Incomplete chemical separation of U and Th prior to electrodeposition and spectral analysis can result in several peak interferences between the U and Th nuclides. Mathematical methods employed for these alpha energy peak corrections, as well as computer programs generated to carry them out, are described by Tomé and Sánchez (1988) and Vargas et al. (1994). Such corrections were not necessary for the samples analyzed in this report, as chemical separation of uranium and thorium was nearly complete, and the detectors yielded high resolution alpha peaks (24 keV Full Width at Half Maximum).

Selective Extractions

Progressive selective leaches (e.g. exchangeable cations, organic matter, free oxides, sand, silt and clay) were performed on the soil samples in order to determine the chemical form of U and Th throughout the soil profiles studied. These leaches were performed on the A-, B- and C-

horizons of both profiles, as each horizon is typically dominated by different pedogenic controls: biological activity and eluviation in the A-horizon, illuviation in the B-horizon, and parent material in the C-horizon. The methodology used to progressively leach the exchangeable cations, organic matter and free oxides is an amended version of that described by Greeman (1992). The mineral fractions were separated using a procedure adapted from Jackson (1956). All leaches were performed at pH 5, as soils have a natural pH range between 4 and 6, and radionuclides in the liquid phase are more stable under acidic conditions (Greeman, 1992). These extraction procedures, described in detail below, are outlined in Figure 5.

Soil samples weighing 5 g are placed in centrifuge tubes. Exchangeable cations are extracted by adding 25 ml of 1M MgCl_2 (pH = 5 with HNO_3), swirling for 15 min., centrifuging for 5 min. at 2000 rpm and decanting the supernatant into a clean centrifuge tube. 10 pCi of ^{232}U - ^{228}Th tracer and 10 ml of 1000 mg/L Fe solution are added to this decantate which is then analyzed for the U and Th nuclides using the total soil procedure beginning with the Fe-oxide precipitation step. Tessier et al. (1979) document the ability of Mg^{2+} to exchange for specifically adsorbed cations at similar concentrations.

Organic matter is then leached from the solids settled during the above centrifuge step by adding 25 ml of 5% NaOCl, heating the sample to 90°C for 30 min., centrifuging, and decanting the supernatant into a clean centrifuge tube. Again 10 pCi of ^{232}U - ^{228}Th tracer and 10 ml of 1000 mg/L Fe solution are added to the decantate which is then analyzed for the U and Th nuclides using the total soil procedure beginning with the Fe-oxide precipitation step. According to Kogel-Knaber and Hatcher (1989), sodium hypochlorite (NaOCl) is a simple oxidizing agent which primarily attacks the electron-rich aromatic and carboxylic functional groups of large organic molecules in geologic samples. Hence, any trace element liberated during this leaching step is probably associated with those functional groups and groups adjacent to them (Greeman, 1992).

The "free" oxides of Fe, Mn, and Al are extracted using dithionite, a reducing agent, in conjunction with the complexing agents citrate and bicarbonate. To the solids settled during the above centrifuge step, 24 ml of 0.3M Na-citrate and 3 ml of 1M Na-bicarbonate are added. Subsequently, the tube is heated to 80 °C, and 1 g of Na-dithionite is stirred in, the sample is centrifuged and the supernatant is decanted into a beaker. Similarly to the previous leaches, 10 pCi of ^{232}U - ^{228}Th tracer is added to the solution.

Early attempts to directly precipitate the Fe-oxides and continue with the total soil procedure from this point yielded very poor recovery of both U and Th. Calculations indicate strong complexation of Fe, U and Th by the citrate in solution. Therefore, the solution is evaporated in the presence of concentrated HNO_3 to destroy the citrate. The sample is then re-dissolved with 2M HCl, and analyzed for the U and Th nuclides using the total soil procedure, beginning with the Fe-oxide precipitation step.

Separation of the mineral fractions according to particle size is accomplished using a modified version of the procedure described by Jackson (1956). The sand-sized particles (43-2000 μm) are separated by wet sieving the remaining sample through a 325 mesh (43 μm) sieve with H_2O . The sands which do not pass through the sieve are then analyzed for the U and Th nuclides using the total soil procedure (from the beginning).

The silt-sized particles (2-43 μm) are separated from the clay fraction by centrifuging, as discussed by Jackson (1956). Samples are initially centrifuged for 13.5 min. at 700 rpm (a 50% time excess) in a 10 cm water column, and the supernatant containing clay particles is decanted into a clean beaker. The sediment remaining in the tubes is then transferred into a single tube, filled to a depth of 10 cm, stirred and centrifuged for 9 min. at 700 rpm, decanting the supernatant into the beaker noted above. Repeating this 9 min. centrifuge step four more times

allows for >97% clay separation (Jackson, 1956, p.139). The silt-sized sediment left in the bottom of the tube after these centrifugings is then oven dried and weighed. This silt-sized material is then analyzed for the U and Th nuclides using the total soil procedure.

The clay suspension from the aforementioned beaker is transferred to several centrifuge tubes filled to a depth of 10 cm and centrifuged for 55 min. at 2400 rpm, settling particles with diameters $>0.06\mu\text{m}$, and the supernatant is decanted into another beaker. The sediment at the bottom of the tubes is transferred to a single tube, the supernatant decanted, and the tube placed in the oven to dry the clays. The fine clays suspended in the remaining solution are flocculated by adding saturated NaCl solution (6N), making the solution $\approx 1\text{N}$ NaCl. These clays are left to gravity settle for at least 24 hrs., then the solution is siphoned off to approximately 1 cm above the settled clays. The remaining solution and clays are transferred to centrifuge tubes and centrifuged for 1 hr. at 2000 rpm. The supernatant should be clear, indicating little suspended clay, hence it may be decanted and discarded. The settled clays are transferred to a single tube, rinsing with 1N NaCl. It is centrifuged for 1 hr. at 2000 rpm and the supernatant decanted and discarded. The settled clays are dried in an oven, then combined with the clays dried in the earlier step. They are collectively weighed and analyzed for the U and Th nuclides using the total soil procedure.

Calibration and Error Determination

Calibration

The activity values obtained for ^{238}U , ^{234}U , and ^{230}Th were calculated from the concentration of the ^{232}U - ^{228}Th tracer, which was diluted from a solution containing $0.1 \mu\text{Ci} \pm 1\%$ supplied by Isotope Products Labs (Burbank, CA) in Oct. 1992. The tracer was diluted in several steps to 5 pCi/ml (Hogue, 1996). Most samples were spiked with 2 ml of this tracer solution, and the associated error for the 10 pCi tracer was calculated to be 2.12%.

Accuracy of the above calibration was tested by analysis of a fresh water lake sediment (SRM 4354) supplied by NIST (Gaithersburg, MD), yielding activity values for ^{238}U , ^{234}U , and ^{230}Th in reasonable agreement with those determined by NIST (Table 2). The activity determined for ^{238}U by the aforementioned alpha-spectrometric method falls within the 95% tolerance limits given by NIST for a 95% confidence interval, based on a standard deviation of 2.51% for 15 measurements. The uncertified ^{230}Th activity value given by NIST falls within 1σ (based on counting error) of that determined by this method. The difference in ^{234}U activity values is noticeably higher, however no error limits were given for the uncertified NIST values, and this deviation may reflect a larger error associated with the ^{234}U activity determination given by NIST.

Error Analysis

Errors in determinations of nuclide activities and recovery are primarily the result of radiation counting errors. These errors are combined according to the error propagation procedures given by Jester (1993, p. 34-37), with the following assumptions:

1. Good chemical separation of uranium and thorium prior to electrodeposition.
2. Background counts in the ^{228}Th and ^{232}U peaks are negligible relative to the total integral counts of these peaks.
3. The lower energy alpha peak of ^{224}Ra does not contribute significant counts to the ^{228}Th peak (More valid when the elapsed time between sample preparation and alpha-spectrometric counting is short, as approximate equilibrium is attained between ^{224}Ra and ^{228}Th in ≈ 1 month).
4. Natural ^{228}Th is in secular equilibrium with ^{232}Th in all samples.
5. Analytical error is small relative to counting error.

Reproducibility

Several replications of total soil analyses were made for all 7 measured horizons of the NY-1 profile. Most of these were made during procedural development, and some had very low tracer yields. Due to the high error associated with low yield samples, data from samples whose uranium or thorium recovery was less than 15% were discarded. From the remaining data set, 17 replications of uranium measurements, and 19 for thorium were available. For ^{238}U , 14 (82%) were within 1 σ counting error, and the remaining 3 (18%) were within 2 σ . For ^{234}U , 12 (71%) were

within 1σ , and the remaining 5 (29%) were within 2σ . ^{230}Th replications were somewhat more deviant, with 13 (68%) within 1σ , 4 (21%) within 2σ , 1 (5%) within 3σ , and 1 extremely anomolous value from a NY-1-2 sample outside 3σ . These results verify that analytical error is dominated by counting error.

RESULTS

Site Characterizations

The preceding methods were applied to samples from two profiles, NY-1 and IL-1, both having Pleistocene till as parent material. These soil profiles, as well as the vegetation growing on them, were sampled in 1989 by E.J. Ciolkosz, R.R. Dobos, A.W. Rose, J.W. Washington and D.J. Greeman. A synopsis of the on-site profile descriptions is given by Hogue (1996). Additional characterization of these sites, and their associated soil properties, was presented in Greeman (1992), and is summarized below and in Table 3.

Site NY-1 is located near Ausable Forks, New York ($44^{\circ}28'21''$ N, $73^{\circ}43'31''$ W). This soil has a frigid Mean Annual Soil Temperature (MAST), between 0 and 8 °C, and an udic moisture regime. The parent materials for this soil profile include Wisconsinan-aged deposits of unstratified till near the surface and ice-contact till at depth. These deposits are derived mainly from granitic gneiss of the Adirondak highlands. In the lowermost C-horizon, a large proportion of highly weathered sedimentary rocks indicates

a different source area. The soil profile is deep (>3m) and well drained. Mature deciduous tree stands indicate that the soil has remained relatively undisturbed for at least 100 years. It is classified as a Haplorthod (USDA Nomenclature), and is an Adams Sandy loam soil type.

Site IL-1 is located in Champagne-Urbana, Illinois (40°05'11" N, 88°13'52" W). The associated MAST is mesic, between 8 and 15 °C, and the moisture regime is udic. The soil is developed on loess which grades into basal till at depth. These deposits are of Wisconsinan Age, and appear to have at least two different source areas. Significant input of organic matter derived from prairie grass has led to the development of a thick A-horizon in this mollisol. The presence of an Ap horizon indicates that it has been farmed in the past. This somewhat poorly drained soil is classified as an Argiudoll, and is a Flanigans Silt loam soil type.

3.2 Reproducibility

An additional check on reproducibility was accomplished by comparing the ^{238}U values of this study to those obtained by neutron activation for the same samples, as presented in Greeman (1992). Figure 6, a plot of ^{238}U activities from total samples obtained from the alpha-spectrometry determinations of this study vs. the neutron activation analyses of Greeman (1992), indicates a good correlation between the total sample values from both profiles, with a correlation coefficient (R) of 0.95.

However, the activity values obtained from alpha-spectrometry are consistently less than those from neutron activation analyses, as evidenced by the slope of 0.76 for the regression line. A similar relationship, with r^2 of 0.98 and slope 0.61, is found between the normalized ^{238}U selective extraction data of Greeman (1992) by neutron activation and determinations by the present method (Hogue, 1996). The two methods therefore appear to have a bias.

Given that the aim of this study is to investigate the chemical form and relative mobility of the U-series nuclides, observable patterns in the nuclide distributions with depth and in select phases are more important than absolute activity values. The strong correlation between the measured ^{238}U activities supports these types of observations, and conclusions drawn from them. The ^{226}Ra values of Greeman (1992) were calibrated independently of the U values, using NIST Ra solution, and are considered correct. Correction of his U values to the present method would further accentuate the disequilibrium he observed.

NY-1 Profile

State of Secular Equilibrium

Disequilibrium between ^{238}U , ^{234}U and ^{230}Th is present throughout the soil profile at site NY-1 (Table 4, Figure 7). ^{234}U consistently exceeds ^{238}U by an average of 8.2% for horizons 2,3,4,5,7, and 10 (A, B, and uppermost C horizons). Likewise, ^{230}Th exceeds ^{238}U by an average of 25.5% for horizons 3,4,5,7, and 10. The extent of

disequilibrium in the order $^{230}\text{Th} > ^{234}\text{U} > ^{238}\text{U}$, along with the activity of these nuclides, increases with depth between horizons 3 through 10. The lowermost C-horizon, NY-1-12, is essentially in equilibrium with respect to uranium and thorium. The ^{226}Ra activities are more variable with depth. However an overall pattern of enrichment relative to its parent and grandparents is observable, with the greatest enrichment in the A- and upper B-horizons (0-30 cm).

Equilibrium State in Select Soil Phases

The pedogenic causes for these disequilibrium conditions were evaluated by performing selective leaches (i.e., exchangeable ion, organic, Fe-oxide, sand, silt and clay fractions) on samples of the A, Bs2, and deepest C horizons (NY-1-2, NY-1-5 and NY-1-12). Activities of ^{238}U , ^{234}U , ^{230}Th and ^{232}Th in the various soil phases of each horizon, are presented in Tables 5, 6 and 7. These activity values were normalized to 100% of those obtained for the total sample (Table 8), and daughter/parent ratios were calculated. A few measured activities which fell below the lower detectable limits of the analyzer ($4.66\sigma_{\text{bkg}}$), were included in these calculations, in the absence of a more acceptable value from a replicated sample. The latter measurements may not be statistically significant, but they are the best approximation available. Because two extraction analyses were performed on NY-1-2

horizon, some phases had more than one representative activity value for a given nuclide. These values were averaged prior to normalization.

Figure 8 and Table 8 display the contribution of each phase to the total activities of ^{238}U , ^{234}U , ^{230}Th , and ^{226}Ra (the latter from Greeman, 1992), as a function of depth in the soil profile. The complete loss of the NY-1-12 U iron-oxide fraction was a result of complexation by the citrate solution, as discussed earlier. Because data from this fraction was lacking, the activity values for this horizon could not be normalized, and are therefore omitted from Figure 8.

In all of the pedogenic phases (i.e. exchangeable ions, organics, and iron-oxides), ^{238}U was consistently depleted with respect to ^{234}U . Within the sand fraction, ^{238}U and ^{234}U are in equilibrium ($\pm 1-6\%$) for all three horizons. In the finer grained mineral fractions (silt and clay), ^{234}U is markedly depleted with respect to ^{238}U in all samples except for the NY-1-5 silt fraction where there is approximate equilibrium between the parent and daughter uranium nuclides.

^{230}Th , on average, is enriched with respect to its uranium ancestors in all phases except in the organics and Fe-oxides. The deficiency of ^{230}Th in the organic fraction of the soil is most notable in the A horizon, where the largest proportion of organic C, 6.26 weight % is present (Table 3). Here, the ^{234}U activity is about 11 times that

for ^{230}Th . The low levels of ^{230}Th in this soil fraction appear dominantly responsible for the deficiency of ^{230}Th with respect to ^{234}U and ^{238}U in this horizon. In the deeper horizons, the depletion is less significant as the weight % organic carbon is below 1% and the $^{234}\text{U}/^{230}\text{Th}$ and $^{238}\text{U}/^{230}\text{Th}$ activity ratios are much lower (between 1.1 and 2).

The occurrence of these three nuclides in the iron oxide phase is quite variable with depth, although the abundance of iron oxides remains relatively constant, between 0.4 and 1.0% (Table 3), throughout the profile. In the A-horizon, the activity distribution in the Fe-oxides is $^{234}\text{U} > ^{230}\text{Th} > ^{238}\text{U}$, with occurrences of 21.7, 17.9, and 16.1% of their total activity, respectively. In the B-horizon, this distribution is $^{234}\text{U} > ^{238}\text{U} > ^{230}\text{Th}$, with occurrences of 21.0, 15.4, and 7.8% of their total activity. The proportion of ^{230}Th in the iron oxides is increased to 25.8% of its total activity in the C-horizon. The un-normalized ^{238}U and ^{234}U values strongly suggest that these nuclides are enriched in the iron-oxides of the C-horizon with respect to the B-horizon as well. However, the equilibrium condition between these nuclides could not be ascertained.

The greatest enrichment of ^{230}Th over uranium occurs in the exchangeable ion fraction, particularly in the A-horizon where 10.2% of the total thorium is in this chemical form. The uranium nuclides have a minimal

occurrence in this phase, below 2.5% of their total activity in all measured horizons. This difference is subdued with depth, as the percentage of ^{230}Th in this chemical form gradually drops to 2.3% in the C-horizon.

The sand, silt and clay fractions all, on average, have excess ^{230}Th , but in the more weathered silt and clay fractions this disequilibrium is more pronounced (Table 8). The average $^{230}\text{Th}/^{234}\text{U}$ ratio in the silt fraction of the A- and B-horizons is 1.50. The $^{230}\text{Th}/^{234}\text{U}$ ratio in the clays of the A-horizon is 1.08, but in the B-horizon where clay content is at a maximum, 4.7 weight %, the ratio is 3.13 (Table 9). This enrichment of clay content and the ^{230}Th concentrations associated with them is reflected by the change from $^{234}\text{U} > ^{238}\text{U} > ^{230}\text{Th}$ in the A-horizon to $^{230}\text{Th} > ^{234}\text{U} > ^{238}\text{U}$ in the B-horizon.

The ^{226}Ra activities tend to mirror those for ^{230}Th in the mineral phases of the A- and B- horizons, except for the A-horizon sands which appear enriched in ^{226}Ra . The ^{226}Ra in the exchangeable ion fraction is enriched relative to ^{230}Th in the A-horizon, but approximate equilibrium exists between exchangeable ^{226}Ra and ^{230}Th in the measured B- and C-horizons. In the Fe-oxide phase, there is an overall depletion of ^{226}Ra with respect to ^{230}Th , ^{234}U and ^{238}U throughout the profile. However, in the organic fraction, ^{226}Ra is markedly enriched with respect to these other nuclides, and maintains relatively high activity

values into the C-horizon, where the weight % organic carbon is quite low (0.02%).

The overall activity of ^{226}Ra is greater than that of ^{238}U , ^{234}U , and ^{230}Th in the pedogenic phases in the A- and B-horizons, with an average pedogenic occurrence of 31%. However, in the C-horizon, the pedogenic occurrence of ^{226}Ra is only 11% (almost entirely organic), and its total pedogenic phase activity is much less than for ^{230}Th . Conversely, the silt and sand fractions of the C-horizon are significantly enriched in ^{226}Ra with respect to ^{230}Th .

IL-1 Profile

State of Secular Equilibrium

Results from analyses for ^{238}U , ^{234}U and ^{230}Th in samples of various horizons of the IL-1 profile are given in Table 10, and are illustrated, along with ^{226}Ra data acquired from Greeman (1992), in Figure 9. A pattern of disequilibrium throughout the profile is revealed, with a general activity trend of $^{226}\text{Ra} > ^{230}\text{Th} > ^{238}\text{U} > ^{234}\text{U}$, and a decrease in the activities of these nuclides with depth. ^{238}U exceeds ^{234}U in all measured horizons by an average of 7.4%. Similarly, ^{230}Th exceeds ^{238}U in all determined horizons by an average of 27.2%, with maximum disequilibrium in the B-horizons. ^{226}Ra exceeds ^{230}Th in the measured A- and B-horizons by an average of 10.0%, but is depleted relative to ^{230}Th in the C-horizon by 14.2%. ^{226}Ra is in excess of both uranium nuclides in all horizons with known activity values.

Equilibrium State in Select Soil Phases

The normalized data from the selective extractions from samples of the A, Bt2, and 2C horizons (IL-1-2, IL-1-7, and IL-1-9) are displayed in Tables 11, 12, and 13 and Figure 10. The normalized values were also incorporated into Table 14. Table 15 shows the normalized activity values within each fraction of IL-1.

The pedogenic phases (i.e. exchangeable ions, organics, and Fe-oxides) are all, on average, enriched in ^{234}U with respect to ^{238}U (Table 14). Conversely, the mineral phases (sand, silt and clay) are all depleted in ^{234}U .

On average for IL-1-2 and IL-1-7, ^{230}Th is enriched relative to the uranium nuclides in all soil fractions except for the organic and silt. In the silt fraction, ^{230}Th activities are intermediate between ^{234}U and ^{238}U , with an average $^{230}\text{Th}/^{238}\text{U}$ ratio of 0.94. The average $^{230}\text{Th}/^{234}\text{U}$ ratio in the organic fraction is 0.43. Maximum thorium enrichment occurs in the iron-oxide fraction, where the average $^{230}\text{Th}/^{234}\text{U}$ ratio is 3.87, and the average ^{230}Th occurrence in this phase is 24% of its total activity. A similar $^{230}\text{Th}/^{234}\text{U}$ activity ratio is observed in the exchangeable ion fraction, but the maximum thorium occurrence in this phase is only 3.2% of the total thorium activity. The average $^{230}\text{Th}/^{234}\text{U}$ activity ratio for clays is 2.093, and for sand this ratio is 1.44.

Summary

The alpha-spectrometric analyses reveal marked disequilibrium conditions between ^{238}U , ^{234}U , ^{230}Th and ^{226}Ra in both soil profiles, which vary with depth and within soil phases. Both profiles are enriched in ^{230}Th relative to ^{234}U , with increased $^{230}\text{Th}/^{234}\text{U}$ activity ratios from the A- to lower B-horizons. ^{234}U is enriched in the soils of NY-1 but depleted in IL-1 soils relative to ^{238}U , with profile averaged $^{234}\text{U}/^{238}\text{U}$ activity ratios of 1.055 and 0.918 respectively. The activity of ^{226}Ra is within 14% of equilibrium with ^{230}Th for all measured horizons of IL-1. However, in NY-1, $^{226}\text{Ra}/^{230}\text{Th}$ ratios range from 0.817 (NY-1-10) to 1.979 (NY-1-3) and vary widely with depth.

Disequilibrium between these U-series nuclides exists in all measured soil phases. Overall, their relative activities in the mineral fractions (sand, silt, clay) are $^{230}\text{Th} \approx ^{226}\text{Ra} > ^{238}\text{U} > ^{234}\text{U}$. However, in the A-horizons, ^{230}Th is depleted with respect to the other nuclides in the sands of NY-1 and the silts of IL-1. Also, in the silts and sands of horizon NY-1-12, the activity of ^{230}Th is about half that of ^{226}Ra . Relative activities in the exchangeable ion fractions of all samples are $^{226}\text{Ra} \geq ^{230}\text{Th} > ^{234}\text{U} > ^{238}\text{U}$. The nuclide distribution in the organic phase is $^{226}\text{Ra} > ^{234}\text{U} > ^{238}\text{U} > ^{230}\text{Th}$. In the iron-oxide fraction, the activity distribution of $^{234}\text{U} \geq ^{238}\text{U} > ^{226}\text{Ra}$ exists. $^{230}\text{Th}/^{234}\text{U}$ iron-oxide activity ratios exceed unity in the IL-1 profile, but are less than one in NY-1.

