

1994-11

**MEASUREMENT OF THE RADON SURFACE FLUX FROM UNDISTURBED SOIL
USING ELECTRET ION CHAMBERS**

Lorin R. Stieff and Paul Kotrappa
Rad Elec Inc.
Frederick, MD

J.E. Rechcigl and Curtis Nobel
Agricultural Research and Education Center
Ona, FL

ABSTRACT

Methods for measuring the radon surface flux without disturbing either the soil or the flux are of interest in geologic studies and in monitoring the radon flux from uranium mine tailings piles. Such flux measurements may also be of use in assessing the radon risk at potential building sites. The flow-through surface flux method does not perturb either the radon flux or the soil. For this method the flux, J , is given by the simple equation

$$J = (R \times F) / (A \times 60) \text{ where :}$$

J is in $\text{pCi} / \text{m}^2 \cdot \text{s}$;

R , the radon concentration, is in pCi / l ;

F , the flow rate, is in $\text{l} / \text{min.}$, and

A , the area of the soil being measured, is in m^2 .

The constant, 60, converts the flow rate from $\text{l} / \text{min.}$ to $\text{l} / \text{sec.}$

This method requires an inverted basin which defines the area of the soil being measured. Equilibrium radon concentrations are established inside the basin between the radon surface flux and a steady, 0.5 to 1.0 l / min , flow of ambient air whose radon concentration is low. The radon concentration in the basin is measured using electret ion chambers. Comparison of preliminary surface flux data from the flow-through technique and data from Large Area Activated Charcoal Canisters measurements suggest that even at very low flux rates the flow-through method can give useful results.

INTRODUCTION

The measurement of the radon surface flux, the rate at which radon atoms cross a unit area of the soil-atmosphere interface per unit time, has been studied for over 60 years. Since the late 1970s indirect methods of measuring surface flux based on the concentrations of radon in soil gas and radium in soil have been the subject of numerous investigations. This interest has been driven by potential problems associated with the accumulation of radon in houses and other large structures. Tanner 1994 has reviewed this recent literature on the measurement of the radon source potential of soils and has divided these studies into two large groups: radon potential mapping and site specific methods. Radon potential mapping involves "surrogate" methods which attempt to establish correlations between rock/soil types and the indoor concentrations of radon. These studies result in the preparation of maps which classify the radon potential of an area as "high-risk", "low-risk" or "normal-risk". The site specific methods may be applied either in the field or to soil samples analyzed in the laboratory. The *in situ* soil measurements have used a variety of soil probes (emanometers) to extract and measure ^{222}Rn in soil gas. These *in situ* measurements usually require additional measurements of soil permeability, moisture content and other factors. The laboratory methods are applied to soil samples brought in from the field, dried and then analyzed for such "invariant" properties as radium activity, dry density, porosity, gas permeability, etc.

In the Tanner review paper (op. cit.) methods for the direct measurement of the radon surface flux such as flow-through and accumulator methods were not included, perhaps, because there are relatively few examples in the literature. In a recent paper describing a new flow-through method, Livingston et al 1990, have combined the use of passive electret ion chambers (EIC's) with the flow-through technique, resulting in a simple, inexpensive instrument that can give direct, long term measurements of the undisturbed radon surface flux. The flow-through method has been described in general terms in NCRP Report No. 70 1984. Mochizuki 1978 includes a description of a flow through instrument (after Wilkening 1960), however, data obtained using the instrument was not presented. Hartley et al 1981 has described a pressure-balanced recirculating system open to the atmosphere in which the radon is adsorbed in an activated carbon trap. The Hartley instrument was used to measure the radon surface flux after asphalt emulsion seals were applied to the surface of test areas at the Grand Junction uranium mill tailings pile. Although the data obtained using the Livingston flow-through instrument is limited, it suggests that the method should find applications in geologic studies, in monitoring the regulatory requirements for mine tailings, and possibly in pre-construction site evaluation.

THE PRINCIPLE OF THE FLOW-THROUGH METHOD

The direct measurement of the radon surface flux using the flow-through method is based on first principles and is given by the following equation:

$$J = (R \times F) / (A \times 60) \text{ where,}$$

J = the radon flux in units of pCi / m² . sec.

R = average radon concentration in the enclosure during the measurement period, units of pCi / l.

