A PRELIMINARY COMPARISON OF RADON SURFACE FLUX MEASUREMENT USING LARGE AREA ACTIVATED CHARCOAL CANISTERS (LAACC) and ELECTRET ION CHAMBERS (EIC)

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ABSTRACT

As part of an ongoing project studying the radiological and environmental effects of the application of phosphogypsum (PG) to agricultural land, a preliminary comparison was made between the base line EPA radon flux measurement technology that uses LAACC's and new methods using EIC's. These studies were made at the Range Cattle Research and Education Center. The radon sources for these tests consisted of six trays (0.61 by 0.92 m) filled with PG (approx. 21pCi g^{-1} ^{226}Ra) -- three to a depth of 7.6 cm and three to a depth of 3.8 cm . The LAACC monitors were exposed for a period of approximately 24 hours and the EIC monitors for approximately two days. This study suggests that both technologies provide comparable results. The problems encountered as well as the advantages and disadvantages of these two technologies are discussed.

INTRODUCTION

In 1990 the University of Florida (UF) initiated a series of research project on the agricultural and environmental impact of the application of by-product phosphogypsum (PG) containing approximately 22 pCi/g of ^{226}Ra (Alcordo and Rechcigl, 1995) to test forage sites at the Range Cattle Research and Educational Center, Institute of Food and Agricultural Sciences, Ona, Florida. The project was supported by the Florida Institute of Phosphate Research (FIPR), Bartow, Florida. The results of this project have been reported in numerous papers including, Rechcigl J.E., et al (1992), Littell, R.C., and Kundu, S.(1992), Alcordo,I.S. and Rechcigl (1993), Rechcigl, J.E., et al (1994), Roessler, C.E., et al,(1994), Stieff, L.R., et al (1994), and Alcordo, I.S., and Rechcigl, J. E. (in press).

There is an interest in the agricultural and other productive uses of by-product PG ($CaSO_4.2H_2O$) from the production of phosphoric acid because of the large quantities being produced --it is estimated that the quantity stored in Florida PG stacks by the year 2000 will exceed 1 billion Mg. Recent studies have shown that PG can be used as a S and Ca soil amendment to meet the Ca and S deficiencies in soil (Alcordo and Rechcigl, 1993, op cit.). However, because of the presence of ^{226}Ra and its decay products (20 to 30 pCi g^{-1} for PG derived from Central Florida phosphate rock), the U. S. Environmental Protection Agency (EPA) places severe restrictions on the use of PG. One objective of the UF studies is to develop data to support a comprehensive assessment of the environmental impact of the application of PG to forage lands.

The major radiological question in the agricultural application of PG is whether there should be concern for indoor Rn in future structures built over land that has been treated with PG repetitively for many years. In the UF, Rn flux is being measured as an indicator of the Rn source term as a function of PG treatment rate and time after application. The primary Rn flux method being used is the EPA baseline method, the Large Area Charcoal Canister (LAACC) technique. Other radiological characteristics being studied but not a subject of this paper include, gamma radiation above ground, ambient atmospheric Rn, and ^{226}Ra and its decay products, ^{210}Pb and ^{210}Po in soil, groundwater, and forage.

In 1991 Rad Elec, Inc. (REI) offered to demonstrate in the field a recently developed, flow -through (EIC) surface flux method, Livingston, et al (1989). The flow-through method is a method that returns to first principles. In the absence of any readably available surface flux facility or standards, the flow-through method offers a means of independently confirming the results LAACC method. Also, the flow-through EIC method has the potential of measuring very low surface flux rates because the measurement time can be extended for days, if necessary, In contrast 24 hr exposure periods are specified for LAACC measurements. At Ona, the counting errors associated with the LAACC field measurements are of the same order of magnitude as many of the very low surface flux measurements themselves.. Between 1991 and 1993 a number of different configurations of the flow-through method were tried at the test sites. These field experiments resulted in the surface flux procedures currently used and have been described by Stieff, L.R., et al (1994). More recently, a passive Rn surface flux method using a modified 960 ml EIC was developed by REI (Stieff, et al, 1996) and field tests of this passive flux monitor have been underway at Ona since 1995.

As part of evaluating the performance of methods in the UF study, a program of comparison of LAACC measurements by two Florida laboratories was set up in September of 1996. This comparison program included using Rn flux test sources consisting of various depths of PG on trays in an indoor setting. Although not a part of the original plan, the establishment of the test sources provided a opportunity for the *ad hoc* inter-comparison of EIC methods with each other and with LAACC methods.