DISCUSSION

Geochemical Controls on Observed Disequilibrium

Preferential Leaching of ^{234}U Relative to ^{238}U

The results from both profiles support the hypothesis that ^{234}U is preferentially leached from minerals during weathering because of lattice damage from alpha-recoil. The $^{234}\text{U}/^{238}\text{U}$ ratios in the NY-1 samples are generally lower in silt and clay than in sands, as expected for this process. In the silt and clay fractions of the NY-1 profile, the average $^{234}\text{U}/^{238}\text{U}$ activity ratios of the A- and B-horizons are 0.92 and 0.83 respectively. The less easily leached sand-size particles in this profile are within the 1σ counting error limits of equilibrium, with normalized $^{234}\text{U}/^{238}\text{U}$ activity ratios of 0.99 in the A-horizon and 0.94 in the B-horizon. The activity ratios from the IL-1 profile show similar average depletions of ^{234}U in the mineral fractions of the A- and B-horizons; 0.91 for sand, 0.91 for silt, and 0.94 for clay.

After uranium is leached from the mineral fractions into the pore water, a portion of it is incorporated into the pedogenic phases (Fe-oxides, organic matter, exchangeable ions and some clays). Given that ^{234}U is leached into the pore fluid at a higher rate, it is not surprising that all the pedogenic fractions, on average, show enrichment of this nuclide with respect to ^{238}U in the normalized activity values of both profiles.

Although having excess ^{234}U in the pedogenic phases, the thickness-weighted, profile-averaged $^{234}\text{U}/^{238}\text{U}$ activity ratio of the measured horizons at site NY-1 is 1.06. This ratio seemingly contradicts the theory of preferential leaching of ^{234}U , which should lead to $^{234}\text{U}/^{238}\text{U}$ less than unity if any uranium is leached from the profile. However, only the soil fraction finer than 2 mm was analyzed in this study. The determinations thus exclude approximately 32% of coarser material that was sieved out of the NY-1 samples during sample preparation. A likely interpretation is that the coarser fraction released excess ^{234}U into the soil matrix, where it has been fixed into the pedogenic phases to give an overall excess of ^{234}U for the < 2 mm fraction. In contrast, the IL-1 profile, which lacks a coarse fraction, has a thickness weighted average $^{234}\text{U}/^{238}\text{U}$ ratio slightly less than unity (0.92).

Preferential Enrichment of ^{230}Th in Primary Minerals and Deficiency in Pedogenic Phases

In the B horizon of NY-1 and in all horizons of IL-1, ^{230}Th exceeds its parent ^{234}U . The average $^{230}\text{Th}/^{234}\text{U}$ is 1.13 for NY-1 and 1.39 for IL-1, with a maximum of 1.68 in the mid-B-horizon of IL-1. This pattern is attributed to the relative immobility of Th compared to U, as a result of stronger adsorption and lower solubility. The pattern extends to the sand, silt and clay fraction of nearly all samples in both profiles, the only exception being the sand

of NY-1-2. This pattern is interpreted to result mainly from preferential loss of U from the primary soil phases.

The $^{230}\text{Th}/^{234}\text{U}$ ratio exceeds 3 in clays of the B horizon of both profiles, and the ^{230}Th concentration within the clay fraction of the B-horizon is higher than in the C-horizon clay (Table 9 and 15). A part of this enrichment is interpreted to be caused by greater leaching from the fine clay particles than from the sands and silts, and a part from preferential incorporation of ^{230}Th in clay formed in the B-horizon soil (pedogenic clay).

In contrast, in the organic phase at both sites, and in the Fe-oxides at NY-1, ^{234}U is enriched relative to ^{230}Th . In Fe-oxides of the A- and B-horizons at NY-1, the $^{230}\text{Th}/^{234}\text{U}$ ratio is 0.44 and 0.78, respectively; in the organic fraction it is 0.09 and 0.52, and at IL-1 the ratio for the organic fraction is 0.46 and 0.40. These major departures from equilibrium are interpreted to result from enrichment of ^{234}U relative to ^{230}Th in soil solutions, and incorporation of this enrichment to the newly formed soil phases.

The Fe-oxides of IL-1 differ from those of NY-1 in having high $^{230}\text{Th}/^{234}\text{U}$ ratios (3.4 and 4.2 for the A- and B-horizons, respectively). A possible cause for this difference from NY-1 is the higher amount of organic matter in the IL-1 soil compared to NY-1, and the stronger affinity of U for organic matter, especially at the higher soil pH of IL-1. A contributing reason may be the

increasingly strong carbonate complexing of U but not Th at pH values above about 6 (Quigley et al., 1996).

A surprising feature is the high $^{230}\text{Th}/^{234}\text{U}$ of the exchangeable fraction in the A- and B-horizons of both profiles. The $^{230}\text{Th}/^{234}\text{U}$ ratio is 4.3, 2.1, 4.7 and 2.8 in these exchangeable fractions, with values exceeding 4 in the A-horizons of both profiles. This anomaly clearly indicates that some ^{230}Th is mobile, probably complexed with an organic ligand, and is perhaps more securely held in exchange sites, especially on organic matter, than is ^{234}U .

Partition of U and Th between Pedogenic Phases

As organic matter, Fe-oxides and clay are formed in the soil by pedogenic processes, and as exchange sites interact with the soil solution, they would be expected to incorporate the components ^{238}U , ^{234}U , ^{230}Th and ^{226}Ra according to an equilibrium constant K_i^j :

$$K_i^j = a_i^j / a_i^{\text{soln}}$$

where a is the chemical activity of the component i in the phase j . The partition of a nuclide between two phases, such as Fe-oxide and exchangeable ions, would then be given by

$$K_i^{\text{exc-Fe}} = K^{\text{exc}} / K^{\text{Fe}} = a_i^{\text{exc}} / a_i^{\text{Fe}} = \lambda_i^{\text{exc}} C_i^{\text{exc}} / \lambda_i^{\text{Fe}} C_i^{\text{Fe}} \quad (\text{x})$$

where λ is an activity coefficient and C the concentration of component i in the indicated phase.

Although this basic process may operate, it is complicated by a number of factors. The value of K may vary appreciably with pH, temperature and competing ions.

In addition, the observed ratio of concentrations may be strongly affected by complexing in solution and interactions in the solid, as well as the degree to which equilibrium is approached. Also, the organic matter may form in the root zone or in the above-ground plant, and be modified from its initial composition during decay and migration. Nevertheless, the ratios of nuclides between pedogenic phases probably reflect approximate affinities of the phases for the nuclides.

The concentrations of the nuclides in the three are summarized in Tables 9 and 15. The concentration in pCi/g within organic matter and Fe-oxide are calculated from the concentration of these two phases measured on the samples (Table 3). The concentration in the exchange sites was estimated in pCi/g of exchangeable cations using the cation exchange capacity and the assumption that the dominant exchangeable ion was Ca for IL-1 and Al for NY-1, as indicated by analyses of exchangeable cations in samples from the profiles.

The partition among phases varies significantly between horizons and between sites. For U in the A-horizon of NY-1, the Fe-oxides are the preferred host with exchange sites a close second and organic matter markedly weaker. For U in the B-horizon, exchange sites have the highest affinity, followed by Fe-oxides, and again with organic matter the weakest. For U in the A-horizon of IL-1, organic matter is slightly preferred to Fe-oxides, and

exchange sites are a markedly weaker host. In the B horizon, organic matter is again the preferred host, and exchange sites are slightly preferred over Fe-oxides. However, all three types of sites have concentrations within a factor of about 4 at IL-1.

For Th at NY-1, the exchange sites have the highest affinity, Fe-oxides are second, and organic matter is clearly the weakest. In IL-1, Fe-oxides have the strongest affinity in the A-horizon, but organic matter increases its affinity downward to become the preferred host in the C-horizon.

For Ra at NY-1, exchange sites are the preferred host. Organic matter also has a strong affinity in the B-horizon, but is similar to Fe-oxides in the A-horizon. Clay generally has the lowest affinity.

Undoubtedly the numerous changes in affinity, as indicated by Tables 9 and 15, result from complex interactions of the factors mentioned above. Important factors probably include differences between field sites and with depth in pH, solution concentration, host-phase properties (i.e., degradation of organic matter) and pathways of incorporation (plant uptake, illuviation of clay, Fe-oxides and organic matter, etc.). The lack of consistent patterns indicates the complexity of these interactions, and suggests caution in interpreting or predicting behavior in soil.

Radium Enrichment by Plant Cycling

The ^{226}Ra activity in the A- and B-horizons of the IL-1 profile exceeds that of the other nuclides by an average of 47.5% for ^{238}U , 58.1% for ^{234}U and 10.0% for ^{230}Th ; however, in the C-horizon, the ^{226}Ra enrichment over its uranium precursors is only 15.5% for ^{238}U and 31.1% for ^{234}U , and ^{226}Ra is depleted relative to ^{230}Th by 12.4%.

These differences in the relative nuclide distributions with depth are inferred to result mostly from a combination of Ra uptake by vegetation, strong affinity of Ra for organic matter and insolubility of U under reduced conditions. Greeman (1992, ms) showed that relative to U and Th, Ra was preferentially mobilized from the B- and C-horizons into the A horizon by vegetation. Sixteen samples of vegetation from the studied sites showed Ra/U ratios averaging 2.8. The A- and B-horizons at IL-1 are above the water table but the C-horizon is often below the water table and contains many, suggesting locally reducing conditions. The U may have been leached from the A- and B-horizons and partly precipitated or adsorbed in the reducing C-horizon.

CONCLUSIONS

1. A method for analysis of ^{238}U , ^{234}U , ^{230}Th , and ^{232}Th in soils has been developed and used to determine the distribution of these nuclides through the soil horizons of a typical mollisol (IL-1) and a typical alfisol (NY-1), previously analyzed for ^{226}Ra by Greeman (1992).

2. Methods for determining these nuclides in selective fractions of these soils (exchangeable ions, organic matter, Fe-oxides, sand, silt and clay) have been developed and applied to representative horizons of these profiles to provide information on the speciation of ^{238}U , ^{234}U and ^{230}Th and to help predict their behavior in soils.

3. Disequilibrium is found to be widespread along the decay series ^{238}U - ^{234}U - ^{230}Th - ^{226}Ra . For 9 total samples the $^{230}\text{Th}/^{234}\text{U}$ ratio ranges from 0.93 to 1.68, and the $^{226}\text{Ra}/^{230}\text{Th}$ ratio from 0.83 to 1.98. Within pedogenic phases, the $^{230}\text{Th}/^{234}\text{U}$ ratio ranges from 0.09 to 0.52 in organic matter, and 0.45 to 4.2 in Fe-oxides. The $^{226}\text{Ra}/^{230}\text{Th}$ ratio is as high as 23 in organic matter.

4. In both profiles ^{234}U is preferentially leached relative to ^{238}U from the residual clay, silt and sand phases (median of 12 fractions 0.89, range 0.74 to 1.03). This disequilibrium is presumably due to α -recoil induced damage of the uranium-bearing mineral grains during the decay process. In contrast, pedogenic phases are enriched in ^{234}U relative to ^{238}U (median of 12 fractions, 1.38, range 0.83 to 2.3). This enrichment occurs by incorporation of the nuclides from the enriched pore solution. In IL-1, the $^{234}\text{U}/^{238}\text{U}$ ratio for the total profile is below unity because of preferential leaching of ^{234}U , but in NY-1 the ratio exceeds one, probably because uranium isotopes were leached from the > 2 mm fraction, which was not analyzed, and fixed in pedogenic phases.

5. In both profiles, ^{230}Th exceeds ^{234}U activity, but this disequilibrium is much more pronounced in IL-1 (ratio of 1.39) than in NY-1 (ratio of 1.13) because of its higher proportion of Fe-oxide and clay, combined with the higher affinity of these phases for ^{230}Th .

6. Preferential uptake and cycling by plants at NY-1 has led to a distribution of ^{226}Ra which appears independent of ^{230}Th throughout the profile, with ^{226}Ra enriched at the surface because plants at this site are Ra-rich, and depleted at depth because of uptake by roots, as well as downward leaching. In contrast, the grasses at IL-1 did not significantly fractionate ^{226}Ra and ^{230}Th with depth, and the zone of nuclide enrichment from plants (0-30 cm) coincides with that for nuclide uptake by plant roots.

7. Clays are a significant pedogenic host for ^{226}Ra and ^{230}Th , and activities of these nuclides are in approximate equilibrium in all measured clay fractions. Both ^{230}Th and ^{226}Ra have a greater affinity for illuvial clays in the B-horizon. Leaching from clays dominates over adsorption onto clays for uranium. A surprising amount of ^{230}Th is exchangeable, perhaps as an organic complex.

8. Iron-oxides are major pedogenic hosts for uranium and thorium but not radium, although some ^{226}Ra adsorption onto iron-oxides is observed in an illuvial horizon (NY-1-5).

9. ^{226}Ra has the greatest association with organic matter. ^{234}U and ^{238}U are preferentially incorporated into

organic matter relative to ^{230}Th at IL-1. At near-neutral pH, $^{234}\text{U}/^{230}\text{Th}$ activity ratios in soil organic matter are similar to their values in the parent vegetation, implying similar affinities between uranium and thorium in organic matter.

9. The results from the NY-1 profile clearly indicate that where vegetation action is significant, and pedogenic hosts limited, ^{226}Ra can be enriched independently of ^{230}Th . However, where vegetation does not selectively fractionate ^{226}Ra between horizons, and pedogenic hosts (i.e., organic material, iron-oxides and clays) are relatively abundant throughout the profile (e.g. IL-1), ^{226}Ra and ^{230}Th activities for a given horizon are near-equilibrium, but the nuclides are likely associated with different soil fractions.

10. Because of extensive and varied disequilibrium in the U decay-series in soils, the abundance of Ra is a much better guide to the radon potential than is abundance of U.

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Table xx. Concentration of normalized ^{238}U , ^{234}U , ^{230}Th and ^{226}Ra within phases (in pCi/g of the phase¹)

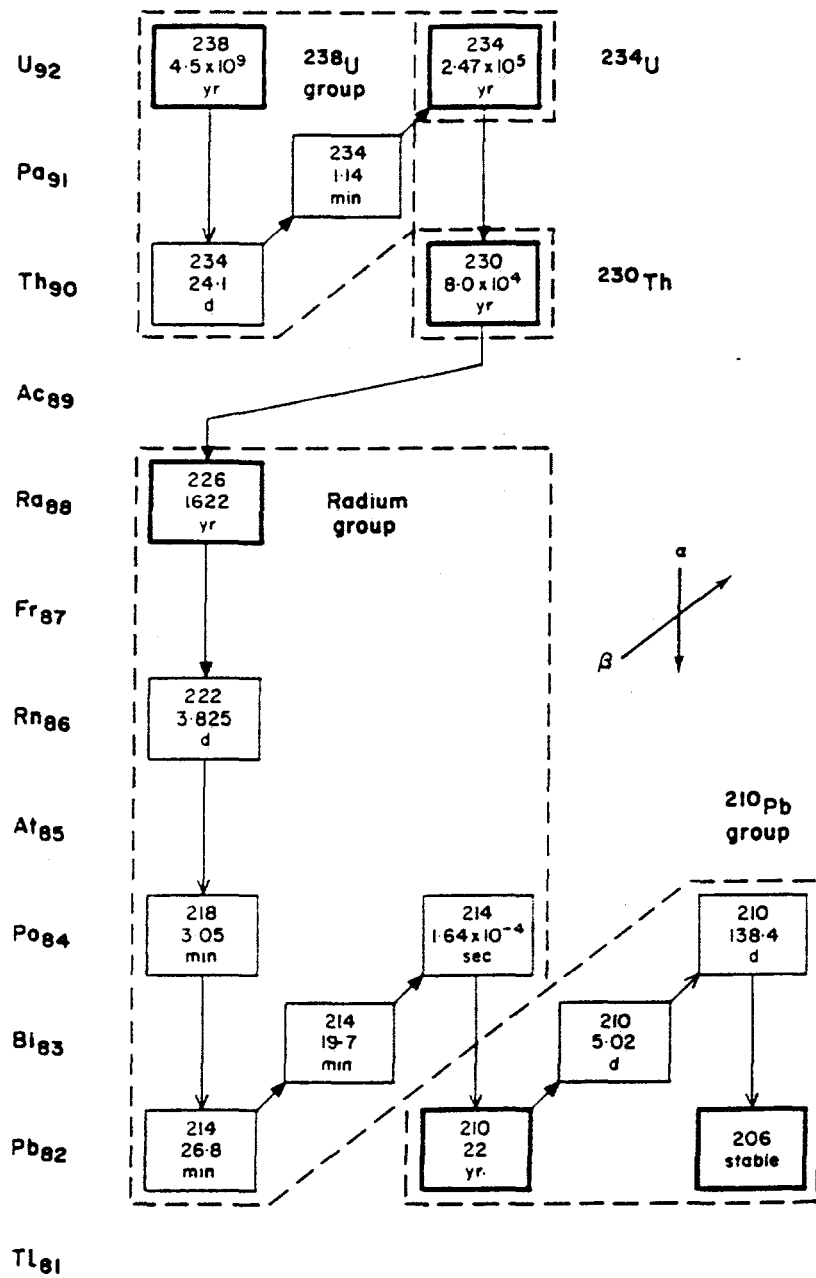


Figure 1. The uranium-238 decay series, after Rose et al. (1979)

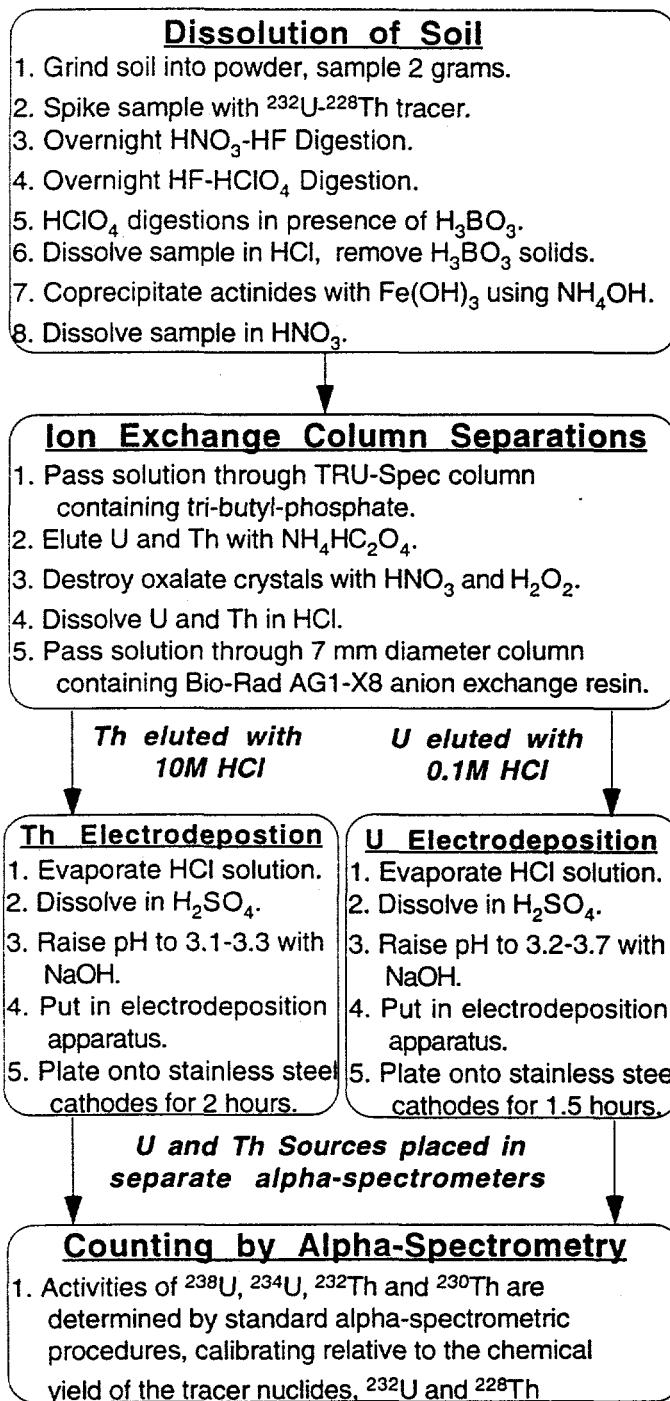


Figure 2. Summary of the procedures involved in the analyses of the total soil sample for ^{238}U , ^{234}U , ^{232}Th and ^{230}Th .

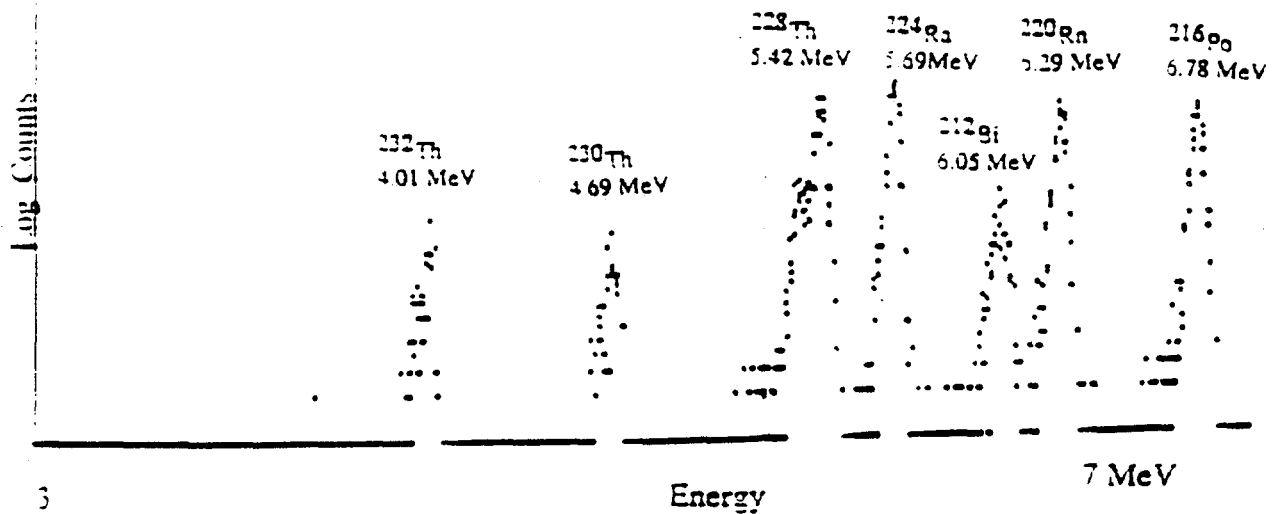


Figure 3. Alpha spectrum. Sample NY-1-2, thorium, with 10 pCi ^{228}Th tracer, 3-7 MeV, EG&G ORTEC analyzer.