F = the flow rate of ambient air through the enclosure in units of l / min. .

A = The area of the ground covered by the enclosure in units of m² .

The numerical constant, 60, converts the flow rate of the ambient air from l / min. to l / sec. Note that the volume of the enclosure does not appear in the equation. However, the volume of the enclosure does effect the time required for the radon surface flux to mix and equilibrate with the flow of ambient air entering the enclosure. In this study the enclosure was a plastic basin 48.9 cm long by 27.95 cm wide and 15.25 cm deep and the equilibrium times relatively short compared to the measurement periods. It should also be noted that this technique does not require any additional soil measurements, such as: soil porosity, density or moisture content. However, the evaluation of flow-through flux data can be improved by the availability of data on soil moisture content.

MEASUREMENT PROCEDURES

The procedures used in this report to measure the radon surface flux using the flow-through method have been somewhat modified from the original procedures described by Livingston et al in an internal report to Rad Elec, Inc. 1990. A drawing of Livingston's experimental equipment is shown in Fig. 1. The principal difference between the two procedures involves the replacement of the compressed air scuba tanks and/or large compressed air cylinders as sources of the air used in the flow-through method. These tanks were difficult to deploy and maintain in any significant numbers in the field and have been replaced with small, precision DC electric pumps supported by a single, fully charged 12 volt car battery. In contrast to the aged air supplied from the tanks, the ambient air supplied by the DC pumps does contain radon and a correction for this radon must be made. The pumps draw 0.09 amps and a car battery can support 12 surface flux units over a 48 hour measurement period. An additional change involves the metal flanges of Figure 1 which has been replaced with separate, rectangular metal frames whose dimensions closely match the opening of the rectangular plastic basins. The welded frames are fabricated from 1.5 inch 90 degree angle iron and are pressed into the earth to define the area of the soil to be measured. Foam gaskets are glued to the open lips of the basins and form a tight seal when the inverted basins are placed on the metal frames. During measurements, the basins are secured to the metal frames with an elastic pongee cord. See Fig. 2. The pumps are enclosed in small, rectangular plastic containers for protection from the environment and are placed on top of the inverted basins. A flow meter connected between the outlet of the pump and the inlet to the basin is used to regulate the air flow. Finally, to minimize the mid-day temperatures inside the

basins , the exteriors of the inverted basins is covered with tightly fitting, commercially available, heavy duty aluminum foil pans.

The largest sets of radon surface flux measurements using the flow-through method have been made at the Agricultural Research and Education Center, University of Florida, Ona Florida.. These measurements are part of a major research program which is studying the improvement in pasture yield as well as the environmental impact of the application of different amounts of slightly radioactive by-product phosphogypsu as a sulfur and calcium soil amendment., Rechcigl et al 1992, Rechcigl et al 1993, and Alcordo and Rechcigl 1994 In addition to pasture yields and detailed radiochemical studies, a major ambient atmospheric radon and gamma background study also has been undertaken using EIC's. The Ona study involves two different bahiagrass pastures, the East and West sites, which involve different types of soil. Each site was divided into 36 plots and phosphogypsum applications of 20 Mg/ ha, 10 Mg/ha and 0g/ ha were spread on the plots according to a statistical design. Surface flux measurements following EPA protocols have been made at approximately two month intervals using the Large Area Activated Charcoal Canisters (LAACC) method. One set of duplicate flow-through surface flux measurements have been completed for the West site.

RESULTS

An example of the radon surface flux and LAACC measurements obtained for a group of six plots from the West site is given in Table 1. The flow-through measurements were made over a period of approximately two days. Because of the low radon flux rates, it was necessary to use the very sensitive, 960 ml EICs (H chambers) and high sensitivity electrets (ST) in order to obtain a sufficiently large electret voltage drops for meaningful measurements. Duplicate H chambers were used within each basin and two flow-through instruments were set up within each of the six plots. The use of duplicate H chambers within each basin and duplicate instrumental setups within each plot provides some reassurance on the performance of this new technique. In addition, the duplicates provide the basis for omitting from the average the very large voltage drop for location 1214 , 166 volts , possibly due to partial accidental discharge of the electret during removal of the electrets in the field. The duplicates also provide the basis for acceptance of the large voltage drops for location 1291 and 1292, 87 and 83 volts, respectively. These higher radon surface flux rates may reflect either some inhomogeneity in the radium content of the phosphogypsum applied to the plot or to the rate of application of the phosphogypsum itself.