EXPERIMENTAL SETUP

The test Rn sources consisted of wooden trays filled to various depths with the same PG used in the field tests. The trays were 61 cm (2 ft) x 91 cm (3 ft) in size with plywood bottoms. The array of sources included triplicate trays for each of three PG depths -- 0, 3.8 cm (1.5 in) and 7.6 cm (3.0 in). The 0 depth PG source consisted of a plain 2 ft x 3 ft plywood board. Hereafter, these will be referred to as the "control", 1.5-in and 3.0-in sources, respectively. The trays, loosely filled with PG were placed on potting tables in a well-ventilated greenhouse at the Ona Research Center, and the surface of the PG was leveled.

The EIC flow-through monitor involves a collection chamber with one open face that is coupled to the Rn-emitting surface. A gentle stream of ambient air is passed through the chamber and the Rn concentration in the incoming air as well as the air inside the collection chamber are measured with EIC's. The collection chambers used in this study consist of clear plastic basins 49 cm by 28.6 cm with an area of approximately 0.14 m^2. The depth of the basins is 15 cm. The basins are placed upside down on the surface of the phosphogypsum and two standard 960 ml, hemispherical EIC's are placed inside the basin with the hemispheres pressed into the surface of the PG. The basins are held firmly in place on the phosphogypsum trays with bungee cords which are secured to the wire mesh of the potting tables. One monitor on the 3 in tray and the opposite monitor on the 1.5 in tray are connected to a 12 volt, DC precision Brailsford pump with tigon tubing and the flow rate for each basin is adjusted to approximately 1 l/m with individual flow meters. In an effort to parallel the LAACC exposure times, the EIC's exposure times were from 24 to 48 hours. Typical voltage drops for the electrets for the 3 in. trays over such exposure periods were approximately 80 to 150 volts, respectively.

The performance of two variations of the new passive EIC surface flux monitors developed by REI were tested at the same time the flow through monitors were in place. The first version of the 960 ml passive monitor (a modified H-chamber) is described in the reference cited above, Stieff, L.R. op cit. (1996) with an electret mounted in the top of the hemisphere and with a tyvek diaphragm covering the approximately 180-cm^2 circular base of the modified H-chamber. In order to minimize contamination of the tyvek diaphragm with phosphogypsum, a paper towel was placed between the monitor and the PG. The paper towel was essentially transparent to the movement of Rn into the monitor. See Figure 2. In the data tables this version of the passive flux monitor is designated as EIC Passive (bare).

The second version of the passive flux monitor is fitted with a circular stainless steel collar which clearly defines the area being monitored and which minimizes the loss of Rn between the edges of the monitor and the surface of the phosphogypsum or the ground when deployed in the field. The results in the data tables from this version are designated EIC Passive(collars).. During the measurement period 10/05/96 to 3/26/97 six sets of "bare" measurements were made and five sets of "collar" measurements were made. During this same period Lab A and Lab B deployed LAACC monitors on 10/21-22/1996 and 2/ 18-20/1997. The measurements made on 10/21-23/1996 , also included a full set of passive "bare" flux measurements.

DATA

Measurements by the three methods were performed between 10/01/96 and 3/26/97. During this time interval LAACC measurements were performed on two occasions (see Table 1), EIC-Flow measurements on seven occasions (see Table 2), and EIC -Passive measurements on six occasions. (see Table 3). Simultaneous measurements by various pairs from the three types of methods were made on a limited number of occasions.

LAACC measurements (Table 1) were performed by two laboratories (designated Lab A and Lab B) for both PG source depths on 10/21-22/96 and for the 1.5-in source on 2/18-20/97. The various results , except for the October, 1996 Lab B results are relatively consistent and are comparable to the EIC results during the overall monitoring period. The October 1996 Lab B results are outside the "consensus" of all the other measurements and appear to be outlying values. The average (and range) of the Rn flux values reported by the LAACC measurements (with the October Lab B results excluded) were 0.34 (0.28-0.41) pCi m^{-2} s^{-1} for the 1.5-in source and 0.69 (0.67-0.73) pCi m^{-2} s^{-1} for the 3.0-in source.