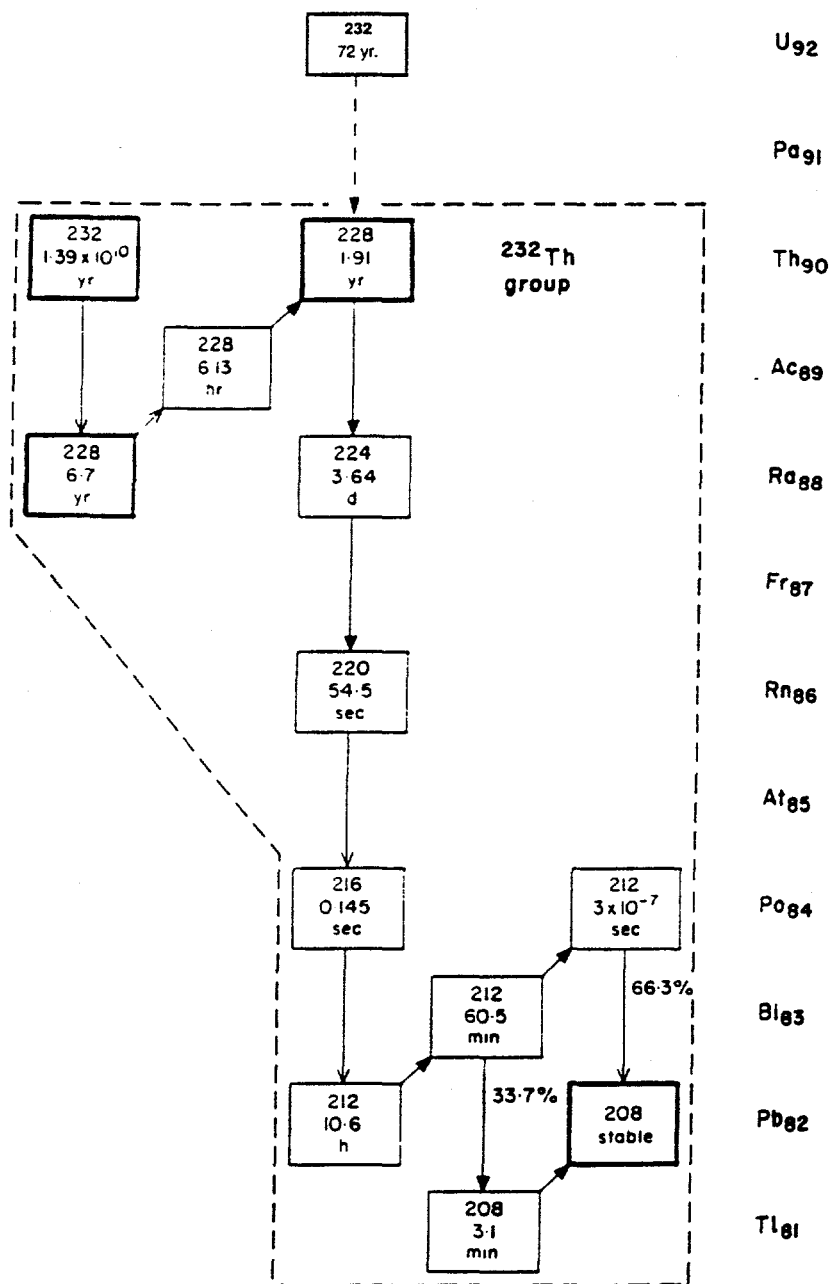


Figure 4. The thorium (4n) decay series, plus ^{232}U decay to ^{228}Th , after Rose et al. (1979)

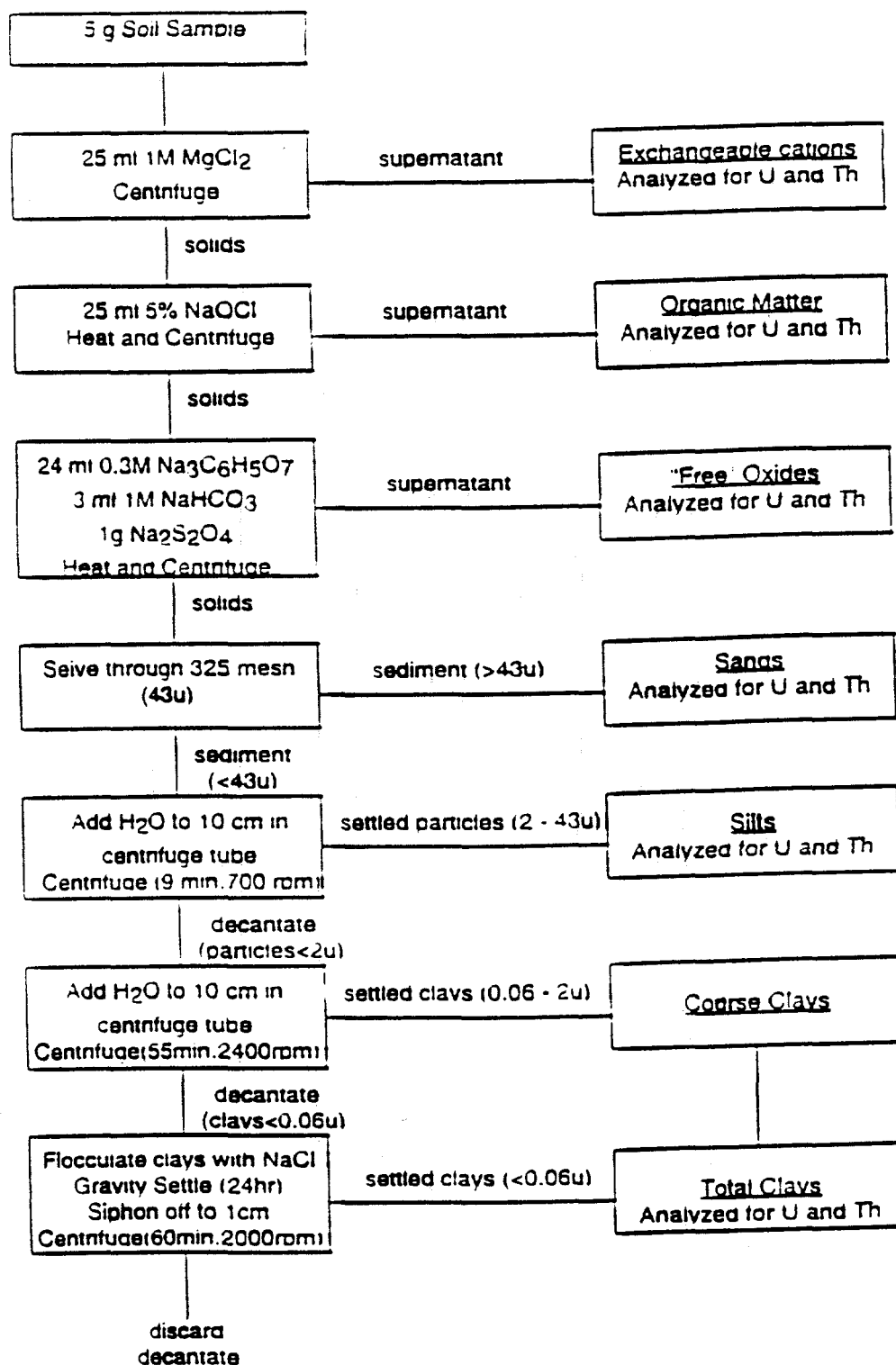
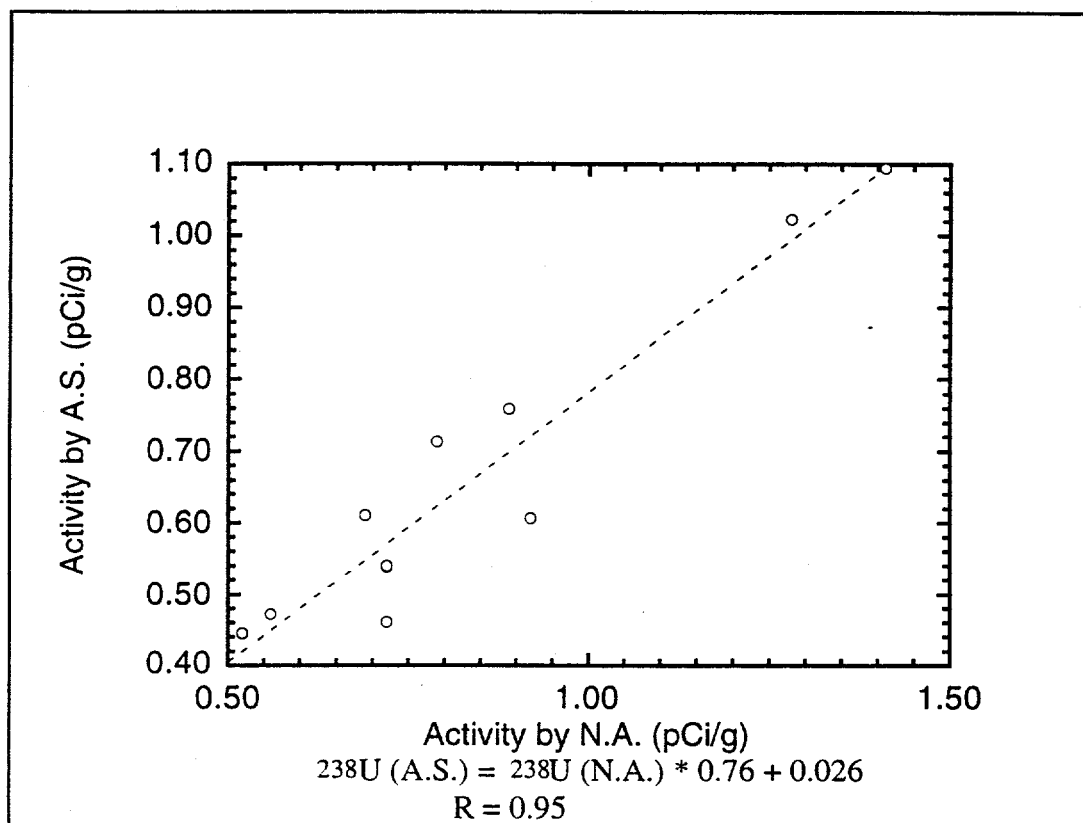


Figure 5. Flow Chart for progressive selective leaches.



-R = correlation coefficient
 A.S. = alpha spectrometry
 N.A. = neutron activation

Figure 6. Total soil sample activity measurements of ^{238}U by alpha spectrometry vs. neutron activation for the NY-1 and IL-1 profiles.
 R = correlation coefficient
 A.S. = alpha spectrometry
 N.A. = neutron activation

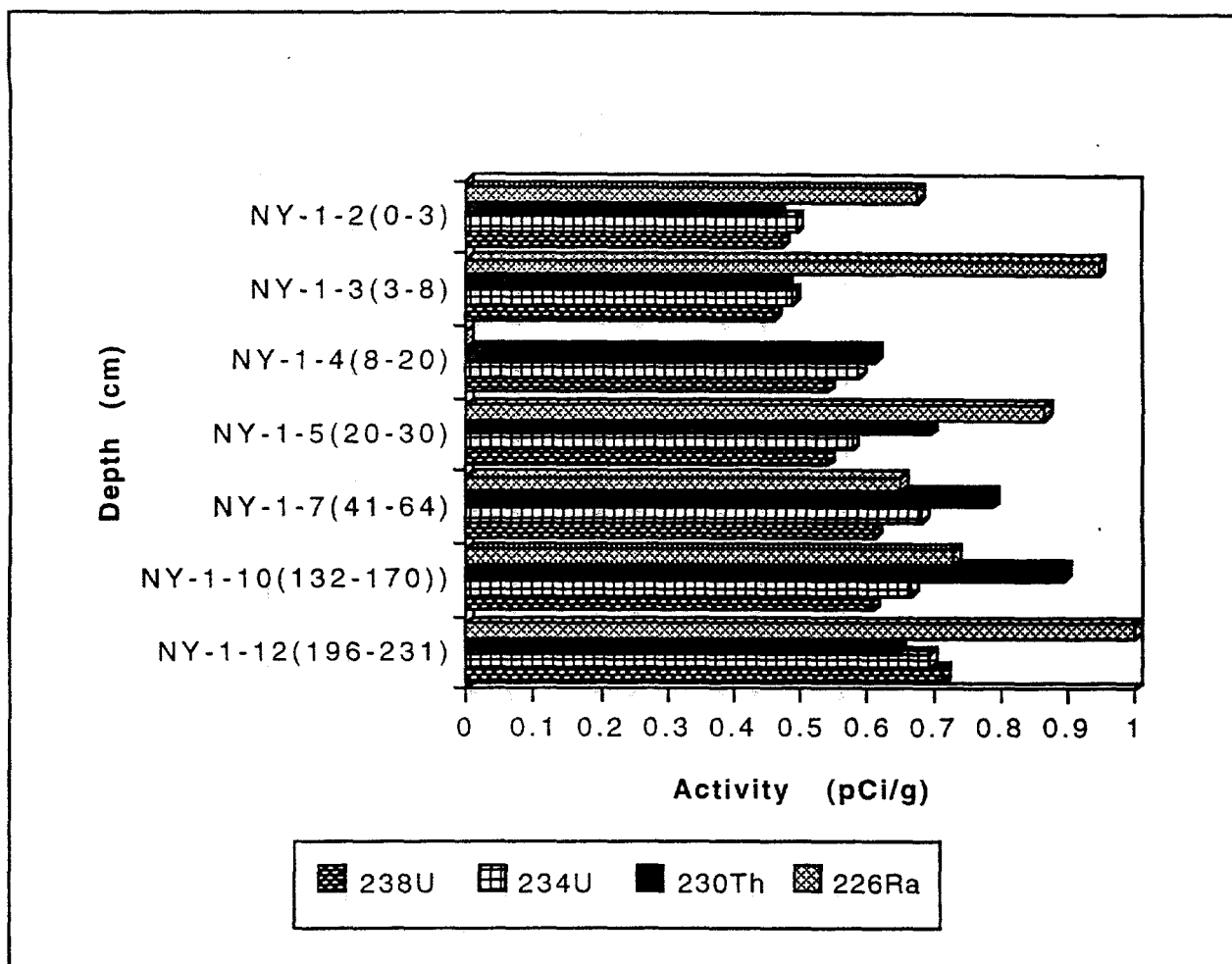


Figure 7. Depth in soil profile vs. activity of ^{226}Ra , ^{230}Th , and ^{234}U relative to their parent ^{238}U for samples from site NY-1.

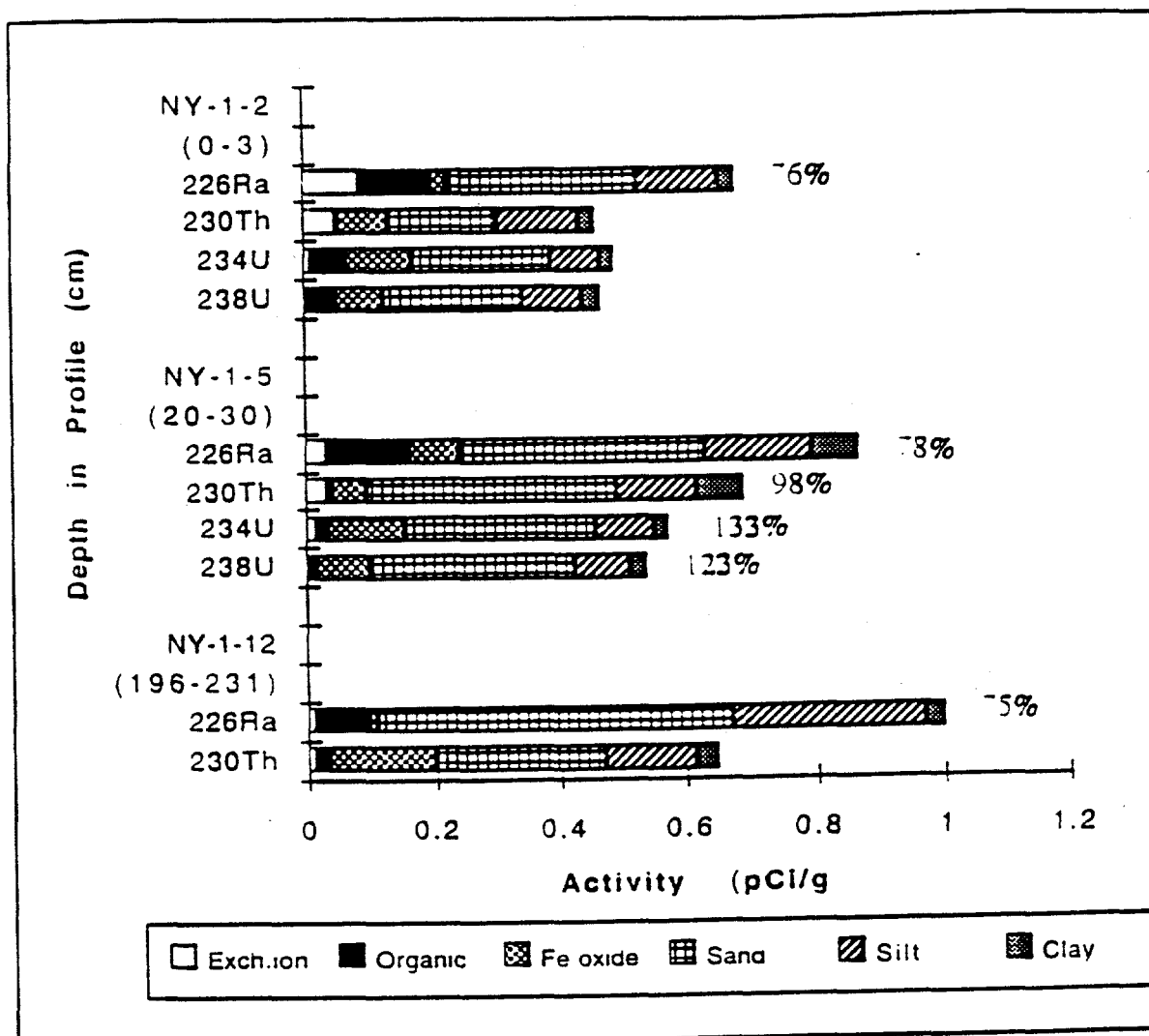


Figure 8. Depth in soil profile vs. activity in selective leaches of ^{226}Ra , ^{230}Th and ^{234}U relative to their parent ^{238}U for samples from site NY-1. The values are normalized to 100% of the activity for each horizon. The analytical totals are shown as % relative to these total values.

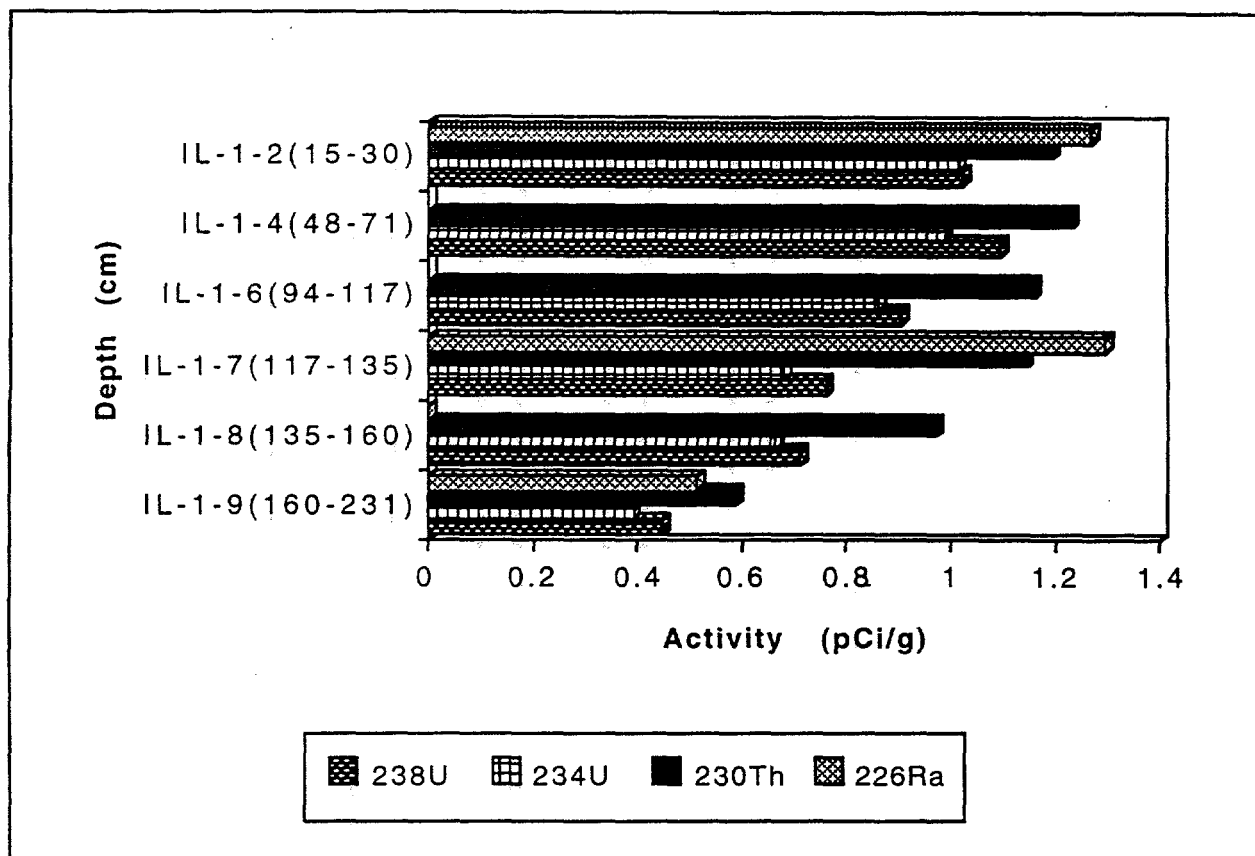


Figure 9. Depth in soil profile vs. activity of ^{226}Ra , ^{230}Th , and ^{234}U relative to their parent ^{238}U for samples from site IL-1.