In addition to the two H- chambers placed inside the basins, one H- chamber was placed on top of each of the inverted basins to measure the concentration of both the ambient atmospheric radon and the gamma backgrounds. Data from these H-chamber was used to correct for the gamma contribution as well as the radon added to the basin from the atmosphere during the measurement period. From Table 1 it can be seen that the voltage drops from the background H-chambers are low but in every case exceed the recommended minimum voltage drop of 10 volts for a useful measurement. The relatively good agreement between the background voltage drops within each of the plots suggests that, inspite of the low voltages, the corrections are probably quite close to the correct values. However, to minimize this uncertainty, future background H-chamber measurements will be extended to four days.

DISCUSSION

From Table 1 it can be seen that there is a reasonably good correlation between the flow-through surface flux values and the amount of phosphogypsum applied to the plots. For example: the plots which did not receive any phosphogypsum (0Mg/ha) have the lowest surface fluxes, 0.052 and 0.035 pCi/ m² .s, the plots receiving 10Mg/ha had fluxes of 0.083 and 0.078, respectively while the plots that received 20Mg/ha had the highest flux rates, 0.118 and 0.227 pCi/ m² .s. There is reasonable agreement between the flux rates for the flow-through and LAACC methods for the plots which did not receive any phosphogypsum, 0.052 and 0.05 for plot 121 and 0.067 and 0.035 for plot 128., respectively. However, the remainder of the LAACC results do not correlate well with the amounts of phosphogypsum applied to the different plots. It should be noted, however, that all of the flow-through flux data for the West site has not been reduced and it is possible that the initial correlation shown in Table 1 will not be supported. Furthermore, measurements for the East site remain to be done. A final assessment of the flow-through

method must await not only additional measurements but also the availability of a surface flux test facility where the performance of the different methods can be compared and calibrated against known surface flux rates.

The first measurements of surface flux using the Livingston/Jester method were made on the phosphogypsum stacks at the IMC plant, New Wales, Florida. Very preliminary data gave flow-through radon surface flux rates of 5.6 pCi / m² .s compared to 18.3 pCi l / m² .s for the LAACCs. Evaluation of this limited data should be deferred because significant changes in field procedures have been made since these measurements were made and until more data is available. Additional measurements are planned at IMC for 1995.

CONCLUSIONS

The results of the first set of flow-through radon surface flux measurements suggest that even at very low flux rates useful comparative data can be obtained. A final assessment of the flow-through surface flux method must await not only additional measurements but also the availability of a surface flux test facility where the performance of different surface flux methods can be compared and calibrated against known surface flux rates.

REFERENCES

Alcorido, I.S., Rechcigl, J.E. Environmental Aspects of Phosphogypsum and other By-product Gypsums. Research and Education Center, University of Florida, Ona, FL 1994

Hartley, J.N.: Freeman, H.D.: Baker, E.G.: Elmore, M.R.: Nelson, D.A.: Voss, C.F., Koehmstedt, P.L. Field Testing of Asphalt Emulsion Radon Barrier System In: Symposium on Uranium Mill Tailings Management, Geotechnical Engineering Program, Civil Engineering Dept. Colorado State University, Fort Collins Co 1981

Livingston, J.V.: Jester, W.A.: Kotrappa, P. E-Perm Measurement of Radon Flux from Undisturbed Soil. 1990 Annual Meeting of the American Nuclear Society, 61 p38-39. (TRANO 62-1 482 1990, ISSN: 0003-018X)

Mochizuki, S.: Sekikawa, T.: Radon-222 Exhalation and Its Variation in Soil Air. Proc. 3d Int. Symp. Natural Radiation Environment, Houston, Texas, Vol. 1, 105-116

National Council on Radiation Protection and Measurement. Exposures from the Uranium Series with Emphasis on Radon and its Daughters. NCRP Report No. 70, 1984

Rechcigl, J.E., Alcorido, I.S., Roessler, C.E., Littell, R.C. Influence of Phosphogypsum on Forage Yield and Quality and on the Environment in a Typical Florida Spodosol Soil (Final Report), Florida Institute of Phosphate Research, Bartow FL 1993