From Table 2 it can be seen that the over all average and the range of the seven EIC flow measurements made on the 1.5-in source trays are 0.31 and (0.26-0.36), respectively. The six flow-through measurements for the 0.3-in trays over the period October, 1996 and March, 1997 are 0.74 and (0.65-0.82).

In evaluating the flow-through method it is important to recognize that two corrections must be made in order to obtain the net voltage drop on the electrets due to the Rn surface flux attributable to the phosphogypsum.. First, it is necessary to correct for the Rn concentration in the ambient air that is continuously pumped through the inverted basins. The ambient atmospheric Rn concentration is obtained using standard 960 ml H-chambers placed at the intake of the 12 volt DC pumps and is approximately 0.3 pCi/l.. The background gamma correction is critical because of the somewhat higher gamma activity associated with the radioactive phosphogypsum and the fact that the H-chambers in the plastic basins are placed directly on the phosphogypsum.. The background gamma activity is measured using standard 960 ml H- chambers similar to those used in the plastic basins. The H-chambers are sealed in Rn-proof, mylar bags and placed on the surface of the phosphogypsum in the same position as the flux monitors. Because the electret voltage drops of the gamma monitors are significantly less than the flux monitors, the exposures are usually from four to six days. The EIC measured gamma backgrounds for the 1.5-and 3.0-in trays were 20.75 and 23.5 uR/h, respectively. From Tables 1 and 2 it can be seen that the 2/18-20/97 control measurements made on the control trays for both the flow through and the LAACC measurements are in good agreement. The Rn surface flux for values the control trays are essentially zero. The very low values for the surface flux measurements obtained on the controls suggests that the corrections for ambient atmospheric Rn and background gamma are close to the proper values

The data for the two passive surface flux methods, "bare" and "collar" are given in Table 3. It can be seen that the over all averages for the 3.0-in trays , 0.52 and 0.76 , and the averages for the 1.5-in trays, 0.35 and 0.50, are not in as good agreement with each other as are the flow through and LAACC averages. A possible explanation for these differences is suggested in the Discussion, below.

In calculating the net electret voltage drop for the two passive EIC flux methods it is also necessary to correct for the PG gamma background. The gamma background is measured with modified H-chambers identical to the passive flux monitors. The background monitors are placed on the surface of the PG with a mylar bag replacing the paper towel between the PG and the monitor. Rn from the PG is unable to enter the H-chamber with the result that drop in voltage of the electret is due predominantly to the background . In general, the 24 hour voltage drops due to the background are relatively small and as a result the background exposure are typically from four to six days.

DISCUSSION

It can be seen from the data tables that the experimental design for the three-way comparison of the flux methods is very imbalanced with only a limited number of simultaneous comparisons. The comparisons are not as powerful as they would have been for a balanced design with simultaneous measurements. However, the EIC-flow data and the Lab a LAACC data suggest that the Rn fluxes from the test-tray sources are relatively constant with time. Thus the overall averages reported by the several methods provide useful preliminary comparisons, even in the absence of simultaneous measurements.

The EIC flow-through measurements were performed on seven occasions between 10/05/96 and 3/26/97, usually for both source depths (see Table 2); these measurements were simultaneous with LAACC measurements for the 1.5-in trays on 2/18-20/97. The ratios of the EIC flow-through results to the LAACC results are presented in Table 4. The ratio of the single simultaneous measurement was 0.94 and the ratio of the overall averages were 0.91 for the 1.5-in source, 1.06 for the 3.0-in source and 0.98 averaged over the two source depths. Thus the two methods were in good agreement with each other, especially when considering systematic errors of approximately 10% associated with the EIC flow-through method and 10 to 20% associated with the LAACC method.

From Table 4 it can be seen that the overall averages for the passive surface flux measurements (bare) for the 1.5- and 3.0-in trays are 0.35 and 0.52 pCi m^{-2} s^{-1} , respectively. These averages can be compared to the overall averages for the 1.5- and 3.0-in trays using the LAACC method of 0.34 and 0.69 pCi m^{-2} s^{-1}, respectively. For this case, the ratio of the passive(bare)/ LAACC overall averages for both the 1.5- and 3.0-in trays are 1.02 and 0.75, respectively. It would appear that at a flux level of 0.35 pCi m^{-2} s^{-1} there was essentially no loss of Rn around the edge of the monitors. However, at the higher flux level associated with the 3.0-in trays, the overall ratio of 0.75 for the "bare" monitors compared to the average LAACC results does suggest that Rn loss around the edges of the monitors may have been significant.