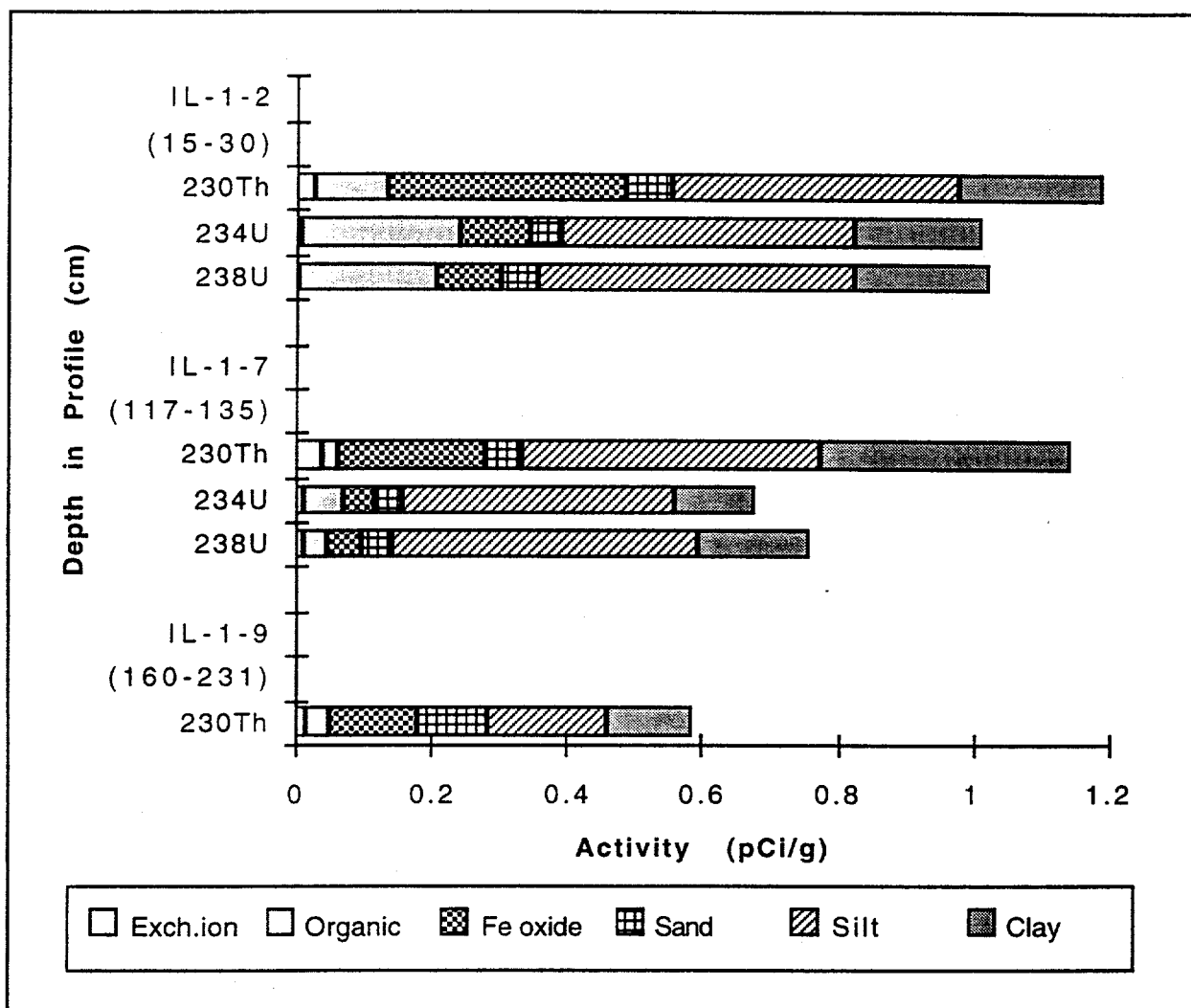


Figure 10. Depth in soil profile vs. activity in selective leaches of ^{230}Th and ^{234}U relative to their parent ^{238}U for samples from site IL-1. The values are normalized to 100% of the activity for each horizon. The analytical totals are shown as % relative to these total values.

Table 1 Alpha decay characteristics of natural uranium and thorium, radiochemical yeild monitor (spike) and some of their alpha-emmitting daughters.*

Radionuclide	Half-life	Energy (MeV) •	Intensity (%)
²³⁸U	4.468 x 10 ⁹ yr	4.147 4.196	23 77
²³⁵U	7.038 x 10 ⁸ yr	4.217 4.325 4.344 4.364 4.370 4.396 4.414 4.502 4.556 4.598	5.7 4.6 1.5 11 6 55 2.1 1.7 4.2 5.0
²³⁴U	2.445 x 10 ⁵ yr	4.724 4.776	27.5 72.5
²³²U	72 yr	5.264 5.320	31.2 68.6
²³²Th	1.405 x 10 ¹⁰ yr	3.952 4.010	23 77
²³⁰Th	7.54 x 10 ⁴ yr	4.621 4.688	23.4 76.3
²²⁸Th	1.913 yr	5.341 5.423	26.7 72.7
²²⁴Ra	3.66 d	5.449 5.686	4.9 95.1
²²⁰Rn	55.6 s	6.288	99.9
²¹⁶Po	0.15 s	6.779	99.9979
²¹²Bi	60.5	6.05 6.09	25 9.6

*Values taken from Browne and Firestone (1986)

•Only energies with intensities > 1% of the total nuclide energy are given.

Table 2. Comparison of activity values obtained
using method in text and those given for
a NIST standard reference material.

SRM 4354	Stated Method	NIST Values *=certified	Tolerance Limits (%)
^{238}U (pCi/g)	0.437 ± 0.016	0.470*	8
^{234}U (pCi/g)	0.441 ± 0.017	0.514	-
^{230}Th (pCi/g)	0.353 ± 0.029	0.351	-

Table 3. Soil properties and ^{226}Ra activity values for the <2 mm fraction from the NY-1 and IL-1 profiles.

Sample	Horiz	Depth cm	Sand wt %	Silt wt %	Clay wt %	Fe-ox wt %	Org.C wt %	^{226}Ra pCi/g
NY-1-2	A	0-3	84	12	3.5	0.6	6.3	0.626
NY-1-3	Bhs	3-8				1.0	1.0	0.946
NY-1-4	Bs1	8-20				0.9	0.7	ND
NY-1-5	Bs2	20-30	82	13	4.7	0.9	0.5	0.845
NY-1-7	CB	41-64				0.7	0.4	0.649
NY-1-10	2C2	132-170				0.4	0.02	0.730
NY-1-12	3C4	196-231	80	17	3.5	0.5	0.02	1.000
IL-1-1	Ap	0-15				1.6	5.61	1.595
IL-1-2	A2	15-30	3.4	58	39	1.5	2.55	1.270
IL-1-4	Bt1	48-71				2.0	0.76	ND
IL-1-5	Bt2	71-94				2.0	0.58	1.649
IL-1-7	Bt4	117-135	22	48	30	1.8	0.33	1.297
IL-1-9	2C	160-231	69	15	17	1.5	0.17	0.514

Note: Data for Fe-oxide, Organic C and ^{226}Ra from Greeman (1992). ND = not determined.

Table 4. Activity values ($\pm 1\sigma$) and tracer yield ($\pm 1\sigma$)
from α -spectrometric analysis of the NY-1
profile.

sample	horizon	^{238}U [pCi/g]	^{234}U [pCi/g]	^{230}Th [pCi/g]	^{232}Th [pCi/g]	Th yield [%]	U yield [%]
NY-1-2	A	.452 \pm .013	.452 \pm .013	.442 \pm .013	.315 \pm .010	69.2	80.7
		.481 \pm .027	.519 \pm .028	1.09 \pm .09*	.460 \pm .0??	24.8	84.1
		.482 \pm .034	.512 \pm .035	.483 \pm .041	.525 \pm .043	38.5	52.5
		(.472)	(.494)	(.463)	(.433)		
NY-1-3	Bhs	.401 \pm .030	.416 \pm .031	.417 \pm .029	.372 \pm .027	63.9	52.5
		.467 \pm .030	.494 \pm .031	.539 \pm .062	.516 \pm .045	34.5	65.0
		.483 \pm .060	.465 \pm .059	-	-	<15	16.8
		.492 \pm .052	.577 \pm .057	-	-	<15	61.2
		(.461)	(.488)	(.478)	(.444)		
NY-1-4	Bs1	.538 \pm .013	.587 \pm .014	.595 \pm .015	.545 \pm .014	70.6	82.6
		-	-	.627 \pm .056	.682 \pm .060	74.8	<15
		(.538)	(.587)	(.611)	(.614)		
NY-1-5	Bs2	.531 \pm .019	.528 \pm .019	.655 \pm .021	.544 \pm .018	43.8	48.7
		.548 \pm .029	.625 \pm .031	.760 \pm .069	.631 \pm .061	24.2	86.4
		-	-	.668 \pm .059	.583 \pm .054	28.5	<15
		(.540)	(.577)	(.694)	(.586)		
NY-1-7	CB	-	-	.695 \pm .023	.644 \pm .022	36.8	<15
		.607 \pm .039	.655 \pm .041	.737 \pm .069	.867 \pm .077	24.1	65.9
		.613 \pm .032	.711 \pm .035	1.033 \pm .090	.572 \pm .059	23.2	82.7
		-	-	.672 \pm .104	.434 \pm .078	21.8	<15
		(.610)	(.683)	(.784)	(.629)		
NY-1-10	2C2	-	-	.713 \pm .038	.670 \pm .036	54.1	<15
		.617 \pm .034	.675 \pm .036	.964 \pm .103	1.225 \pm .120	15.8	71.0
		.597 \pm .046	.655 \pm .049	1.001 \pm .092	1.508 \pm .090	46.2	78.9
		(.607)	(.665)	(.893)	(1.134)		
NY-1-12	3C4	.726 \pm .053	.629 \pm .048	.587 \pm .066	.764 \pm .079	19.1	36.5
		.616 \pm .037	.650 \pm .038	.706 \pm .054	.560 \pm .047	35.5	61.6
		.683 \pm .041	.696 \pm .042	-	-	<15	55.0
		.826 \pm .082	.799 \pm .080	-	-	<15	47.8
		(.713)	(.693)	(.647)	(.662)		
Profile TWA		.616	.650	.735	.774		

*This ^{230}Th value was omitted from the calculation of horizon average due to its extreme variance from the other ^{230}Th activity determinations.

() represents average activity for the horizon.

TWA = Thickness Weighted Average

Table 5. Activity values ($\pm 1\sigma$) and tracer yield ($\pm 1\sigma$)
from α -spectrometric analysis of selective
leaches from the NY-1-2 horizon.

sample	soil fraction	^{238}U (pCi/g)	^{234}U (pCi/g)	^{230}Th (pCi/g)	^{232}Th (pCi/g)	U yield (%)	Th yield (%)
NY-1-2 A	Exch.ion	.010 \pm .002	.016 \pm .002	.110 \pm .007	.053 \pm .005	62.1 \pm 2.7	17.3 \pm 0.5
	Organic	.049 \pm .005	.048<.080	.008 \pm .001	.003 \pm .001	15.8 \pm 0.7	109 \pm 3.0
	Fe-oxide	.093 \pm .011	.139 \pm .014	.134 \pm .017	.030 \pm .008	8.7 \pm 0.4	4.3 \pm 0.2
	Sand	.242 \pm .011	.255 \pm .011	.277 \pm .019	.065 \pm .008	75.9 \pm 3.6	14.9 \pm 0.5
	Silt	.118 \pm .007	.101 \pm .009	.290 \pm .010	.048 \pm .003	50.5 \pm 2.5	46.2 \pm 1.3
	Clay	.044 \pm .005	.025<.026	.039 \pm .004	.024 \pm .003	64.1 \pm 2.9	45.6 \pm 1.4
NY-1-2 B	Exch.ion	.009 \pm .001	.011 \pm .002	.040 \pm .005	.021 \pm .003	46.5 \pm 2.0	12.1 \pm 0.3
	Organic	.063<.072	.093<.107	-	-	10.0 \pm 0.5	0
	Fe-oxide	-	-	-	-	0	0
	Sand	.300 \pm .032	.315 \pm .039	.281 \pm .008	.137 \pm .005	11.4 \pm 0.5	61.4 \pm 1.7
	Silt	.070 \pm .006	.051<.060	.124 \pm .009	.052 \pm .005	48.8 \pm 2.3	43.4 \pm 1.3
	Clay	.030 \pm .003	.031 \pm .004	.047 \pm .003	.022 \pm .002	57.2 \pm 2.6	32.9 \pm 0.9
NY-1-2 average	Exch.ion	.010	.014	.075	.037	54.3	14.7
	Organic	.049	.071	.008	.003	12.9	54.5
	Fe-oxide	.093	.139	.134	.030	4.4	2.2
	Sand	.271	.285	.279	.084	43.7	38.2
	Silt	.118	.101	.207	.050	49.7	44.8
	Clay	.037	.031	.043	.023	60.7	39.3
	Σ phases	0.578	0.641	0.746	0.227		

Values to the left of the "<" represent measurements which fell below the lower limit of detection (LLD), shown on the right, which is defined by Jester (1993) as 4.66 times the standard deviation of the background activity (σ_{bkg}).

- represents soil phases for which no activity data is available (0% analytical recovery of tracer)

**Table 6. Activity values ($\pm 1\sigma$) and tracer yield ($\pm 1\sigma$)
from α -spectrometric analysis of selective
leaches from the NY-1-5 horizon.**

soil fraction	^{238}U (pCi/g)	^{234}U (pCi/g)	^{230}Th (pCi/g)	^{232}Th (pCi/g)	U yield (%)	Th yield (%)
Exch.ion	.007<.008	.019 \pm .003	.029 \pm .003	.008 \pm .001	73.8 \pm 3.2	60.1 \pm 1.7
Organic	.014<.070	.026<.093	.010 \pm .001	.002 \pm .001	20.2 \pm 1.0	84.6 \pm 2.4
Fe-oxide	.102 \pm .022	.161 \pm .030	.053 \pm .006	.023 \pm .004	9.6 \pm 0.4	16.3 \pm 0.5
Sand	.398 \pm .013	.406 \pm .014	.391 \pm .013	.466 \pm .015	60.2 \pm 2.7	73.9 \pm 2.2
Silt	.110 \pm .009	.124 \pm .014	.125 \pm .007	.083 \pm .006	32.2 \pm 1.5	34.6 \pm 1.0
Clay	.033 \pm .003	.031 \pm .004	.070 \pm .004	.061 \pm .003	75.9 \pm 3.5	75.6 \pm 2.1
Σ phases	0.663	0.767	0.678	0.643		

**Table 7. Activity values ($\pm 1\sigma$) and tracer yield ($\pm 1\sigma$)
from α -spectrometric analysis of selective
leaches from the NY-1-12 horizon.**

soil fraction	^{238}U (pCi/g)	^{234}U (pCi/g)	^{230}Th (pCi/g)	^{232}Th (pCi/g)	U yield (%)	Th yield (%)
Exch.ion	.011<.012	.013 \pm .003	.016 \pm .003	.008 \pm .001	71.3 \pm 3.1	55.3 \pm 1.5
Organic	.017<.023	.036<.051	.026 \pm .002	.004 \pm .001	71.1 \pm 3.1	88.3 \pm 2.4
Fe-oxide	-	-	.205 \pm .037	.106 \pm .026	0	2.8 \pm 0.2
Sand	.294 \pm .015	.301 \pm .014	.333 \pm .013	.174 \pm .008	53.3 \pm 2.4	35.2 \pm 1.0
Silt	.151 \pm .011	.122 \pm .013	.177 \pm .010	.095 \pm .007	39.3 \pm 1.8	25.6 \pm 0.8
Clay	.029 \pm .003	.035 \pm .005	.038 \pm .004	.039 \pm .004	66.6 \pm 3.0	46.8 \pm 1.4
Σ phases	.502+Fe-Ox	.507+Fe-Ox	0.795	0.426		

Values to the left of the "<" represent measurements which fell below the lower limit of detection (LLD), shown on the right, which is defined by Jester (1993) as 4.66 times the standard deviation of the background activity (σ_{bkg}).

- represents soil phases for which no activity data is available (0% analytical recovery of sample)

Table 8. Normalized ^{238}U , ^{234}U , ^{230}Th and ^{226}Ra activity values and daughter/parent activity ratios in select phases of horizons from site NY-1.

Horizon	Nuclide	Ex.ion	Organic	Fe-Ox	Sand	Silt	Clay
NY-1-2	^{238}U	0.008	0.040	0.076	0.221	0.096	0.030
	^{234}U	0.011	0.055	0.107	0.219	0.078	0.024
	^{230}Th	0.047	0.005	0.083	0.173	0.128	0.026
	^{226}Ra	0.088	0.115	0.023	0.304	0.128	0.024
	$\frac{^{234}\text{U}}{^{238}\text{U}}$	1.375	1.375	1.408	0.991	0.813	0.800
	$\frac{^{230}\text{Th}}{^{234}\text{U}}$	4.273	0.091	0.776	0.790	1.641	1.083
	$\frac{^{226}\text{Ra}}{^{230}\text{Th}}$	1.872	23.00	0.277	1.757	1.00	0.923
NY-1-5	^{238}U	0.006	0.011	0.083	0.324	0.090	0.027
	^{234}U	0.014	0.019	0.121	0.305	0.093	0.023
	^{230}Th	0.030	0.010	0.054	0.400	0.127	0.072
	^{226}Ra	0.031	0.130	0.082	0.389	0.164	0.071
	$\frac{^{234}\text{U}}{^{238}\text{U}}$	2.333	1.727	1.458	0.941	1.033	0.852
	$\frac{^{230}\text{Th}}{^{234}\text{U}}$	2.142	0.526	0.446	1.311	1.366	3.130
	$\frac{^{226}\text{Ra}}{^{230}\text{Th}}$	1.033	13.00	1.519	0.973	1.291	0.986
NY-1-12	^{230}Th	0.013	0.021	0.167	0.271	0.144	0.031
	^{226}Ra	0.012	0.082	0.015	0.560	0.300	0.032
	$\frac{^{226}\text{Ra}}{^{230}\text{Th}}$	0.923	3.905	0.090	2.066	2.083	1.032
	^{230}Th						

^{226}Ra activities are from Greeman (1992).

Table 9. Concentrations of normalized nuclide activities within fractions for the measured horizons of the NY-1 profile. Values are calculated as the normalized nuclide content divided by the mass of the phase.

Sample	^{238}U (pCi/g)	^{234}U (pCi/g)	^{230}Th (pCi/g)	^{226}Ra (pCi/g)
NY-1-2				
Sand	0.26	0.26	0.20	0.36
Silt	0.80	0.65	1.06	1.0
Clay	0.85	0.68	0.74	0.7
Fe-ox.	13.	18	14	3.8
Org. C	0.6	0.9	0.1	1.8
Exch.	5.7	7.8	33	62
NY-1-5				
Sand	0.40	0.37	0.49	0.47
Silt	0.69	0.71	0.97	1.2
Clay	0.57	0.48	1.5	1.5
Fe-ox.	9	13	6.	9
Org. C	2.2	3.8	2.0	26
Exch.	18	42	90	93
NY-1-12				
Sand			0.34	0.84
Silt			0.9	1.8
Clay			0.9	0.9
Fe-ox.			33	3
Org. C			>100*	>100*
Exch.			38	35

*Organic matter in NY-1-12 is recorded as 0.02%, which is so low that it is probably not the dominant host for nuclides released in this step.

Source of data: Mass of phases is based on Table 3 and data in Greeman (1992). The mass of exchangeable ions was assumed to be Al, because acidity was dominant in the exchange sites.

Table 10. Activity values ($\pm 1\sigma$) and tracer yield ($\pm 1\sigma$)
from α -spectrometric analysis of the IL-1
profile.

sample	horizon	^{238}U (pCi/g)	^{234}U (pCi/g)	^{230}Th (pCi/g)	^{232}Th (pCi/g)	Th yield (%)	U yield (%)
IL-1-2	Ap	1.023 \pm .029	1.013 \pm .031	1.191 \pm .052	.693 \pm .036	17.9 \pm 0.6	83.1 \pm 3.6
IL-1-4	A2	1.095 \pm .030	.988 \pm .034	1.231 \pm .036	.811 \pm .026	68.7 \pm 1.9	93.7 \pm 4.1
IL-1-6	Bt1	.903 \pm .027	.860 \pm .026	1.159 \pm .044	.726 \pm .032	20.7 \pm 0.6	74.3 \pm 3.3
IL-1-7	Bt2	.759 \pm .024	.680 \pm .023	1.143 \pm .041	1.025 \pm .038	57.6 \pm 1.7	83.7 \pm 3.8
IL-1-8	Bt4	.712 \pm .023	.664 \pm .024	.970 \pm .044	.645 \pm .034	31.2 \pm 1.0	63.6 \pm 2.8
IL-1-9	2C	.445 \pm .016	.392 \pm .022	.587 \pm .019	.377 \pm .014	51.0 \pm 1.4	72.7 \pm 3.2
Profile TWA		0.711	0.653	0.910	.612		

TWA = Thickness Weighted Average

Table 11. Activity values ($\pm 1\sigma$) and tracer yield ($\pm 1\sigma$)
from α -spectrometric analysis of selective
leaches from the IL-1-2 horizon.

soil fraction	^{238}U (pCi/g)	^{234}U (pCi/g)	^{230}Th (pCi/g)	^{232}Th (pCi/g)	Th yield (%)	U yield (%)
Exch.ion	.005 \pm .001	<.008	.033 \pm .003	.023 \pm .003	49.2 \pm	56.6 \pm
Organic	.214 \pm .031	.250 \pm .034	.125 \pm .053	.069 \pm .016	2.0 \pm	4.9 \pm
Fe-oxide	.102 \pm .006	.111 \pm .009	.411 \pm .030	.328 \pm .015	38.8 \pm	64.6 \pm
Sand	.055 \pm .007	.048 \pm .006	.082 \pm .007	.031 \pm .004	43.7 \pm	42.1 \pm
Silt	.497 \pm .015	.463 \pm .019	.492 \pm .021	.393 \pm .017	48.3 \pm	73.6 \pm
Clay	.213 \pm .010	.201 \pm .010	.249 \pm .018	.156 \pm .016	46.3 \pm	54.9 \pm
Σ phases	1.086	1.079	1.392	1.000		

**Table 12. Activity values ($\pm 1\sigma$) and tracer yield ($\pm 1\sigma$)
from α -spectrometric analysis of selective
leaches from the IL-1-7 horizon.**

soil fraction	^{238}U (pCi/g)	^{234}U (pCi/g)	^{230}Th (pCi/g)	^{232}Th (pCi/g)	Th yield (%)	U yield (%)
Exch.ion	.012 \pm .001	.015 \pm .003	.038 \pm .004	.010 \pm .002	31.1 \pm 0.9	65.4 \pm 2.9
Organic	.034<.039	.065 \pm .013	.023 \pm .002	.023 \pm .002	76.4 \pm 2.1	47.2 \pm 2.1
Fe-oxide	.054 \pm .005	.049 \pm .008	.221 \pm .008	.181 \pm .007	74.5 \pm 2.1	85.6 \pm 3.7
Sand	.048 \pm .010	.049<.095	.058 \pm .003	.040 \pm .003	49.3 \pm 1.4	36.1 \pm 1.6
Silt	.471 \pm .015	.459 \pm .016	.447 \pm .016	.417 \pm .015	38.7 \pm 1.2	63.9 \pm 2.8
Clay	.170 \pm .007	.136 \pm .007	.372 \pm .041	.198 \pm .030	2.6 \pm 0.1	77.7 \pm 3.4
Σ phases	0.789	0.773	1.159	0.869		

Values to the left of the "<" represent measurements which fell below the lower limit of detection (LLD), shown on the right, which is defined by Jester (1993) as 4.66 times the standard deviation of the background activity (σ_{bkg}).