Rechcigl, J.E., Alcorido, I.S., Roessler, C.E. Stieff, L.R. Methods of Measuring Ambient Atmospheric Radon and Radon Surface Flux associated with Phosphogypsum Treatment of a Florida Spodosol Soil. Commun Soil Sci. Plant Anal., 23(17-20).2581-2594 1992

Tanner, A.B.: Measurement and Determination of Radon Source Potential, A Literature Review. NISTIR 5399, National Institute of Standards and Technology, United States Department of Commerce, Gaithersburg, MD 20899, 1994

TABLE 1: RADON SURFACE FLUX MEASUREMENTS FOR THE PERIOD 4/05/94 TO 4/07/94, ONA ,FLORIDA

LOCATION	dVOLT	RN CONC pCi/l	RN CONC AVG, pCi/l	BKG CONC pCi/l	CORR RN CONC. pCi/l	FLOW RATE l/min	SUR FLUX pCi m ² s ⁻¹	AVG SF FL pCi m ² s ⁻¹	LAACC pCi m ² s ⁻¹	APPLICATION Mg/ha
1191	25	1.19							0.02 (a)	
1192	25	1.19	1.19							
1193	12			0.6	0.59	1.0	0.072			
1194	30	1.44							0.03 (b)	
1195	29	1.42	1.43						Avg	
1196	15			0.69	0.74	1.05	0.095	0.083	0.025	10
1214	166	8.53	Omit							
1215	21	1.02	1.02						0.06 (a)	
1216	14			0.68	0.34	1.0	0.041			
1211	23	1.21							0.04 (b)	
1212	23	1.16	1.185						Avg	
1213	14			0.64	0.545	0.95	0.063	0.052	0.05	0
1201	30	1.47								
1202	30	1.44	1.455						0.08 (a)	
1203	13			0.6	0.855	1.0	0.104			
1204	32	1.59							0.05 (b)	
1205	32	1.76	1.675						Avg	
1206	13			0.61	1.075	1.0	0.131	0.118	0.065	20
1284	27	1.37								
1285	29	1.45	1.41						0.04 (a)	
1286	17			0.86	0.55	0.85	0.057			
1281	24	1.15							0.03 (b)	
1283	24	1.17	1.16						Avg	
1282	14			0.65	0.51	1.25	0.078	0.067	0.035	0
1294	29	1.6								
1295	27	1.3	1.145						0.07 (a)	
1296	13			0.66	0.485	1.0	0.059			
1291	87	4.2							0.04 (b)	
1292	83	3.97	4.085						Avg	
1293	18			0.85	3.235	1.0	0.395	0.227	0.055	20
1304	44	2.21								
1305	42	2.11	2.16						0.01 (a)	
1306	13			0.66	1.5	0.45	0.082			
1301	36	1.98							0.02 (b)	
1302	49	2.24	2.11						Avg	
1303	11			0.59	1.52	0.40	0.074	0.078	0.015	10