**Table 13. Activity values ($\pm 1\sigma$) and tracer yield ($\pm 1\sigma$)
from α -spectrometric analysis of selective
leaches from the IL-1-9 horizon.**

soil fraction	^{238}U (pCi/g)	^{234}U (pCi/g)	^{230}Th (pCi/g)	^{232}Th (pCi/g)	Th yield (%)	U yield (%)
Exch.ion	.005 \pm .001	.006 \pm .002	.022 \pm .003	.007 \pm .002	71.6 \pm 2.1	90.4 \pm 3.9
Organic	-	-	.051 \pm .004	0	62.2 \pm 1.8	0
Fe-oxide	.032 \pm .004	.045 \pm .007	.196 \pm .010	.106 \pm .007	41.9 \pm 1.3	58.1 \pm 2.6
Sand	.122 \pm .007	.119 \pm .008	.150 \pm .009	.130 \pm .009	39.9 \pm 1.2	47.9 \pm 2.1
Silt	.127 \pm .014	.104 \pm .018	.266 \pm .018	.256 \pm .016	24.2 \pm 0.8	17.5 \pm 0.9
Clay	.133 \pm .007	.126 \pm .007	.189 \pm .017	.219 \pm .016	18.2 \pm 0.6	47.4 \pm 2.1
Σ phases	0.419+org	0.400+org	0.874	0.718		

- represents soil phases for which no activity data is available (0% analytical recovery of sample)

Table 14. Normalized ^{238}U , ^{234}U , and ^{230}Th activity values (pCi/g) and daughter/parent activity ratios in select phases of horizons from IL-1.

Horizon	Nuclide	Ex.ion	Organic	Fe-Ox	Sand	Silt	Clay
IL-1-2	^{238}U	0.005	0.202	0.096	0.052	0.468	0.201
	^{234}U	0.006	0.235	0.104	0.045	0.435	0.189
	^{230}Th	0.028	0.107	0.352	0.070	0.421	0.213
	$\frac{^{234}\text{U}}{^{238}\text{U}}$	1.200	1.163	1.083	0.865	0.929	0.940
	$\frac{^{230}\text{Th}}{^{234}\text{U}}$	4.667	0.455	3.385	1.556	0.968	1.127
IL-1-7	^{238}U	0.012	0.033	0.052	0.046	0.453	0.163
	^{234}U	0.013	0.057	0.043	0.043	0.404	0.120
	^{230}Th	0.037	0.023	0.218	0.057	0.441	0.367
	$\frac{^{234}\text{U}}{^{238}\text{U}}$	1.083	1.727	0.827	0.935	0.891	0.736
	$\frac{^{230}\text{Th}}{^{234}\text{U}}$	2.846	0.403	4.192	1.326	1.092	3.058
IL-1-12	^{230}Th	0.015	0.034	0.132	0.101	0.179	0.127

^{226}Ra values are from Greeman (1992).

Table 15. Concentrations of normalized nuclide activities within fractions for the measured horizons of the IL-1 profile. Values are calculated as the normalized nuclide content divided by the mass of the phase.

Sample	²³⁸ U (pCi/g)	²³⁴ U (pCi/g)	²³⁰ Th (pCi/g)
IL-1-2			
Sand	1.5	1.3	2.1
Silt	0.80	0.75	0.72
Clay	0.51	0.48	0.54
Fe-ox.	6.4	6.9	23
Org. C	7.9	9.2	4.2
Exch.	1.6	1.9	9
IL-1-7			
Sand	0.21	0.19	0.26
Silt	0.94	0.84	0.92
Clay	0.54	0.40	1.22
Fe-ox.	2.9	2.4	12
Org. C	10	17	7
Exch.	3.5	3.7	1
IL-1-9			
Sand			0.14
Silt			1.19
Clay			0.75
Fe-ox.			9
Org. C			20
Exch.			10

Source of data: Based on normalized nuclide data, and mass of phases in table 3 and in Greeman (1992). The mass of exchangeable ions was calculated by assuming that Ca was the dominant exchangeable ion, as found by measurements.

Appendix C
ANALYTICAL DATA FOR SOIL PROFILES

= cation exchange capacity.

[illegible]

6-10-8								0.13	4.83
6-10-9	4.7	2	7.8	0.21	0.8	0.8	9.6	0.14	4.34
8-99-2	4.4	29	16.3	0.17	0.6	1.5	17.3	4.48	1.2
8-99-3	4.6	55	9.0	0.07	0.3	0.5	9.9	0.98	1.2
8-99-4								0.43	
8-99-5	4.7	21	5.3	0.10	0.4	0.8	6.5	0.26	1.6
8-99-6								0.13	
8-99-7	5.1	20	5.3	0.32	2.3	2.5	10.4	0.10	2.3
8-99-8								0.10	
8-99-9	5.5	27	0.15	0.15	3.4	3.5	9.8	0.08	2.0
8-99-10								0.11	
8-99-11								0.08	
8-99-12								0.09	1.6
TN-1-2	5.3	12	4.1	0.35	1.3	3.0	8.8	3.10	1.5
TN-1-3	5.1	7	4.1	0.17	0.6	1.0	5.9	1.36	1.7
TN-1-4	4.9	7	4.1	0.12	0.5	0.8	5.5	0.66	2.2
TN-1-5								0.34	
TN-1-6	5.1	1	15.1	0.28	1.0	0.8	17.0	0.26	7.0
TN-1-7								0.10	
TN-1-8	5.0	1	16.3	0.17	0.5	0.8	16.5	0.17	6.8
TN-1-9								0.22	
TN-1-10								0.08	6.0
TN-1-11	4.9	1	7.8	0.07	0.3	0.5	8.7	0.05	3.7
14-84-1			14.7	0.43	0.4	3.5	19.1	2.49	1.98
14-84-2			8.2	0.32	0.4	4.0	13.0	0.92	2.18
14-84-3			7.9	0.19	0.2	2.3	10.6	0.39	2.30
14-84-4			10.7	0.13	0.2	1.8	12.9	0.32	2.36
14-84-5			12.8	0.13	0.2	1.7	14.9	0.34	2.38
14-84-6			11.2	0.10	0.2	1.5	13.0	0.26	2.64
14-84-7			9.3	0.08	0.2	1.2	10.8	0.25	1.75
14-84-8			8.0	0.13	0.4	1.9	10.5	0.37	3.05
14-84-9			8.4	0.18	0.3	1.9	10.9	0.41	4.47

For samples 14-80, 14-81, 14-82 and 14-83, See Washington (1991)

Table C2. Radon concentration at the field sites
Values in pCi/L

<u>Site 14-84</u>	Deep	Shallow
11/30/88-3/15/89	1574	
3/15/89-6/13/89	1445	687
6/13/89-8/9/89	1426	394
8/9/89/10/11/89	981	1025
10/11/89-1/9/90	736	440
1/9/90-3/14/90	976	238

<u>Site TN-1</u>		
7/25/89-10/7/89	1112	1324
10/7/89-1/20/90	1166	1184
1/20/90-4/5/90	1760	1117
4/5/90-7/17/90	982	1050

<u>Site IL-1</u>		
6/27/89-9/11/89	1232	
9/11/89-11/9/89	761	529
11/9/89-2/10/90	696	
2/10/90-7/14/90	389	696

<u>Site NY-1</u>		
8/22/89-10/22/89	401	123
10/22/89-1/29/90	376	155
1/29/90-5/1/90	444	312

<u>Site NC-1</u>		
9/1/88-1/1/89	8674	
1/1/89-5/10/89	3471	
5/10/89-7/12/89	5795	2578
7/12/89-10/6/89	5476	2840
10/6/89-1/3/90	2006	888
1/3/90-3/23/90	2101	924
3/23/90-7/13/90	2180	992

<u>Site 6-10</u>		
8/22/88-11/29/88	1490	
11/29/88-4/5/89	1091	948
4/5/89-6/11/89	1447	796
6/11/89-8/10/89	1556	
8/10/89-10/9/89	1045	426
10/9/89-1/8/90	332	466
1/8/90-3/6/90	503	405
3/6/90-6/20/90	656	323
6/20/90-8/28/90	1127	549

<u>Site 6-11</u>		
8/22/88-11/29/88	691	
11/29/88-4/5/89	699	432
4/5/89-6/11/89	324	450
6/11/89-8/10/89	300	
8/10/89-10/9/89	226	257

10/9/89-1/8/90	157	377
1/8/90-3/6/90	237	327
3/6/90-6/20/90	180	262
6/20/90-8/28/90	276	249

Site 8-99		
8/1/89-10/9/89	1278	877
10/9/89-1/8/90	455	953
1/8/90-3/12/90	170	1261
3/12/90-6/21/90	102	637
6/21/90-8/29/90	1070	515

Table C3. Particle size distribution, coarse fragments, density and organic C for samples from the 14-80, 14-81, 14-82 and 14-83 profiles.

Sample #	SOIL PROPERTIES										
	Horizon	Depth	Particle	Texture	Structure	Roots	Coarse Frags (cm)				P.S.D.A.
	Desig.	(cm)	Ped				25.4-7.6	7.6-1.9	1.9-0.2	0.2	Sa (%) Si (%)
			Colour								
<hr/>											
8001	A	0.0-7.6 10YR 2/1		sil	2fgr M,12		0	94.78	180.5	1019.3	19.2 73.4
8002	E	7.6-33.0 10YR 6/3		sil	1fSBK M,12		0	15.82	352.0	2350.3	18.9 64.8
8003	EB	33.0-45.77.5YR 5/6		sic	2fSBK C,12		0	88.78	381.5	1594.4	14.8 54.8
8004	Bt1	45.7-61.05YR 5/6		c	3fABK C,12		0	24.90	464.5	2553.0	9.7 40.8
8005	Bt2	61.0-88.95YR 4/6		c	3mABK C,12		0	258.50	634.0	2019.5	9.1 26.3
8006	Bt3	88.9-125.5YR 5/6		c	3mABK F,1		0	198.78	567.0	2767.0	14.9 19.7
8007	Bt4	125.5-149.5YR 5/6		cl	1mSBK F,1		0	35.72	410.5	2397.3	27.9 13.4
8008	Bt5	149.9-182.5YR 5/6		sic	2mSBK F,1		0	50.00	917.5	2796.3	9.4 17.5
8009	BC	182.9-2547.5YR 5/6		sic	1cSBK		0	37.86	293.0	3367.6	4.1 45.5
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8101	A	0.0-7.6 10YR 2/1		sil	1fSBK M,12		0	0.00	160.5	2579.3	12.2 69.8
8102	E	7.6-25.4 10YR 5/3		sil	1fSBK M,12		0	0.00	94.0	3121.7	9.9 72.5
8103	EB	25.4-35.610YR 5/4		sic	1fSBK C,1		0	0.00	93.5	4066.0	8.8 71.0
8104	Bt	35.6-63.610YR 5/6		sic	2fSBK C,1		0	0.00	460.5	3785.0	9.1 64.6
8105	Bx1	63.6-104.10YR 4/4		sil	2vcPR F,1		0	0.00	652.5	4842.0	22.4 58.1
8106	Bx2	104.1-139.10YR 4/4		sil	2vcPR		0	88.11	894.0	4761.5	23.5 57.5
8107	BC	139.7-1627.5YR 4/6		cl	1mSBK		0	235.55	2736.5	3057.5	47.7 24.3
8108	2C1	162.6-1937.5YR 5/6		sic	1mPL		0	0.00	210.5	5426.0	10.6 58.2
8109	2C2	193.0-2237.5YR 5/6		sic	1mPL		0	43.51	213.0	4446.0	6.6 55.4
8110	2C3	223.5-2597.5YR 5/6		sic	MA		0	202.29	1678.0	3177.3	20.8 44.0
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8201	A	0.0-7.6 10YR 2/2		sil	2fGR			40.86	194.0	2376.5	48.6 41.9
8202	E	7.6-22.9 10YR 5/4		sil	1fSBK			152.29	405.5	4402.0	47.4 39.3
8203	Bw1	22.9-40.710YR 5/4		l	1fSBK			414.29	642.0	3789.0	48.2 38.5
8204	Bw2	40.7-61.010YR 5/6		sil	1mSBK			99.41	410.5	4999.0	42.9 42.2
8205	Bx1	61.0-78.710YR 5/3		l	1vcPR			370.95	1202.0	3935.0	57.4 31.3
8206	Bx2	78.7-104.10YR 5/6		sl	2vcPR			766.05	1064.0	3607.0	63.9 30.1
8207	Bx3	104.1-132.10YR 4/6		sl	3vcPR			2054.70	1314.5	3233.0	67.2 19.1
8208	Bx4	132.1-1577.5YR 5/6		sl	1coSBK			2486.30	949.5	4529.5	58.5 26.2
8209	Bx5	157.5-20310YR 4/4		sl	3vcPR			405.67	1467.0	4611.5	68.1 30.3
8210	Bx6	203.2-24310YR 4/4		sl	3vcPR			486.58	1330.0	4205.0	70.0 21.1
8211	2C	243.8-30410YR 5/4		ls	MA			4387.90	2689.0	2980.0	77.3 13.2
<hr/>											
8303	E	2.5-7.6 7.5YR 5/2		ls	1fSBK			431.96	723.0	3225.0	68.1 30.3
8304	Bhs	7.6-12.7 7.5YR 3/3		sl	1fSBK			48.88	343.0	2189.5	57.8 35.2
8305	Bs	12.7-22.910YR 5/6		sl	1fSBK			69.10	195.0	4646.5	57.6 33.8
8306	Bt1	22.9-43.210YR 5/5		sl	1fSBK			180.00	676.5	3808.5	58.4 29.9
8307	Bt2	43.2-71.110YR 5/5		sl	1mSBK			1071.90	835.0	4104.5	59.8 29.6
8308	Bt3	71.1-94.05YR 5/6		l	2fSBK			1380.60	1499.5	3450.0	51.9 22.9
8309	Bt4	94.0-121.5YR 5/3		cl	1mSBK		677.74	2454.80	1204.5	4749.5	54.1 23.9
8310	Bc	121.9-1377.5YR 5/6		sl	1mSBK		1402.90	1368.00	866.5	3310.5	64.3 21.3
8311	C1	137.2-16710YR 5/6		ls	SGR		4591.90	3851.20	1236.5	2530.5	69.3 30.3
8312	C2	167.6-20710YR 5/6		ls	SGR			5597.60	1735.5	1826.0	72.2 18.7

[illegible]

#	Organic C /		Slods				CoLE	/			
	C1 (%)	C2 (%)	1/3 Bar Bulk Rho H2O (%)	Dry Rho (gm/cm3)	Porosity (%)	Sma Rho (gm/cm3)		U (ppm)	V +/-	Th (ppm)	Th +/-
8001	7.3	2.42						4.41	0.13	10	1.3
8002	16.3	0.40						5.70	0.11	8.5	0.1
8003	10.4	0.43						6.27	0.15	~3.74	
8004	50.6	0.14						7.39	0.21	11	0.1
8005	54.6	0.13						7.19	0.19	~10.00	
8006	55.4	0.06						7.96	0.24	~4.14	
8007	58.7	0.07						6.57	0.20	11	0.1
8008	73.1	0.04						8.02	0.24	~6.04	
8009	50.4	0.05						7.86	0.24	11	0.1
8101	18.0	6.86						3.30	0.13	4.21	
8102	17.4	0.45						3.75	0.15	5.40	
8103	20.2	0.20						3.99	0.16	4.27	
8104	26.3	0.12						4.03	0.17	6.48	
8105	19.4	0.00						3.55	0.14	8.44	
8106	19.0	0.01						3.90	0.16	6.98	
8107	28.0	0.09						3.80	0.09	8.6	0.3
8108	31.1	0.07						4.35	0.13	5.98	
8109	38.0	0.01						3.90	0.12	7.37	
8110	35.3	0.06						4.61	0.14	2.48	
8201	9.5	9.30						2.01	0.10	3.10	
8202	13.4	0.56	17.53	1.58	1.62	38.40	2.54	0.010	2.21	0.11	3.98
8203	13.3	0.21	15.62	1.64	1.67	36.10	2.50	0.008	1.94	0.11	6.17
8204	14.9	0.14	15.07	1.65	1.68	36.10	2.53	0.007	2.55	0.11	6.27
8205	11.2	0.56	14.87	1.67	1.71	24.69	2.51	0.007	2.41	0.10	3.17
8206	5.0	0.00	11.29	1.36	1.88	26.40	2.51	0.003	2.01	0.10	3.91
8207	13.6	0.14	12.11	1.86	1.89	25.48	2.52	0.004	2.20	0.10	6.37
8208	15.3	0.08	12.95	1.89	1.90	25.68	2.42	0.004	2.29	0.11	6.21
8209	1.6	0.16	10.05	1.94	1.95	22.76	2.52	0.002	NA	NA	NA
8210	8.9	0.27	10.37	1.93	1.97	24.15	2.36	0.007	1.91	0.10	4.99
8211	9.5	0.09	13.63	1.82	1.84	22.02	2.54	0.002	1.63	0.11	5.36
8303	1.6	0.33						1.63	0.10	2.40	
8304	7.0	1.18						1.59	0.10	3.64	
8305	8.6	0.65	17.15	1.35	1.37	46.85	2.34	0.006	1.69	0.10	3.53
8306	11.7	0.09	13.95	1.75	1.81	30.96	2.50	0.011	2.01	0.10	5.60
8307	10.4	0.01	13.64	1.78	1.82	29.70	2.50	0.007	2.35	0.11	5.44
8308	25.2	0.01	16.67	1.69	1.76	31.86	2.56	0.013	2.69	0.12	7.12
8309	22.0	0.00	13.10	1.80	1.85	30.18	2.49	0.009	2.78	0.11	7.07
8310	14.4	0.00	13.73	1.75	1.81	27.97	2.47	0.011	2.05	0.10	5.92
8311	0.4	0.00						2.06	0.10	3.81	
8312	9.2	0.03						2.06	0.10	5.74	

Table C4. Particle size distribution analysis for soil (<2mm) by methods of Ciolkosz et al. (1988), by R.R. Dobos.

PENN STATE SOIL CHARACTERIZATION LAB: PARTICLE SIZE DISTRIBUTION ANALYSIS													
IDENT	VCS	CS	MS	FS	VFS-1	VFS-2	TOTAL S	TOTAL C	TOTAL SI	50-5	5-2	20-2	LAB FIELD
IL0101	0.5	0.9	2.6	4.1	2.1	1.9	12.2	23.2	64.6	57.4	7.2	38.0	SIL SIL
IL0102	0.0	0.3	1.2	1.3	0.4	0.5	3.8	28.8	67.4	58.3	9.1	43.3	SICL SIL
IL0103	0.0	0.3	0.7	0.9	0.2	0.3	2.5	31.2	66.4	58.5	7.9	44.4	SICL SIL
IL0104	0.0	0.2	0.5	0.7	0.3	0.4	2.1	35.7	62.2	53.4	8.8	43.3	SICL SIL
IL0105	0.1	0.1	0.5	0.4	0.2	0.3	1.6	34.6	63.7	57.7	6.0	39.9	SICL SIL
IL0106	0.1	0.2	1.0	1.0	0.2	0.4	3.0	30.0	67.0	62.9	4.1	37.4	SICL SIL
IL0107	0.1	0.3	1.2	1.0	0.2	0.3	3.1	33.7	63.3	61.7	1.5	33.7	SICL SIL
IL0108	1.5	3.7	11.5	10.8	2.2	1.6	31.2	21.2	47.6	39.6	8.0	30.7	L SIL
IL0109	1.5	6.1	22.9	26.6	4.9	2.5	64.6	14.5	20.9	16.9	4.0	11.7	L SIL
IDENT	VCS	CS	MS	FS	VFS-1	VFS-2	TOTAL S	TOTAL C	TOTAL SI	50-5	5-2	20-2	LAB FIELD
NV0102	7.8	17.1	27.6	20.9	5.3	3.4	82.1	2.3	15.6	14.7	0.9	6.8	LS LS
NV0103	3.9	17.4	27.1	20.5	5.1	4.4	78.4	3.7	18.0	16.0	2.0	8.5	LS LS
NV0104	9.3	15.9	24.6	20.9	5.5	3.6	79.9	4.1	16.0	12.7	3.3	7.7	LCOS SL
NV0105	10.0	22.3	28.0	18.6	3.8	2.0	84.7	3.6	11.7	9.7	2.0	6.1	LCOS SL
NV0106	7.1	14.0	24.7	23.7	5.8	4.4	79.7	4.7	15.6	11.9	3.6	8.1	LS LS
NV0107	11.3	17.0	23.5	21.5	5.3	4.0	82.6	3.2	14.2	12.6	1.6	5.6	LCOS LS
NV0108	4.6	11.8	21.3	27.0	9.3	6.8	80.8	2.0	17.3	15.5	1.7	5.9	LS LS
NV0109	5.3	11.2	20.4	26.6	7.9	7.6	79.0	0.5	20.5	18.5	2.0	7.7	LS LS
NV0110	7.3	15.2	27.6	27.2	6.7	3.5	87.5	0.8	11.7	10.2	1.6	3.5	S LS
NV0111	3.4	9.1	18.3	27.8	10.4	7.5	76.5	1.7	21.8	18.9	2.9	8.9	LS LS
NV0112	4.0	9.1	17.9	27.5	10.1	8.0	76.5	2.4	21.1	20.3	0.8	7.6	LS LS
IDENT	VCS	CS	MS	FS	VFS-1	VFS-2	TOTAL S	TOTAL C	TOTAL SI	50-5	5-2	20-2	LAB FIELD
089902	2.3	2.1	5.1	19.2	7.0	6.6	42.3	10.9	46.8	40.3	6.5	23.5	L SIL
089903	2.2	2.0	5.3	21.9	7.8	6.6	45.9	11.0	43.1	36.6	6.5	21.2	L SIL
089904	2.6	2.6	6.0	22.2	7.0	6.7	47.0	11.6	41.4	34.9	6.5	20.5	L SIL
089905	6.1	4.7	6.0	12.0	4.1	5.3	38.3	14.4	47.3	41.3	6.0	24.4	L SIL
089906	6.2	4.7	3.4	3.5	2.1	4.3	24.1	22.4	53.5	44.0	9.5	33.7	SIL CL
089907	3.8	3.8	2.9	3.4	2.4	3.3	19.4	25.7	54.9	43.4	11.5	36.3	SIL CL
089908	6.2	4.7	3.1	3.1	2.0	3.8	22.9	21.5	55.6	45.5	10.0	35.3	SIL CL
089909	6.5	5.0	3.2	2.6	1.7	3.3	22.3	21.2	56.4	45.2	11.2	37.7	SIL CL
089910	7.0	5.6	4.3	3.4	2.2	2.9	25.3	18.7	55.9	45.1	10.8	37.2	SIL CL
089911	6.9	6.8	5.1	4.3	2.2	3.8	29.1	17.4	53.5	44.0	9.5	33.4	SIL CL
089912	11.4	9.0	5.9	4.1	2.4	3.1	36.0	15.2	48.8	41.2	7.6	28.9	L CL

PENN STATE SOIL CHARACTERIZATION LAB: PARTICLE SIZE DISTRIBUTION ANALYSIS

IDENT	VCS	CS	MS	FS	VFS-1	VFS-2	TOTAL S	TOTAL C	TOTAL SI	50-5	5-2	20-2	LAB	FIELD
TN0102	1.8	1.8	15.6	17.7	1.8	2.0	40.8	19.6	39.6	29.4	10.2	30.2	L	SIL
TN0103	0.9	1.1	13.7	19.3	2.2	1.7	39.0	20.9	40.1	30.3	9.8	28.7	L	SIL
TN0104	0.3	0.9	13.9	16.1	1.7	1.8	34.8	27.7	37.6	27.6	10.0	28.1	CL	SICL
TN0105	0.2	0.3	4.7	6.6	0.8	0.8	13.4	67.2	19.4	12.4	7.0	15.6	C	C
TN0106	0.2	0.3	3.0	4.0	0.8	0.8	9.2	76.0	14.8	7.6	7.3	12.9	C	C
TN0107	0.0	0.2	2.7	3.4	0.4	0.5	7.3	77.3	15.4	9.0	6.4	12.1	C	C
TN0108	0.1	0.1	1.3	1.7	0.3	0.5	4.0	76.8	19.2	12.4	6.8	15.5	C	C
TN0109	0.1	0.1	1.5	2.8	0.8	0.7	6.1	73.5	20.5	12.0	8.5	16.5	C	C
TN0110	0.1	0.2	1.3	2.9	1.1	1.3	6.9	66.7	26.5	18.6	7.8	22.0	C	SICL
TN0111	1.1	7.0	32.7	12.1	0.4	0.2	53.5	43.1	3.4	2.2	1.2	2.4	SC	SL

IDENT	VCS	CS	MS	FS	VFS-1	VFS-2	TOTAL S	TOTAL C	TOTAL SI	50-5	5-2	20-2	LAB	FIELD
061002 0	3.9	3.6	6.8	12.5	5.3	3.8	36.0	19.5	44.5	33.4	11.1	31.1	L	SIL
061003 0	5.5	5.3	7.7	12.4	4.6	4.5	40.1	8.2	51.6	28.2	23.4	39.4	SIL	SIL
061004 0	5.7	6.9	8.4	13.6	4.9	3.7	43.1	19.5	37.4	26.5	11.0	25.6	L	SIL
061005 0	9.4	8.4	7.9	10.6	4.1	3.7	44.0	18.1	37.9	23.8	14.1	28.2	L	SIL
061006 0	10.6	11.2	7.1	5.7	1.9	1.9	38.5	32.5	29.0	14.5	14.4	23.3	CL	CL
061007 0	10.6	10.2	6.9	6.6	2.7	2.3	39.5	35.3	25.2	14.4	10.8	19.3	CL	SICL
061008 0	11.0	12.5	8.3	7.0	2.2	2.3	43.3	35.0	21.7	12.5	9.2	15.3	CL	CL
061009 0	13.8	13.6	9.8	8.1	2.3	2.4	50.0	33.1	16.9	11.0	6.0	12.0	SCL	SIC

PENN STATE SOIL CHARACTERIZATION LAB - PARTICLE SIZE DISTRIBUTION ANALYSIS

IDENT	VCS	CS	MS	FS	VFS-1	VFS-2	TOTAL S	TOTAL C	TOTAL SI	50-5	5-2	20-2	LAB	FIELD
061102 0	4.6	9.5	11.5	12.2	3.7	3.9	45.3	19.2	35.5	27.1	8.5	24.1	L	SIL
061103 0	5.9	10.0	11.0	12.6	4.2	3.2	46.9	14.1	39.0	27.4	11.6	27.3	L	SIL
061104 0	7.7	10.7	11.7	12.2	3.9	3.7	50.0	6.9	43.1	24.1	19.0	21.0	SL	SIL
061105 0	6.0	9.5	11.3	14.6	3.5	3.5	48.4	20.6	31.0	24.9	6.1	16.1	L	SIL
061106 0	5.4	12.1	13.3	14.9	5.6	4.2	55.3	17.8	26.8	21.2	5.7	14.6	SL	SIL
061107 0	5.9	10.8	15.2	19.5	6.4	5.9	63.6	16.2	20.2	17.0	3.2	9.3	SL	L
061108 0	16.8	17.2	14.2	13.0	4.2	3.1	69.1	11.3	19.6	15.9	3.6	10.1	COSL	L
061109 0	5.7	12.8	17.2	19.7	6.0	5.6	67.0	10.0	23.0	19.4	3.6	10.4	SL	L
061110 0	2.2	10.5	16.4	22.4	7.3	5.2	63.9	9.7	26.4	21.6	4.8	13.3	FSL	L
061111 0	2.6	10.7	15.7	18.4	6.4	6.4	60.2	8.1	31.8	26.5	5.2	17.3	FSL	L

PENN STATE SOIL CHARACTERIZATION LAB - PARTICLE SIZE DISTRIBUTION ANALYSIS

IDENT	VCS	CS	MS	FS	VFS-1	VFS-2	TOTAL S	TOTAL C	TOTAL SI	50-5	5-2	20-2	LAB	FIELD
NC102	0	23.2	21.0	14.4	10.8	3.3	75.1	7.7	17.2	13.6	3.6	7.7	COSL	LCOS
NC103	0	23.0	23.7	14.2	9.0	2.4	74.7	9.2	16.1	12.5	3.6	7.6	COSL	COSL
NC104	0	15.8	23.3	15.4	11.2	2.9	70.7	8.8	20.5	15.7	4.8	10.0	COSL	COSL
NC105	0	8.2	8.6	6.2	5.8	2.1	33.6	45.7	20.7	17.5	3.2	10.9	C	CL
NC106	0	9.1	9.9	7.4	7.3	3.3	39.6	37.6	22.8	15.6	7.3	17.0	CL	CL
NC107	0	9.6	14.1	11.0	10.6	4.7	54.0	28.9	17.2	10.5	6.6	19.5	SCL	CL
NC108	0	11.3	14.6	11.7	10.1	4.2	55.6	19.6	24.8	20.0	4.8	17.2	COSL	CL
NC109	0	11.9	15.6	12.3	11.4	4.3	63.1	8.0	31.9	21.0	10.8	24.8	COSL	L
NC110	0	13.0	15.4	13.1	11.3	4.3	60.8	8.4	30.8	28.4	2.4	21.2	COSL	COSL
NC111	0	17.7	16.7	14.3	12.0	4.2	70.0	3.2	26.8	21.6	5.2	16.0	COSL	SL

PENN STATE SOIL CHARACTERIZATION LAB - PARTICLE SIZE DISTRIBUTION ANALYSIS

IDENT	VCS	CS	MS	FS	VFS-1	VFS-2	TOTAL S	TOTAL C	TOTAL SI	50-5	5-2	20-2	LAB	FIELD
148401	0	0.1	0.4	2.8	13.9	8.0	34.7	18.5	46.8	39.7	7.1	21.0	L	SIL
148402	0	0.1	0.2	1.9	11.5	7.9	30.5	17.1	52.4	40.2	12.2	30.1	SIL	SIL
148403	0	0.1	0.1	0.9	10.8	7.6	29.6	20.9	49.4	43.4	6.0	25.8	L	SIL
148404	0	0.1	1.2	10.3	7.2	6.4	27.9	21.8	50.3	43.0	7.3	23.0	SIL	SIL
148405	0	0.3	1.3	3.7	12.3	6.2	30.2	22.2	47.6	41.2	6.5	27.0	L	SIL
148406	0	0.1	0.3	4.6	15.6	8.0	36.1	18.4	45.5	39.9	5.6	21.6	L	SIL
148407	0	0.1	0.7	8.0	20.3	8.1	46.2	15.5	38.2	32.7	5.6	19.5	L	SIL
148408	0	4.7	9.0	16.2	13.0	4.5	51.9	16.8	31.3	25.7	5.6	17.2	L	LCOS
148409	0	4.8	8.6	12.8	11.0	4.2	46.6	16.9	36.5	26.4	10.1	22.1	L	LCOS

Table C5. Bulk density, porosity, percentage of coarse fragments and water held for various tensions, by R.R. Dobos using methods of Ciolkosz et al., (1988).

PENN STATE SOIL CHARACTERIZATION LAB - BULK DENSITY AND WATER DETERMINATIONS

IDENT	BULK DENSITY		COARSE FRAGMENTS		COEFF OF LIN EXTENS	TOTAL POROS ESTIM (VOL%)	WATER RETENTION				TOTAL SOIL					
	TOTAL	<2 MM	VOL (VOL%)	WGHT (WT%)			<2 MM MATERIAL		1/3 AT 15 AT		TOTAL SOIL					
							1/3 AT	15 AT	AVAIL	AVAIL	1/3 AT	15 AT	AVAIL	AVAIL		
----- (G/CM3) ----- (WT%) ----- (VOL%) ----- (VOL%) ----- (WT%) ----- (VOL%) ----- (VOL%) ----- (CM/CM)																
148401A	1.26	1.25	2.54	0.5	1.1	0.014	52.6	27.7	10.8	16.9	21.1	27.4	10.7	16.7	21.0	0.21
148401C	1.36	1.36	0.0	0.0	0.0	0.015	48.9	23.0	10.8	12.2	16.6	23.0	10.8	12.2	16.6	0.17
148401D	1.35	1.35	0.0	0.0	0.0	0.015	49.2	22.4	10.8	11.7	15.7	22.4	10.8	11.7	15.7	0.16
148402A	1.33	1.33	0.0	0.0	0.0	0.016	49.7	20.6	10.1	10.5	13.9	20.6	10.1	10.5	13.9	0.14
148402B	1.53	1.53	2.20	0.2	0.2	0.013	42.2	17.6	10.1	7.5	11.4	17.6	10.1	7.5	11.4	0.11
148402C	1.45	1.45	0.0	0.0	0.0	0.012	45.3	17.7	10.1	7.5	10.9	17.7	10.1	7.5	10.9	0.11
148403A	1.42	1.42	0.0	0.0	0.0	0.006	46.4	20.3	10.2	10.1	14.4	20.3	10.2	10.1	14.4	0.14
148403C	1.51	1.51	0.0	0.0	0.0	0.010	43.2	19.4	10.2	9.2	13.8	19.4	10.2	9.2	13.8	0.14
148403D	1.45	1.45	0.0	0.0	0.0	0.011	45.1	20.7	10.1	10.6	15.4	20.7	10.1	10.6	15.4	0.15
148404A	1.51	1.51	0.0	0.0	0.0	0.007	42.9	19.3	10.0	9.3	14.0	19.3	10.0	9.3	14.0	0.14
148404C	1.48	1.48	0.0	0.0	0.0	0.010	44.2	20.0	10.0	10.0	14.8	20.0	10.0	10.0	14.8	0.15
148404D	1.47	1.47	0.0	0.0	0.0	0.011	44.6	19.9	10.0	9.9	14.6	19.9	10.0	9.9	14.6	0.15
148405A	1.48	1.48	0.0	0.0	0.0	0.009	44.1	19.2	10.3	8.9	13.2	19.2	10.3	8.9	13.2	0.13
148405B	1.41	1.41	0.0	0.0	0.0	0.008	46.7	19.4	10.3	9.1	12.9	19.4	10.3	9.1	12.9	0.13
148505D	1.46	1.46	0.0	0.0	0.0	0.002	45.0	19.3	10.3	9.0	13.1	19.3	10.3	9.0	13.1	0.13
148406A	1.38	1.38	0.0	0.0	0.0	0.008	47.8	19.9	9.3	10.6	14.7	19.9	9.3	10.6	14.7	0.15
148406B	1.35	1.35	2.22	0.1	0.1	0.003	49.1	20.5	9.3	11.2	15.1	20.5	9.3	11.2	15.0	0.15
148406C	1.41	1.41	0.0	0.0	0.0	0.007	46.6	19.4	9.3	10.1	14.3	19.4	9.3	10.1	14.3	0.14
148407A	1.42	1.42	0.0	0.0	0.0	0.012	46.3	18.1	7.4	10.7	15.3	18.1	7.4	10.7	15.3	0.15
148407B	1.36	1.36	0.0	0.0	0.0	0.021	48.7	15.7	7.4	8.3	11.2	15.7	7.4	8.3	11.2	0.11
148407C	1.54	1.48	2.55	5.3	8.8	0.004	42.0	14.0	7.4	6.6	9.8	12.8	6.7	6.1	9.3	0.10

061003B	1.46	1.19	2.30	23.8	37.6	0.002	45.0	34.3	9.4	24.9	29.7	21.4	5.9	15.5	22.6	0.36
061003C	1.51	1.07	2.24	37.8	56.0	0.006	42.9	36.5	9.4	27.0	28.9	16.0	4.2	11.9	16.0	0.41
061003D	1.58	1.22	2.32	32.2	47.4	0.008	40.5	29.0	9.4	19.5	23.8	15.2	5.0	10.3	16.2	0.31
061004A	1.54	1.25	2.29	27.9	41.5	0.035	41.9	29.2	8.6	20.6	25.7	17.1	5.0	12.0	18.5	0.32
061004B	1.50	1.27	2.36	20.8	32.8	0.011	43.5	27.5	8.6	18.9	24.0	18.5	5.8	12.7	19.0	0.28
061004D	1.44	1.18	2.40	21.7	36.0	0.012	45.5	25.2	8.6	16.6	19.6	16.1	5.5	10.6	15.3	0.24
061005A	1.67	1.36	2.32	32.1	44.7	0.008	36.9	25.9	10.0	15.9	21.6	14.3	5.5	8.8	14.7	0.27
061005C	1.66	1.16	2.27	45.3	61.8	0.018	37.3	43.0	10.0	33.0	38.3	16.4	3.8	12.6	21.0	0.55
061005D	1.81	1.31	2.24	53.9	66.7	0.035	31.7	37.4	10.0	27.5	35.9	12.5	3.3	9.2	16.6	0.50
061006A	1.82	1.12	2.36	56.5	73.1	0.022	31.3	54.3	14.7	39.6	44.5	14.6	4.0	10.6	19.4	0.72
061006C	1.84	1.11	2.31	61.1	76.5	0.013	30.4	53.3	14.7	38.6	43.0	12.5	3.5	9.1	16.7	0.71
061006D	1.68	1.06	2.22	53.2	70.5	0.013	36.7	49.0	14.7	34.2	36.2	14.5	4.3	10.1	17.0	0.57
061007A	1.86	1.29	2.27	58.1	70.6	0.005	29.8	44.6	14.8	29.8	38.6	13.0	4.3	8.7	16.2	0.55
061007C	1.81	1.19	2.28	56.5	71.3	0.029	31.9	48.1	14.8	33.3	39.7	13.8	4.3	9.6	17.3	0.60
061007B	1.87	1.17	2.24	65.7	78.6	0.004	29.5	51.6	14.8	36.8	43.0	11.1	3.2	7.9	14.8	0.69
061008A	1.87	1.20	2.28	62.3	75.8	0.021	29.3	51.1	14.1	37.1	44.7	12.4	3.4	9.0	16.8	0.69
061008B	1.80	1.17	2.18	62.5	75.6	0.010	32.2	56.5	14.1	42.5	49.6	13.8	3.4	10.4	18.6	0.76
061008C	1.73	1.09	2.33	51.6	69.5	0.028	34.6	49.5	14.1	35.4	38.7	15.1	4.3	10.8	18.7	0.61

IDENT	BULK DENSITY		COARSE FRAGMENTS			COEFF OF LIN EXTENS	TOTAL POROS ESTIM (VOL%)	<2 MM MATERIAL			WATER RETENTION			TOTAL SOIL		
	TOTAL	<2 MM	DENS	VOL (VOL%)	WGHT (WT%)			1/3 AT	15 AT	AVAIL (VOL%)	1/3 AT	15 AT	AVAIL (VOL%)	1/3 AT	15 AT	AVAIL (VOL%)
NC103A	1.59	1.51	2.62	7.7	12.6	0.007	39.9	11.0	3.5	7.5	11.3	9.6	3.1	6.6	10.4	0.12
NC103B	1.63	1.54	2.57	8.4	13.3	0.001	38.6	10.9	3.5	7.4	11.5	9.5	3.0	6.4	10.5	0.12
NC103D	1.71	1.62	2.62	8.9	13.6	0.001	35.4	9.8	3.5	6.3	10.2	8.4	3.0	5.4	9.3	0.11
NC104B	1.64	1.57	2.57	6.9	10.8	0.011	38.1	12.9	3.9	9.0	14.1	11.5	3.5	8.0	13.1	0.15
NC104C	1.65	1.58	2.60	6.8	10.7	0.008	37.6	11.7	3.9	7.8	12.4	10.5	3.5	7.0	11.5	0.13
NC104D	1.65	1.59	2.56	7.0	10.8	0.011	37.6	13.3	3.9	9.4	14.9	11.9	3.5	8.4	13.8	0.15
NC105A	1.39	1.34	2.60	3.7	7.0	0.021	47.6	29.6	20.5	9.1	12.2	27.6	19.1	8.5	11.7	0.13
NC105C	1.41	1.37	2.52	3.3	5.9	0.024	46.9	27.1	20.5	6.6	9.0	25.5	19.3	6.2	8.7	0.09
NC105D	1.30	1.25	2.51	3.4	6.6	0.041	51.0	29.7	20.5	9.1	11.5	27.7	19.2	8.5	11.1	0.12
NC106A	1.39	1.31	2.70	5.5	10.7	0.019	47.5	30.0	18.8	11.2	14.7	26.8	16.8	10.0	13.9	0.16
NC106B	1.33	1.29	2.54	2.8	5.4	0.021	49.9	29.4	18.8	10.6	13.6	27.8	17.8	10.0	13.3	0.14
NC106C	1.37	1.28	2.56	6.8	12.6	0.029	48.3	26.7	18.8	7.9	10.2	23.4	16.4	6.9	9.5	0.11
NC107A	1.39	1.34	2.82	3.7	7.5	0.017	47.4	26.1	14.7	11.4	15.3	24.1	13.6	10.6	14.7	0.16
NC107C	1.37	1.32	2.58	3.9	7.3	0.023	48.4	30.2	14.7	15.5	20.4	28.0	13.6	14.4	19.7	0.21
NC107D	1.45	1.38	2.62	5.4	9.8	0.014	45.4	26.5	14.7	11.9	16.4	23.9	13.2	10.7	15.5	0.17
NC108A	1.44	1.42	2.57	2.1	3.8	0.011	45.5	25.0	11.9	13.0	18.5	24.0	11.5	12.5	18.1	0.19
NC108B	1.44	1.38	2.79	4.3	8.4	0.001	45.6	24.1	11.9	12.2	16.8	22.1	10.9	11.2	16.1	0.18
NC108C	1.31	1.28	2.44	2.3	4.3	0.057	50.5	27.6	11.9	15.7	20.1	26.4	11.4	15.0	19.7	0.21
NC109B	1.49	1.39	2.72	7.2	13.1	0.011	43.9	24.3	9.2	15.1	20.9	21.1	8.0	13.1	19.4	0.22
NC109C	1.39	1.33	2.63	4.1	7.8	0.016	47.6	26.8	9.2	17.5	23.4	24.7	8.5	16.2	22.4	0.24
NC109D	1.44	1.37	2.77	4.8	9.3	0.018	45.8	24.6	9.2	15.4	21.0	22.3	8.4	13.9	20.0	0.22
NC110A	1.32	1.24	2.61	5.5	10.9	0.015	50.3	28.3	8.2	20.1	24.9	25.2	7.3	17.9	23.6	0.26
NC110C	1.31	1.25	2.53	4.9	9.5	0.016	50.4	27.1	8.2	18.9	23.6	24.6	7.5	17.1	22.5	0.25
NC110D	1.29	1.21	2.63	5.6	11.5	0.014	51.5	24.1	8.2	15.9	19.1	21.3	7.3	14.0	18.1	0.20
NC111A	1.33	1.20	2.58	9.3	18.1	0.002	49.9	24.9	5.6	19.3	23.1	20.4	4.6	15.8	21.0	0.26
NC111B	1.38	1.28	2.82	6.4	13.1	0.002	48.1	24.2	5.6	18.6	23.8	21.0	4.8	16.2	22.3	0.26
NC111C	1.27	1.16	2.65	7.6	15.8	0.013	52.1	23.2	5.6	17.6	20.4	19.5	4.7	14.8	18.8	0.22