Measurement Period -1.94 days, Chamber Type- H, Electret Type ST

a- LAACC measured 2/28/94, b-LAACC measured 5/09/94

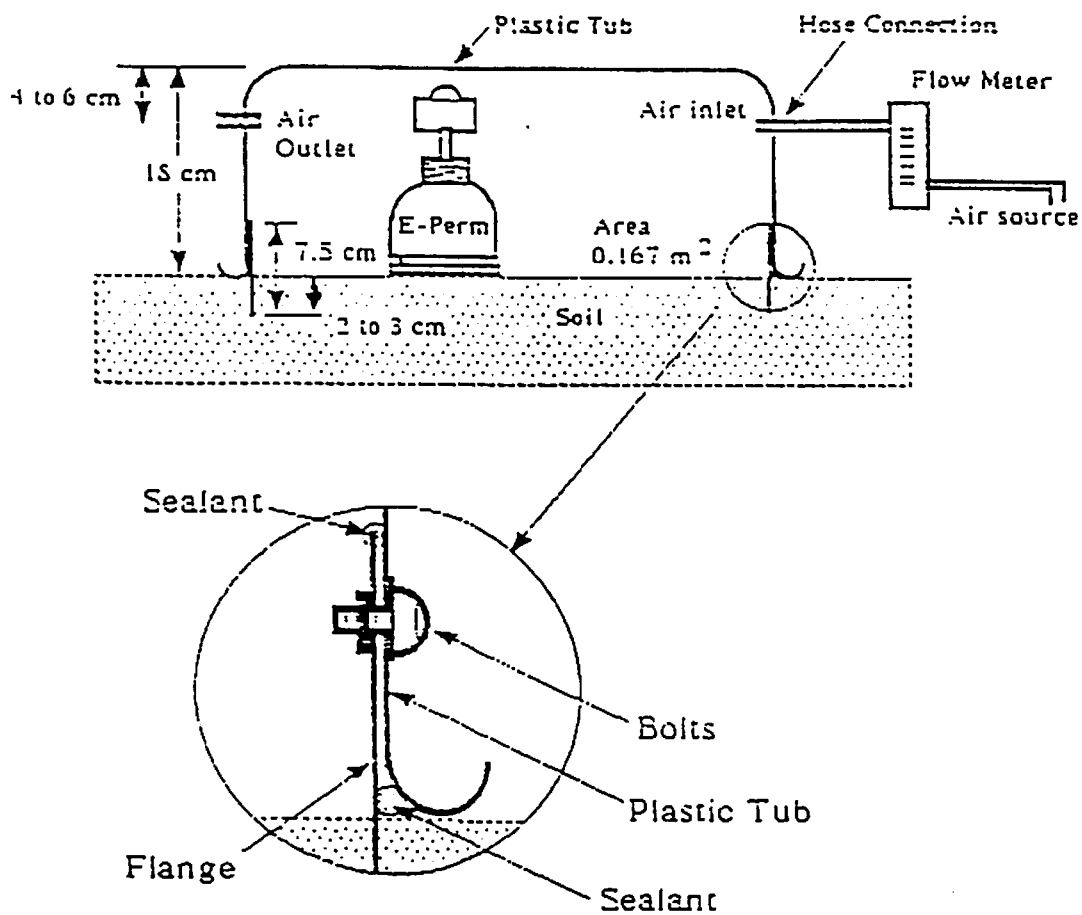


Fig. 1: The Livingston Surface Flux Monitor

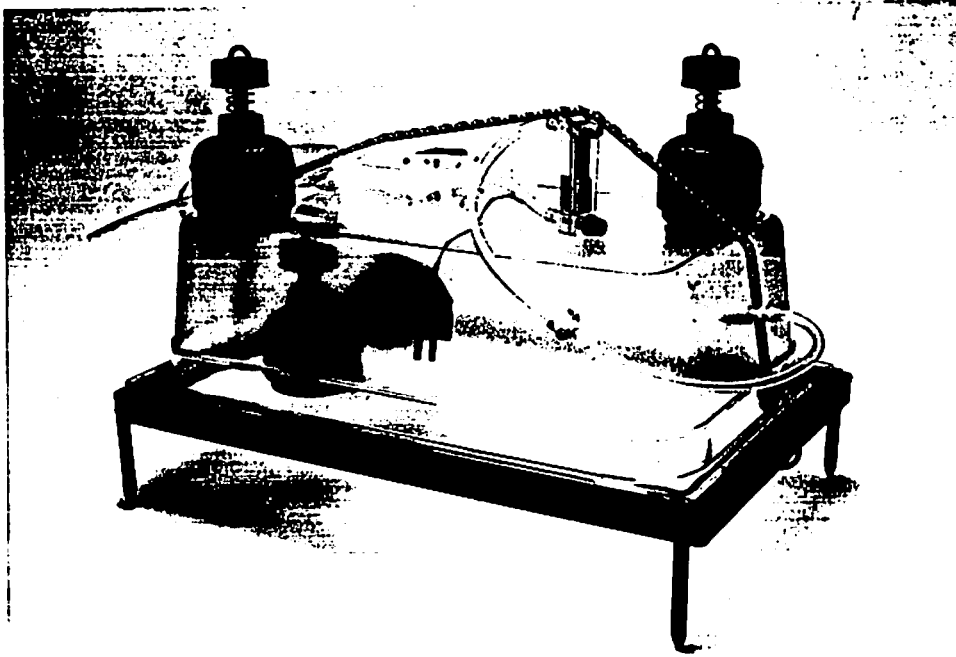


Fig. 2: The Ona Surface Flux Monitor