PENN STATE SOIL CHARACTERIZATION LAB - BULK DENSITY AND WATER DEYE RMINATIONS

IDENT	BULK DENSITY		COARSE FRAGMENTS			COEFF OF LIN EXTENS	TOTAL POROS ESTIM (VOL%)	<2 MM MATERIAL			WATER RETENTION			TOTAL SOIL			
	TOTAL SOIL	<2 MM (G/CM3)	DENS	VOL (VOL%)	WGHT (WT%)			1/3 AT (WT%)	15 AT (WT%)	AVAIL (VOL%)	1/3 AT (WT%)	15 AT (WT%)	AVAIL (VOL%)	1/3 AT (WT%)	15 AT (WT%)	AVAIL (VOL%)	
NY0108B	1.90	1.77	2.62	15.1	20.9	0.004	28.4	4.3	1.2	3.1	5.4	5.4	3.4	1.0	2.4	4.6	0.06
NY0108C	1.92	1.84	2.61	9.7	13.2	0.007	27.7	4.4	1.2	3.1	5.8	5.8	3.8	1.1	2.7	5.2	0.06
NY0108E	1.90	1.82	2.57	11.7	15.8	0.003	28.1	5.6	1.2	4.4	8.0	8.0	4.7	1.0	3.7	7.0	0.08
NY0109A	1.96	1.86	2.69	12.5	17.2	0.003	25.9	6.9	1.0	6.0	11.1	11.1	5.7	0.8	4.9	9.7	0.12
NY0109D	1.73	1.66	2.64	7.7	11.7	0.012	34.6	6.0	1.0	5.0	8.3	8.3	5.3	0.8	4.4	7.7	0.09
NY0109E	1.73	1.68	2.63	5.2	7.9	0.003	34.7	5.1	1.0	4.1	6.9	6.9	4.7	0.9	3.8	6.6	0.07
NY0110B	1.83	1.75	2.66	8.1	11.8	0.009	31.1	5.2	0.8	4.5	7.8	7.8	4.6	0.7	4.0	7.2	0.08
NY0110D	1.94	1.93	2.04	7.9	8.3	0.005	26.9	6.9	0.8	6.1	11.8	11.8	6.3	0.7	5.6	10.9	0.12
NY0110E	1.96	1.89	2.64	8.6	11.6	0.002	26.2	6.5	0.8	5.7	10.8	10.8	5.7	0.7	5.1	9.9	0.11
NY0111C	1.75	1.68	2.59	7.0	10.3	0.005	34.1	6.9	1.1	5.8	9.8	9.8	6.2	1.0	5.2	9.1	0.10
NY0111E	1.73	1.64	2.55	9.1	13.5	0.040	34.9	7.1	1.1	6.0	9.9	9.9	6.2	1.0	5.2	9.0	0.10
NY0112B	2.17	1.75	2.72	42.9	53.9	0.006	18.2	6.0	1.1	4.8	8.5	8.5	2.8	0.5	2.2	4.8	0.11
NY0112C	1.83	1.76	2.55	8.4	11.8	0.005	31.1	6.3	1.1	5.1	9.1	9.1	5.5	1.0	4.5	8.3	0.09
NY0112E	1.79	1.72	2.55	8.8	12.6	0.001	32.4	6.2	1.1	5.0	8.6	8.6	5.4	1.0	4.4	7.9	0.09
061104B	1.43	1.39	2.47	3.5	6.1	0.012	45.9	20.7	9.6	11.0	15.4	15.4	19.4	9.0	10.4	14.9	0.16
061104C	1.47	1.25	2.49	17.8	30.1	0.008	44.4	23.4	9.6	13.8	17.3	17.3	16.4	6.7	9.7	14.2	0.20
061105B	1.44	1.41	2.36	3.3	5.4	0.017	45.6	22.3	10.4	11.9	16.7	16.7	21.1	9.9	11.2	16.2	0.17
061105C	1.44	1.40	2.36	3.9	6.4	0.006	45.6	19.3	10.4	8.9	12.5	12.5	18.1	9.8	8.3	12.0	0.13
061105D	1.37	1.34	2.38	3.0	5.1	0.016	48.2	21.0	10.4	10.6	14.2	14.2	19.9	9.9	10.0	13.8	0.15
061106A	1.42	1.40	2.29	2.7	4.3	0.014	46.4	22.5	10.3	12.2	17.1	17.1	21.5	9.8	11.7	16.6	0.17
061106B	1.36	1.34	2.10	2.5	3.9	0.001	48.8	22.2	10.3	11.9	15.9	15.9	21.3	9.9	11.4	15.5	0.16
061106C	1.42	1.40	2.09	3.5	5.2	0.002	46.3	22.2	10.3	11.9	16.6	16.6	21.0	9.7	11.3	16.0	0.17
061107B	1.35	1.20	2.15	15.2	24.3	0.018	49.1	30.2	10.3	19.9	24.0	24.0	22.9	7.8	15.1	20.3	0.27
061107C	1.36	1.32	2.31	4.2	7.2	0.005	48.6	20.8	10.3	10.6	13.9	13.9	19.4	9.5	9.8	13.3	0.14
061107D	1.51	1.23	2.21	28.4	41.6	0.002	43.2	32.5	10.3	22.2	27.3	27.3	19.0	6.0	13.0	19.5	0.33
061108A	1.26	1.22	2.17	4.8	8.2	0.002	52.3	21.9	7.8	14.1	17.2	17.2	20.1	7.2	12.9	16.3	0.18
061108C	1.22	1.13	2.27	7.4	13.9	0.000	54.1	26.6	7.8	18.8	21.3	21.3	22.9	6.8	16.2	19.7	0.23
061108D	1.30	1.21	2.03	10.3	16.1	0.012	51.1	27.1	7.8	19.3	23.4	23.4	22.8	6.6	16.2	21.0	0.25
061109A	1.30	1.19	2.09	12.6	20.2	0.012	50.8	29.9	8.1	21.8	25.9	25.9	23.8	6.5	17.4	22.6	0.28
061109C	1.35	1.34	2.34	1.2	2.1	0.012	49.0	23.9	8.1	15.8	21.1	21.1	23.4	8.0	15.4	20.8	0.21
061109D	1.22	1.22	2.11	0.1	0.1	0.037	54.0	23.7	8.1	15.5	18.9	18.9	23.6	8.1	15.5	18.9	0.19
061110A	1.39	0.81	2.12	44.6	68.0	0.005	47.5	69.5	7.3	62.2	50.1	50.1	22.3	2.4	19.9	27.7	0.87
061110B	1.27	1.21	2.10	7.0	11.5	0.007	52.2	25.3	7.3	17.9	21.6	21.6	22.4	6.5	15.9	20.1	0.23
061110D	1.37	1.36	2.29	1.9	3.2	0.001	48.2	21.2	7.3	13.8	18.7	18.7	20.5	7.1	13.4	18.4	0.19
061111C	1.14	1.13	2.34	0.9	1.8	0.074	57.0	31.8	6.6	25.2	28.4	28.4	31.3	6.5	24.8	28.2	0.29
061111D	1.25	1.24	2.45	0.1	0.3	0.000	53.0	24.0	6.6	17.4	21.6	21.6	23.9	6.6	17.3	21.6	0.22

PENN STATE SOIL CHARACTERIZATION LAB - 1 - .ITY AND WATER DETE RMINATIONS

IDENT	BULK DENSITY		COARSE FRAGMENTS			COEFF OF LIN EXTENS	IAL PROS ESTIM (VOL%)	<2 MM MATERIAL			WATER RETENTION			TOTAL SOIL			
	TOTAL SOIL	<2 MM (G/CM3)	DENS	VOL (VOL%)	WGHT (WT%)			1/3 AT (WT%)	15 AT (WT%)	AVAIL (VOL%)	1/3 AT (WT%)	15 AT (WT%)	AVAIL (VOL%)	1/3 AT (WT%)	15 AT (WT%)	AVAIL (VOL%)	CM/CM)
IL0101A	1.08	1.08	0.0	0.0	0.0	0.030	59.2	29.9	13.3	16.7	18.0	29.9	13.3	16.7	18.0	0.18	
IL0101C	1.13	1.13	0.0	0.0	0.0	0.024	57.4	28.4	13.3	15.2	17.2	28.4	13.3	15.2	17.2	0.17	
IL0101E	1.06	1.06	0.0	0.0	0.0	0.025	59.9	30.4	13.3	17.1	18.2	30.4	13.3	17.1	18.2	0.18	
IL0102B	1.29	1.29	0.0	0.0	0.0	0.039	51.4	28.8	13.0	15.8	20.4	28.8	13.0	15.8	20.4	0.20	
IL0102D	1.23	1.23	0.0	0.0	0.0	0.031	53.5	27.1	13.0	14.1	17.3	27.1	13.0	14.1	17.3	0.17	
IL0102E	1.22	1.22	0.0	0.0	0.0	0.027	53.9	27.1	13.0	14.1	17.3	27.1	13.0	14.1	17.3	0.17	
IL0103C	1.21	1.21	0.0	0.0	0.0	0.031	54.2	26.2	14.0	12.2	14.8	26.2	14.0	12.2	14.8	0.15	
IL0103D	1.22	1.22	0.0	0.0	0.0	0.044	54.1	27.3	14.0	13.3	16.2	27.3	14.0	13.3	16.2	0.16	
IL0103E	1.15	1.15	0.0	0.0	0.0	0.033	56.6	29.0	14.0	15.0	17.2	29.0	14.0	15.0	17.2	0.17	
IL0104A	1.28	1.28	0.0	0.0	0.0	0.053	51.8	27.4	16.7	10.7	13.7	27.4	16.7	10.7	13.7	0.14	
IL0104B	1.26	1.26	0.0	0.0	0.0	0.051	52.6	29.5	16.7	12.8	16.1	29.5	16.7	12.8	16.1	0.16	
IL0104D	1.29	1.29	0.0	0.0	0.0	0.045	51.4	29.1	16.7	12.4	16.0	29.1	16.7	12.4	16.0	0.16	
IL0105A	1.27	1.27	0.0	0.0	0.0	0.046	51.9	29.9	15.0	14.9	19.0	29.9	15.0	14.9	19.0	0.19	
IL0105C	1.26	1.26	0.0	0.0	0.0	0.040	52.4	29.7	15.0	14.7	18.5	29.7	15.0	14.7	18.5	0.18	
IL0105E	1.28	1.28	0.0	0.0	0.0	0.043	51.5	29.2	15.0	14.2	18.2	29.2	15.0	14.2	18.2	0.18	
IL0106A	1.26	1.26	0.0	0.0	0.0	0.035	52.4	29.0	13.5	15.5	19.5	29.0	13.5	15.5	19.5	0.20	
IL0106C	1.22	1.22	0.0	0.0	0.0	0.048	53.9	29.6	13.5	16.1	19.7	29.6	13.5	16.1	19.7	0.20	
IL0107B	1.22	1.22	0.0	0.0	0.0	0.036	53.8	29.1	15.6	13.4	16.4	29.1	15.6	13.4	16.4	0.16	
IL0107C	1.28	1.28	0.0	0.0	0.0	0.029	51.6	25.9	15.6	10.2	13.1	25.9	15.6	10.2	13.1	0.13	
IL0107E	1.29	1.29	3.10	0.2	0.4	0.034	51.2	26.9	15.6	11.3	14.5	26.8	15.6	11.2	14.5	0.15	
IL0108A	1.44	1.44	3.10	0.2	0.5	0.019	45.6	17.6	9.4	8.2	11.7	17.5	9.4	8.1	11.7	0.12	
IL0108D	1.49	1.46	2.32	3.2	5.0	0.022	44.0	18.6	9.4	9.1	13.3	17.6	8.9	8.7	12.9	0.14	
IL0108E	1.40	1.39	2.09	1.0	1.4	0.023	47.1	23.6	9.4	14.2	19.7	23.2	9.3	13.9	19.5	0.20	
IL0109C	1.45	1.44	2.43	1.0	1.7	0.012	45.3	15.8	6.2	9.6	13.8	15.6	6.1	9.4	13.7	0.14	
IL0109D	1.49	1.44	2.48	4.4	7.3	0.023	43.9	16.3	6.2	10.1	14.5	15.1	5.8	9.3	13.9	0.15	
IL0109E	1.49	1.46	2.41	3.3	5.4	0.012	43.6	14.6	6.2	8.4	12.3	13.8	5.9	7.9	11.9	0.13	

IDENT	BULK DENSITY		COARSE FRAGMENTS			COEFF OF LIN EXTENS	TOTAL POROS ESTIM (VOL%)	---<2 MM MATERIAL---				WATER RETENTION---				TOTAL SOIL---			
	TOTAL SOIL	---(G/CM3)---	DENS	VOL (VOL%)	WGHT (WT%)			1/3 AT	15 AT	AVAIL (VOL%)	1/3 AT	15 AT	AVAIL (WT%)	1/3 AT	15 AT	AVAIL (VOL%)	AVAIL (CM/CM)		
TNO103B	1.57	1.55	2.42	2.1	3.3	0.010	40.8	16.9	8.2	8.8	13.6	16.4	7.9	8.5	13.3	0.14			
TNO103C	1.58	1.52	2.45	6.9	10.7	0.013	40.4	17.6	8.2	9.4	14.2	15.7	7.3	8.4	13.3	0.15			
TNO103D	1.57	1.57	3.67	0.1	0.2	0.009	40.7	16.9	8.2	8.8	13.8	16.9	8.2	8.7	13.7	0.14			
TNO104A	1.56	1.56	0.0	0.0	0.0	0.014	41.0	16.7	10.3	6.5	10.1	16.7	10.3	6.5	10.1	0.10			
TNO104B	1.58	1.58	0.0	0.0	0.0	0.018	40.5	16.6	10.3	6.4	10.1	16.6	10.3	6.4	10.1	0.10			
TNO104C	1.60	1.60	0.0	0.0	0.0	0.014	39.6	17.7	10.3	7.4	11.9	17.7	10.3	7.4	11.9	0.12			
TNO105A	1.33	1.33	0.0	0.0	0.0	0.031	49.9	30.3	24.3	6.0	7.9	30.3	24.3	6.0	7.9	0.08			
TNO105B	1.29	1.29	0.0	0.0	0.0	0.029	51.4	31.1	24.3	6.8	8.7	31.1	24.3	6.8	8.7	0.09			
TNO105C	1.33	1.33	2.83	0.2	0.4	0.038	49.8	27.9	24.3	3.6	4.7	27.8	24.2	3.5	4.7	0.05			
TNO106A	1.27	1.27	0.0	0.0	0.0	0.015	52.1	33.5	28.1	5.5	6.9	33.5	28.1	5.5	6.9	0.07			
TNO106B	1.33	1.33	0.0	0.0	0.0	0.017	50.0	32.4	28.1	4.3	5.7	32.4	28.1	4.3	5.7	0.06			
TNO106D	1.21	1.21	1.74	1.0	1.5	0.010	54.2	36.4	28.1	8.3	10.1	35.9	27.7	8.2	10.0	0.10			
TNO107A	1.16	1.16	0.0	0.0	0.0	0.023	56.1	37.1	28.4	8.7	10.1	37.1	28.4	8.7	10.1	0.10			
TNO107D	1.06	1.06	0.0	0.0	0.0	0.090	59.9	34.5	28.4	6.1	6.5	34.5	28.4	6.1	6.5	0.07			
TNO108A	1.16	1.16	0.0	0.0	0.0	0.036	56.2	38.1	27.7	10.4	12.1	38.1	27.7	10.4	12.1	0.12			
TNO108B	1.17	1.17	0.0	0.0	0.0	0.019	55.9	39.1	27.7	11.4	13.3	39.1	27.7	11.4	13.3	0.13			
TNO108C	1.13	1.13	0.0	0.0	0.0	0.015	57.4	39.4	27.7	11.7	13.2	39.4	27.7	11.7	13.2	0.13			
TNO109B	1.23	1.23	0.0	0.0	0.0	0.044	53.4	35.7	29.0	6.7	8.3	35.7	29.0	6.7	8.3	0.08			
TNO109C	1.20	1.20	0.0	0.0	0.0	0.027	54.6	38.7	29.0	9.7	11.6	38.7	29.0	9.7	11.6	0.12			
TNO109E	1.12	1.12	0.0	0.0	0.0	0.038	57.9	35.5	29.0	6.5	7.3	35.5	29.0	6.5	7.3	0.07			
TNO110B	1.12	1.12	0.0	0.0	0.0	0.041	57.7	38.3	17.2	21.1	23.7	38.3	17.2	21.1	23.7	0.24			
TNO110D	1.04	1.04	0.0	0.0	0.0	0.032	60.7	39.6	17.2	22.4	23.3	39.6	17.2	22.4	23.3	0.23			
TNO111A	1.29	1.16	1.94	16.6	25.0	0.011	51.4	26.2	4.2	21.9	25.4	19.6	3.2	16.5	21.2	0.28			
TNO111C	1.35	0.98	1.70	51.3	64.7	0.009	49.0	37.1	4.2	32.9	32.3	13.1	1.5	11.6	15.7	0.44			
TNO111D	1.46	1.46	0.0	0.0	0.0	0.025	45.0	18.3	4.2	14.1	20.5	18.3	4.2	14.1	20.5	0.21			

PENN STATE SOIL CHARACTERIZATION LAB - BULK DENSITY AND WATER RETENTION

IDENT	BULK DENSITY		COARSE FRAGMENTS			COEFF OF LIN EXTENS	TOTAL POROS ESTIM (VOL%)	<2 MM MATERIAL			WATER RETENTION			TOTAL SOIL			
	TOTAL SOIL	<2 MM	DENS	VOL (VOL%)	WGHT (WT%)			1/3 AT	15 AT (WT%)	AVAIL (VOL%)	1/3 AT	15 AT (WT%)	AVAIL (VOL%)	1/3 AT	15 AT (WT%)	AVAIL (VOL%)	CM/CM
089903A	1.18	1.14	2.65	2.7	6.0	0.006	55.4	20.1	8.8	11.3	12.9	18.9	8.3	10.6	12.5	0.13	
089903C	1.27	1.21	2.61	3.8	7.9	0.005	52.2	19.2	8.8	10.4	12.6	17.7	8.1	9.6	12.1	0.13	
089903E	1.30	1.26	2.65	2.8	5.7	0.001	50.8	17.8	8.8	9.0	11.4	16.8	8.3	8.5	11.1	0.12	
089904B	1.59	1.53	2.59	5.6	9.1	0.004	40.0	15.1	6.3	8.8	13.5	13.7	5.7	8.0	12.8	0.14	
089904C	1.67	1.48	2.56	17.7	27.0	0.002	36.8	16.3	6.3	10.1	14.9	11.9	4.6	7.3	12.3	0.17	
089904D	1.58	1.48	2.55	8.8	14.3	0.004	40.5	15.3	6.3	9.0	13.4	13.1	5.4	7.7	12.2	0.14	
089905A	1.47	1.39	2.59	7.3	12.9	0.009	44.4	16.4	8.8	7.6	10.5	14.3	7.7	6.6	9.8	0.11	
089905C	1.83	1.58	2.60	24.5	34.7	0.009	31.0	19.1	8.8	10.2	16.2	12.4	5.8	6.7	12.2	0.19	
089905E	1.60	1.51	2.59	8.5	13.7	0.008	39.6	15.2	8.8	6.4	9.6	13.1	7.6	5.5	8.8	0.10	
089906B	1.77	1.61	2.61	16.2	23.8	0.013	33.1	17.3	10.1	7.2	11.6	13.2	7.7	5.5	9.7	0.13	
089906C	1.87	1.62	2.56	27.0	36.9	0.001	29.3	17.4	10.1	7.3	11.7	11.0	6.4	4.6	8.6	0.14	
089906D	1.79	1.61	2.58	18.0	26.0	0.013	32.6	17.9	10.1	7.8	12.6	13.3	7.5	5.8	10.3	0.14	
089907A	1.79	1.70	2.62	9.7	14.2	0.017	32.6	16.8	9.5	7.2	12.3	14.4	8.2	6.2	11.1	0.13	
089907B	1.85	1.71	2.54	17.1	23.4	0.015	30.1	17.1	9.5	7.5	12.9	13.1	7.3	5.8	10.7	0.14	
089907D	1.78	1.62	2.58	16.5	24.0	0.016	33.0	18.7	9.5	9.1	14.8	14.2	7.3	6.9	12.3	0.16	
089908B	1.81	1.65	2.61	17.2	24.8	0.011	31.7	16.9	9.0	7.9	13.0	12.7	6.8	5.9	10.7	0.14	
089908D	1.88	1.55	2.62	30.3	42.4	0.012	29.2	18.6	9.0	9.6	14.9	10.7	5.2	5.5	10.4	0.18	
089908E	1.88	1.65	2.59	24.8	34.1	0.013	28.9	17.5	9.0	8.5	14.1	11.6	5.9	5.6	10.6	0.16	
089909A	1.86	1.77	2.60	11.5	16.1	0.015	29.7	16.4	8.9	7.4	13.2	13.8	7.5	6.2	11.6	0.14	
089909B	1.92	1.68	2.61	25.2	34.3	0.008	27.7	15.9	8.9	6.9	11.7	10.4	5.9	4.6	8.7	0.13	
089909C	1.81	1.67	2.58	14.9	21.3	0.024	31.8	19.4	8.9	10.5	17.5	15.3	7.0	8.2	14.9	0.19	
089910C	1.89	1.59	2.62	28.6	39.8	0.002	28.8	19.6	7.9	11.8	18.7	11.8	4.7	7.1	13.4	0.22	
089910D	2.05	1.66	2.62	39.8	51.0	0.002	22.8	20.5	7.9	12.7	21.1	10.1	3.8	6.2	12.7	0.26	
089910E	1.90	1.57	2.61	31.7	43.5	0.008	28.2	19.0	7.9	11.1	17.5	10.7	4.4	6.3	11.9	0.21	
089911A	1.78	1.57	2.55	21.3	30.5	0.016	32.8	17.2	7.4	9.8	15.5	12.0	5.2	6.8	12.2	0.18	
089911D	1.82	1.57	2.52	26.2	36.3	0.001	31.3	19.7	7.4	12.3	19.3	12.5	4.7	7.8	14.2	0.22	
089911E	2.00	1.67	2.43	42.8	52.1	0.005	24.6	18.6	7.4	11.2	18.6	8.9	3.5	5.3	10.7	0.22	
089912A	1.87	1.25	2.63	45.1	63.5	0.004	29.4	25.8	5.7	20.1	25.0	9.4	2.1	7.3	13.7	0.38	
089912C	2.14	1.58	2.52	59.3	69.9	0.011	19.3	24.0	5.7	18.3	29.0	7.2	1.7	5.5	11.8	0.39	
089912D	1.99	1.61	2.64	37.2	49.4	0.001	24.8	21.8	5.7	16.1	25.8	11.0	2.9	8.1	16.2	0.32	

Table C6. Total chemical analysis of soils, by method of Suhr and Gong (1983). See Ciolkosz et al. (1993) for additional analyses of Pennsylvania soils.

THE PENNSYLVANIA STATE UNIVERSITY
INTER-OFFICE CORRESPONDENCE

Date: April 18, 1988

From: N. H. Suhr, 311 Mineral Sciences

To: A. W. Rose, 332 Deike

Subj: Spectrochemical Analysis of Soil Samples

Our Sample No.	88-140	88-141	88-142	88-143	88-144	88-145	88-146	88-147
Your Sample No.	4-8001	8002	8003	8004	8005	8006	8007	8008
SiO ₂	67.2%	80.1%	75.2%	65.2%	55.3%	56.4%	58.6%	54.9%
Al ₂ O ₃	6.87	8.24	11.1	16.5	21.5	20.5	20.4	21.5
TiO ₂	0.85	1.04	0.96	0.82	0.66	0.63	0.57	0.70
Fe ₂ O ₃	3.04	3.53	4.44	5.81	7.05	6.85	6.50	7.22
MgO	0.49	0.63	0.94	1.62	1.91	1.93	1.86	2.09
CaO	0.56	0.15	0.11	0.14	0.16	0.13	0.15	0.13
MnO	0.440	0.115	0.061	0.049	0.065	0.075	0.083	0.102
Na ₂ O	0.24	0.27	0.23	0.13	0.07	0.06	0.05	0.06
K ₂ O	1.58	2.28	2.84	3.89	4.23	4.20	3.64	4.00
P ₂ O ₅	0.23	0.11	0.07	0.08	0.11	0.08	0.09	0.14
L.O.I. (950°C)	18.7	3.37	4.33	6.39	8.22	8.29	8.30	9.03
TOTALS	100.2%	99.8%	100.3%	100.6%	99.3%	99.1%	100.2%	99.9%

Our Sample No.	88-148	88-149	88-150	88-151	88-152	88-153	88-154	88-155
Your Sample No.	8009	8010	8101	8102	8103	8104	8105	8106
SiO ₂	59.2%	56.8%	68.6%	78.8%	76.5%	73.6%	75.2%	76.8%
Al ₂ O ₃	18.9	20.0	8.70	9.13	10.1	11.6	10.7	10.4
TiO ₂	0.61	0.61	0.81	0.91	0.90	0.84	0.84	0.80
Fe ₂ O ₃	5.64	6.73	2.87	3.08	3.59	4.74	4.58	4.27
MgO	2.40	1.93	0.69	0.61	0.71	0.92	0.91	0.95
CaO	0.11	0.13	0.47	0.17	0.15	0.15	0.23	0.32
MnO	0.059	0.070	0.600	0.166	0.076	0.058	0.069	0.075
Na ₂ O	0.06	0.06	0.49	0.57	0.55	0.53	0.64	0.74
K ₂ O	6.07	4.14	1.77	2.10	2.24	2.48	2.48	2.58
P ₂ O ₅	0.10	0.12	0.21	0.11	0.09	0.09	0.08	0.07
L.O.I. (950°C)	5.85	8.34	14.9	3.69	3.71	4.38	3.86	3.52
TOTALS	99.0%	98.9%	100.1%	99.3%	98.6%	99.4%	99.6%	100.5%

Our Sample No.	88-156	88-157	88-158	88-159	88-160	88-161	88-162	88-163
Your Sample No.	8107	8108	8109	8110	8111	8112	8201	8202
SiO ₂	76.0%	76.2%	74.3%	73.9%	75.9%	80.0%	71.2%	79.3%
Al ₂ O ₃	10.5	10.7	11.5	11.3	10.7	9.29	5.27	9.70
TiO ₂	0.53	0.72	0.76	0.69	0.79	0.88	0.72	0.79
Fe ₂ O ₃	5.67	3.37	3.63	4.19	4.49	2.81	1.37	2.01
MgO	1.06	1.02	1.13	1.18	0.94	0.60	0.22	0.37
CaO	0.17	0.16	0.14	0.13	0.24	0.16	0.09	0.06
MnO	0.064	0.065	0.036	0.082	0.066	0.134	0.049	0.021
Na ₂ O	0.18	0.17	0.13	0.10	0.71	0.65	0.20	0.26
K ₂ O	2.67	3.14	3.05	2.95	2.56	2.32	0.88	1.53
P ₂ O ₅	0.07	0.06	0.06	0.07	0.08	0.09	0.09	0.05
L.O.I. (950°C)	3.99	3.50	4.47	4.66	3.28	3.68	18.7	4.91
TOTALS	100.9%	99.1%	99.2%	99.3%	99.8%	100.6%	98.8%	99.0%

88-164	88-165	88-166	88-167	88-168	88-169	88-170	88-184
8203	8204	8205	8206	8207	8208	8209	8313
76.3%	75.7%	78.6%	83.3%	74.4%	69.8%	75.7%	81.5%
11.8	12.5	11.0	8.72	13.0	14.2	11.6	7.09
0.80	0.79	0.70	0.62	0.71	0.72	0.78	0.67
2.79	3.64	3.35	2.33	4.44	6.43	3.57	2.57
0.53	0.58	0.48	0.30	0.35	0.47	0.53	0.29
0.08	0.07	0.06	0.07	0.05	0.08	0.12	0.05
0.018	0.021	0.019	0.031	0.044	0.075	0.044	0.011
0.29	0.33	0.26	0.20	0.17	0.15	0.34	0.20
1.93	2.15	1.97	1.63	2.39	2.46	2.26	1.16
0.06	0.04	0.05	0.05	0.06	0.07	0.08	0.06
4.57	4.33	3.89	2.59	4.46	5.29	4.18	5.82
99.2%	100.2%	100.4%	99.8%	100.1%	99.7%	99.2%	99.4%

88-171	88-172	88-173	88-174	88-175	88-176	88-177
8210	8211	8212	8303	8304	8305	8306
76.8%	78.3%	80.5%	89.9%	78.8%	76.8%	80.3%
12.0	10.9	9.67	3.75	8.40	10.9	9.88
0.69	0.51	0.75	0.80	0.75	0.74	0.53
3.87	3.91	2.64	0.83	2.61	2.70	2.90
0.48	0.42	0.43	0.16	0.33	0.49	0.46
0.08	0.05	0.08	0.04	0.06	0.07	0.06
0.042	0.047	0.031	0.007	0.012	0.022	0.018
0.26	0.12	0.25	0.14	0.24	0.28	0.22
2.31	2.02	1.80	0.73	1.30	1.60	1.69
0.07	0.07	0.05	0.03	0.08	0.09	0.04
4.13	3.95	3.38	2.50	6.65	5.91	3.94
100.7%	100.3%	99.6%	98.9%	99.2%	99.6%	100.0%

THE PENNSYLVANIA STATE UNIVERSITY
INTER-OFFICE CORRESPONDENCE

Date: January 31, 1980
 From: N. H. Suhr, 311 Mineral Sciences
 To: A. W. Rose, 332 Deike
 Subj: Spectrochemical Analysis of Soils (As Received)

Our No.	89-13	89-14	89-15	89-16	89-17	89-18
Your No.	6-10-2	6-10-5	6-10-7	6-11-5	6-11-8	14-84-3
L.O.I. (750°C)	18.5%	5.74%	6.75%	6.80%	6.73%	4.31%
SiO ₂	63.5	64.8	57.1	61.0	59.0	76.8
Al ₂ O ₃	9.90	16.9	20.8	20.0	21.9	11.0
TiO ₂	0.81	0.97	0.89	0.45	0.27	0.80
Fe ₂ O ₃	3.50	6.15	7.97	5.25	4.70	4.36
MgO	0.65	1.29	1.59	0.33	0.14	0.75
CaO	0.19	0.15	0.27	0.21	0.27	0.16
MnO	0.28	0.05	0.13	0.05	0.06	0.08
Na ₂ O	0.27	0.20	0.07	0.29	0.25	0.46
K ₂ O	1.27	3.44	4.49	4.61	6.19	1.91
P ₂ O ₅	<0.05	<0.05	<0.05	<0.05	<0.05	0.08
SO ₃	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
TOTALS	98.9%	99.7%	100.1%	99.0%	99.5%	100.7%

A.W. Rose -2-

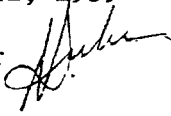
Our No.	89-19	89-20	89-21	89-22	89-23
Your No.	14-84-5	14-84-8	NC-1-5	NC-1-6	NC-1-10
L.O.I. (750°C)	4.07%	4.03%	11.9%	10.7%	8.02%
SiO ₂	76.0	77.3	52.7	55.2	62.5
Al ₂ O ₃	11.4	10.1	25.7	25.5	21.5
TiO ₂	0.80	0.59	0.41	0.33	0.31
Fe ₂ O ₃	4.57	5.44	4.97	4.75	3.02
MgO	0.73	0.63	0.54	0.59	0.75
CaO	0.09	0.12	0.27	0.16	0.23
MnO	0.07	0.07	0.03	0.03	0.05
Na ₂ O	0.43	0.35	0.11	0.13	0.18
K ₂ O	1.97	1.84	2.15	2.52	3.87
P ₂ O ₅	0.08	0.14	0.08	<0.05	<0.05
SO ₃	<0.05	<0.05	<0.05	<0.05	<0.05
TOTALS	100.2%	100.6%	98.9%	99.9%	100.4%

Analyst: H. Gong

NHS/vc

cc: H. Gong

THE PENNSYLVANIA STATE UNIVERSITY
INTER-OFFICE CORRESPONDENCE

Date: February 21, 1989
From: N. H. Suhr 
To: A. W. Rose

Subj: Spectrochemical Analysis of Soils (as received basis)

Our No.	89-46	89-47	89-48	89-49	89-50	89-51	89-52	89-53
Your No.	6-10-4	6-10-7X	6-10-8	6-11-3	6-11-6	6-11-9	6-11-10	6-11-11
L.O.I. (750°C)	5.88%	6.71%	6.76%	9.23%	7.57%	7.05%	6.90%	7.89%
SiO ₂	71.0	58.1	57.6	61.5	58.8	59.7	59.9	57.2
Al ₂ O ₃	13.7	20.4	19.9	17.9	21.7	20.8	21.1	21.2
TiO ₂	0.96	0.95	0.80	0.52	0.44	0.33	0.30	0.48
Fe ₂ O ₃	4.75	7.72	7.84	4.35	4.49	4.90	4.86	5.91
MgO	1.11	1.41	1.41	0.31	0.21	0.31	0.21	0.38
CaO	0.11	0.21	0.07	0.19	0.22	0.23	0.16	0.28
MnO	0.047	0.105	0.133	0.168	0.049	0.076	0.058	0.102
Na ₂ O	0.28	0.11	0.15	0.41	0.31	0.30	0.31	0.22
K ₂ O	2.54	4.78	4.57	4.60	5.44	6.23	5.82	5.33
P ₂ O ₅	0.11	0.11	0.12	0.11	0.07	0.07	0.05	0.07
SO ₃	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
TOTALS	100.5%	100.6%	99.4%	99.3%	99.3%	100.0%	99.7%	99.1%

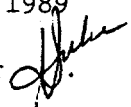
Analyst: H. Gong

NHS/vc

cc: H. Gong

THE PENNSYLVANIA STATE UNIVERSITY
INTER-OFFICE CORRESPONDENCE

Date: March 13, 1989

From: N. H. Suhr 

To: A. W. Rose

Subj: Spectrochemical Analyses of Soils (as-received basis)

Our No.	89-74	89-75	89-76	89-77	89-78	89-79	89-80
Your No.	6-10-3	6-10-4X	6-10-6	6-10-9	6-11-2	6-11-4	6-11-7
L.O.I. (750°C)	6.73%	5.59%	6.02%	6.46%	14.60%	6.32%	6.93%
SiO ₂	72.9	71.0	57.8	56.0	56.5	64.6	57.3
Al ₂ O ₃	12.6	14.1	20.4	20.2	16.8	17.3	21.9
TiO ₂	1.04	0.95	0.97	0.98	0.58	0.51	0.35
Fe ₂ O ₃	3.97	4.78	7.30	8.04	4.89	4.81	5.53
MgO	0.82	1.00	1.34	1.66	0.33	0.34	0.15
CaO	0.20	0.25	0.32	0.24	0.33	0.29	0.32
MnO	0.12	0.04	0.05	0.17	0.21	0.06	0.08
Na ₂ O	0.25	0.24	0.05	0.05	0.35	0.33	0.27
K ₂ O	1.85	2.55	4.72	4.29	4.22	4.51	5.62
P ₂ O ₅	0.11	0.12	0.14	0.13	0.18	0.13	0.09
SO ₃	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
TOTALS	100.6%	100.6%	99.1%	98.2%	99.0%	99.2%	98.5%

Our No.	89-81	89-82	89-83	89-84	89-85	89-86	89-87
Your No.	14-84-1	14-84-2	14-84-4	14-84-6	14-84-7	14-84-9	NC-1-2
L.O.I. (750°C)	7.12%	4.22%	3.83%	3.71%	3.12%	4.29%	3.58%
SiO ₂	75.8	77.3	75.5	76.9	81.6	73.0	85.0
Al ₂ O ₃	8.84	10.3	10.9	10.3	8.29	10.5	6.40
TiO ₂	0.82	0.87	0.84	0.84	0.72	0.63	0.29
Fe ₂ O ₃	3.65	4.03	4.61	4.70	3.32	7.17	0.96
MgO	0.57	0.65	0.77	0.75	0.53	0.76	0.10
CaO	0.19	0.32	0.21	0.22	0.13	0.23	0.06
MnO	0.09	0.10	0.08	0.08	0.01	0.07	0.01
Na ₂ O	0.43	0.45	0.45	0.47	0.39	0.36	0.13
K ₂ O	1.59	1.76	1.95	1.76	1.46	1.99	2.09
P ₂ O ₅	0.13	0.10	0.10	0.11	0.09	0.21	0.05
SO ₃	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
TOTALS	99.2%	100.1%	99.2%	99.8%	99.7%	99.2%	98.7%

Our No.	89-88	89-89	89-90	89-91	89-92	89-93	89-94
Your No.	NC-1-3	NC-1-4	NC-1-7	NC-1-8	NC-1-9	NC-1-11	6-11-6X
L.O.I. (750°C)	2.97%	3.29%	8.82%	8.54%	8.03%	8.29%	8.58%
SiO ₂	84.3	84.5	59.4	61.2	61.8	63.4	57.2
Al ₂ O ₃	7.43	7.69	22.4	21.1	20.1	19.6	22.4
TiO ₂	0.31	0.36	0.46	0.44	0.44	0.44	0.41
Fe ₂ O ₃	1.19	1.27	4.34	4.06	3.60	3.34	4.53
MgO	0.12	0.16	0.77	0.77	0.79	1.04	0.25
CaO	0.08	0.04	0.22	0.28	0.22	0.22	0.26
MnO	0.00	0.00	0.04	0.03	0.03	0.05	0.04
Na ₂ O	0.13	0.13	0.13	0.13	0.15	0.13	0.27
K ₂ O	2.26	2.25	2.80	2.95	3.47	3.30	5.62
P ₂ O ₅	0.06	0.07	0.05	0.04	0.05	0.04	0.09
SO ₃	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
TOTALS	98.9%	99.8%	99.4%	99.5%	98.7%	99.8%	99.6%

Analyst: H. Gong

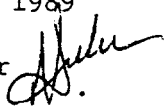
NHS/vc

cc: H. Gong

THE PENNSYLVANIA STATE UNIVERSITY

INTER-OFFICE CORRESPONDENCE

Date: April 13, 1989

From: N. H. Suhr 

To: A. W. Rose

Subj: Spectrochemical Analyses of Soils (as received basis)

Our No.	89-235	89-236	89-237	89-238	89-239	89-240
Your No.	NC-1-2 (+10)	NC-1-11 (+10)	6-10-3 (rocks)	6-10-3 (E +10)	6-11-4 (rocks)	6-11-4 (+10)
L.O.I. (750°C)	2.00%	3.73%	5.14%	5.97%	2.88%	5.46%
SiO ₂	93	81	68.3	64.1	66.6	65.1
Al ₂ O ₃	3.11	10.9	14.9	16.1	16.4	16.4
TiO ₂	0.15	0.14	0.80	0.81	0.28	0.51
Fe ₂ O ₃	0.80	1.37	6.37	7.41	3.30	5.65
MgO	0.11	0.27	1.22	1.29	0.22	0.29
CaO	0.06	0.12	0.23	0.23	1.11	0.53
MnO	<0.05	<0.05	0.09	0.17	<0.05	0.10
Na ₂ O	0.00	0.09	0.41	0.41	3.19	0.87
K ₂ O	1.04	2.22	2.93	3.30	5.70	4.65
P ₂ O ₅	<0.05	<0.05	0.12	0.17	<0.05	0.14
SO ₃	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
TOTALS	100.3%	99.8%	100.5%	100.0%	99.7%	99.7%

A. W. Rose -2-

Our No.	89-241	89-242	89-243	89-244	89-245
Your No.	6-11-9 (rocks)	6-11-9 (+10)	14-84-8 (+10)	14-89-8 (rocks)	14-89-08 (rocks)
L.O.I. (750°C)	6.14%	7.28%	3.29%	1.74%	1.95%
SiO ₂	59.1	61.3	79	89	89
Al ₂ O ₃	19.9	18.8	8.32	4.25	5.84
TiO ₂	0.41	0.41	0.61	0.33	0.22
Fe ₂ O ₃	6.01	5.98	5.54	3.08	1.23
MgO	0.23	0.17	0.62	0.28	0.21
CaO	0.27	0.21	0.16	0.09	0.10
MnO	0.05	0.07	0.05	<0.05	0.06
Na ₂ O	0.29	0.20	0.43	0.29	<0.05
K ₂ O	6.63	4.85	1.62	0.94	1.33
P ₂ O ₅	0.07	0.09	0.11	0.06	0.05
SO ₃	<0.05	<0.05	<0.05	<0.05	<0.05
TOTALS	99.1%	99.4%	99.8%	100.1%	99.9%

Analyst: H. Gong

NHS/vc

cc: H. Gong

THE PENNSYLVANIA STATE UNIVERSITY
INTER-OFFICE CORRESPONDENCE

Date: May 1, 1990
From: N. H. Suhr, *NH* Mineral Sciences
To: Dan Greeman; 227 Deike
Subj: Spectrochemical Analysis of Soils (values on ash basis)

Our No.	90-212	90-213	90-214	90-215	90-216	90-217	90-218
Your No.	8-99-2	8-99-3	8-99-5	8-99-7	8-99-9	8-99-12	TN-1-2
HTA (800°C)	11.9%	4.65%	3.97%	4.65%	3.91%	4.46%	8.83%
SiO ₂	82.4	83.1	77.1	69.2	71.9	70.1	91.0
Al ₂ O ₃	8.93	8.01	12.7	16.6	15.0	15.5	5.22
TiO ₂	0.85	0.74	0.97	0.96	0.94	1.01	0.68
Fe ₂ O ₃	3.51	3.04	5.29	6.34	5.97	6.02	2.08
MgO	0.56	0.54	1.05	1.28	1.18	1.29	0.23
CaO	0.30	0.27	0.12	0.13	0.26	0.18	0.31
MnO	0.09	0.08	0.05	0.09	0.11	0.09	0.17
SrO	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
BaO	0.04	0.03	0.04	0.05	0.05	0.05	0.03
Na ₂ O	0.77	0.74	0.77	0.81	0.93	0.90	0.07
K ₂ O	1.49	1.43	2.59	3.52	3.12	3.33	0.58
P ₂ O ₅	0.15	0.10	0.13	0.15	0.17	0.11	0.05
SO ₃	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
TOTALS	99.1%	98.1%	100.8%	99.1%	99.6%	98.6%	100.4%

THE PENNSYLVANIA STATE UNIVERSITY
INTER-OFFICE CORRESPONDENCE

Date: May 14, 1990 *[Signature]*
 From: N. H. Suhr, *[Signature]* Mineral Sciences
 To: Dan Greenman, 227 Deike
 Subj: Spectrochemical Analysis of Soils

Our No.	90-268	90-269	90-270	90-271	90-272	90-273
Your No.	NY-1-2	NY-1-7	NY-1-10	NY-1-12	8-99-P	8-99-9R
HTA (800°C)	71.6%	87.1%	98.2%	99.7%	95.5%	95.8%
SiO ₂	80.0	77.5	78.0	81.2	71.8	70.9
Al ₂ O ₃	8.17	9.55	7.98	7.85	15.1	15.8
TiO ₂	0.84	0.82	1.00	0.64	1.00	0.97
Fe ₂ O ₃	5.00	5.50	7.09	4.10	6.44	6.50
MgO	0.50	0.68	0.53	0.42	1.19	1.29
CaO	0.82	0.95	0.96	0.86	0.22	0.29
MnO	0.04	0.04	0.04	0.03	0.04	0.08
SrO	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
BaO	0.03	0.04	0.03	0.04	0.04	0.04
Na ₂ O	2.28	2.37	2.29	2.11	1.01	0.99
K ₂ O	2.84	2.98	2.90	2.90	2.86	3.35
P ₂ O ₅	0.04	0.04	0.04	0.03	0.04	0.08
SO ₃	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
TOTALS	100.6%	100.5%	100.9%	100.2%	99.7%	100.3%

Dan Greenman - 2 -

Our No.	90-274	90-275	90-276	90-277	90-278	90-279
Your No.	IL-1-5	IL-1-7	IL-1-9	TN-1-3	TN-1-8	TN-Rock 1
HTA (800°C)	92.3%	94.4%	97.0%	94.9%	89.5%	56.5%
SiO ₂	73.7	77.5	84.5	91.0 <i>E</i>	62.6 <i>TD</i>	12.6
Al ₂ O ₃	13.6	11.4	8.00	5.66 <i>E</i>	25.3 <i>LD</i>	1.95
TiO ₂	0.69	0.65	0.27	0.67	0.63	0.05
Fe ₂ O ₃	5.50	4.57	3.24	2.18	8.15	0.47
MgO	1.22	1.09	0.72	0.26	1.03	32.4
CaO	0.82	0.92	0.65	0.06	0.34	50.6
MnO	0.10	0.10	0.07	0.15	0.03	0.02
SrO	<0.01	<0.01	<0.01	<0.01	<0.01	0.02
BaO	0.05	0.05	0.03	0.02	0.01	0.01
Na ₂ O	0.99	1.12	0.98	0.06	0.03	0.08
K ₂ O	2.24	2.14	2.05	0.36	1.37	0.54
P ₂ O ₅	0.10	0.10	0.07	0.15	0.04	0.02
SO ₃	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
TOTALS	99.0%	99.6%	100.6%	100.6%	99.5%	98.8%

NOTE: Results on ash basis

Analyst: H. Gong

NHS/vc

cc: A. Rose
H. Gong

THE PENNSYLVANIA STATE UNIVERSITY

INTER-OFFICE CORRESPONDENCE

Date: May 1, 1990
 From: N. H. Suhr, *NH* Mineral Sciences
 To: Dan Greeman; 227 Deike
 Subj: Spectrochemical Analysis of Soils (values on ash basis)

Our No.	90-212	90-213	90-214	90-215	90-216	90-217	90-218
Your No.	8-99-2	8-99-3	8-99-5	8-99-7	8-99-9	8-99-12	TN-1-2
HTA (800°C)	11.9%	4.65%	3.97%	4.65%	3.91%	4.46%	8.83%
SiO ₂	82.4	83.1	77.1	69.2	71.9	70.1	91.0
Al ₂ O ₃	8.93	8.01	12.7	16.6	15.0	15.5	5.22
TiO ₂	0.85	0.74	0.97	0.96	0.94	1.01	0.68
Fe ₂ O ₃	3.51	3.04	5.29	6.34	5.97	6.02	2.08
MgO	0.56	0.54	1.05	1.28	1.18	1.29	0.23
CaO	0.30	0.27	0.12	0.13	0.26	0.18	0.31
MnO	0.09	0.08	0.05	0.09	0.11	0.09	0.17
SrO	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
BaO	0.04	0.03	0.04	0.05	0.05	0.05	0.03
Na ₂ O	0.77	0.74	0.77	0.81	0.93	0.90	0.07
K ₂ O	1.49	1.43	2.59	3.52	3.12	3.33	0.58
P ₂ O ₅	0.15	0.10	0.13	0.15	0.17	0.11	0.05
SO ₃	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
TOTALS	99.1%	98.1%	100.8%	99.1%	99.6%	98.6%	100.4%

D. Geenman -2-

Our No.	90-219	90-220	90-221	90-222	90-223	90-224	90-225	90-226
Your No.	TN-1-4	TN-1-6	TN-1-11	NY-1-3	NY-1-5	IL-1-1	IL-1-2	IL-1-3
HTA (800°C)	4.40%	10.1%	6.12%	3.40%	2.08%	12.1%	8.57%	7.93%
SiO ₂	86.2	62.4	78.5	78.7	77.4	75.0	78.0	76.6
Al ₂ O ₃	7.21	25.3	14.4	8.82	9.49	11.6	11.2	12.0
TiO ₂	0.70	0.57	0.24	0.79	0.79	0.79	0.78	0.75
Fe ₂ O ₃	2.74	7.96	4.02	4.84	5.03	5.31	3.93	4.12
MgO	0.29	0.82	0.35	0.47	0.59	0.78	0.75	0.89
CaO	0.09	0.05	0.02	0.84	0.90	1.30	0.87	0.70
MnO	0.09	0.04	0.03	0.03	0.01	0.13	0.15	0.15
SrO	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
BaO	0.02	0.02	0.02	0.04	0.04	0.06	0.07	0.07
Na ₂ O	0.06	0.03	0.04	2.26	2.30	0.89	0.90	0.89
K ₂ O	0.63	1.30	0.81	2.74	2.93	2.32	2.35	2.36
P ₂ O ₅	0.04	<0.02	0.04	0.11	0.05	0.12	0.10	0.09
SO ₃	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
TOTALS	98.1%	98.5%	98.5%	99.6%	99.5%	98.3%	99.1%	98.6%

Analyst: H. Gong

NHS/vc

cc: A. Rose
H. Gong

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