Table 4 shows that the overall averages for the passive(collars) monitors for the 1.5- and 3.0-in trays are 0.50 and 0.76 pCi m^{-2} s^{-1}, respectively compared to the LAACC overall averages of 0.34 and 0.69 pCi m^{-2} s^{-1} ,respectively. For these passive monitors, the ratio of the overall averages for the passive(collar)/LAACC fluxes are 1.45 and 1.10.. In lieu of specific recalibrations of the monitors with collars, these preliminary flux calculations have been made using the original calibration coefficients developed for the "bare" passive flux monitors . It does not seem unreasonable that the collar might have result in somewhat more Rn entering the monitors per unit time if the collars performed as designed. It may be significant that the overall average values for the passive (collar) /flow through ratio, 0.76/0.74, is 1.02.

CONCLUSIONS

A preliminary comparison of the Rn surface flux measurements in the range of 0.3 to 1.0 pCi m^{-2} s^{-1} using LAACC, flow through and passive EIC methods suggest that all three methods are in reasonable agreement with each other. The best overall agreement for both the 3.0-and 1.5-in phosphogypsum trays is between the LAACC and flow through methods. The passive (bare) method is systematically low for the 3.0-in trays and the passive(collar) method is systematically high for both the 1.5 and 3.0-in trays when compared to the LAACC results.

This preliminary comparison of the three different flux methods strongly suggests that a balanced, carefully designed experiment would provide a much stronger basis for evaluating the limitations and strengths of the different methods.

The flow-through EIC method has the advantage of being a method that is based on first principles and potentially could become the reference method for Rn surface flux measurements.

Both the passive and flow through EIC methods are capable of long term integrated surface flux measurements and thus have the potential for making possible the measurement of very low Rn surface flux rates. This capability could become a useful research tool.

Additional comparative measurements should help resolve the optimum design for the passive flux monitor.

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Table 1. Summary of Radon Surface Flux Measurements made on Test Phosphogypsum Flux Trays using Large Area Activated Charcoal Canisters (LAACC)

PG. Tray		LAACC Lab A	LAACC Lab A	LAACC Lab B	LAACC Lab B
Depth in		10/21-22/96 pCi $m^{-2}s^{-1}$	2/18-20/97 pCi $m^{-2}s^{-1}$	10/21-22/96	2/18-20/97
3.0-1		0.73		0.21	
3.0-2		0.67		0.2	
3.0-3		0.68		0.31	
	Avg.	0.693		0.24	
1.5-1		0.34	0.366, 0.391	0.13	0.350 0.267
1.5-2		0.34	0.386, 0.368	0.17	0.325, 0.270
1.5-3		0.36	0.364, 0.408	0.17	0.278, 0.298
	Avg.	0.347	0.379	0.157	0.298
Control					
3.0-0			0.007-0.003		
1.5-0			0.004-0.001		
1.5-0			0.006-0.008		

Note: A new batch of charcoal was used by Lab B for the 2/18-20/1997 LAACC measurements.

Table 2. Summary of the Radon Surface Flux Measurements Made on the Ona Phosphogypsum Trays Using Flow Through Methods.

PG. Tray Depth in		Flow Through 10/5-7/96 pCi $m^{-2}s^{-1}$	Flow Through 1/14-16/97 pCi $m^{-2}s^{-1}$	Flow Through 2/7-10/97 pCi $m^{-2}s^{-1}$	Flow Through 2/10-12/97 pCi $m^{-2}s^{-1}$	Flow Through 2/12-14/97 pCi $m^{-2}s^{-1}$	Flow Through 2/18-20/97 pCi $m^{-2}s^{-1}$	Flow Through 3/25-26/97 pCi $m^{-2}s^{-1}$	Over All Avg
3.0-1		0.882	0.79	0.738	0.745	0.725		0.567	
3.0-2		0.808	0.751	0.556	0.663	0.686		0.640	
3.0-3		0.762	0.811	0.662	0.797	0.893		0.79	
	Avg.	0.817	0.784	0.652	0.735	0.768		0.666	0.737
1.5.-1		0.290	0.382	0.318	0.293	0.382	0.345	0.258	
1.5.2		0.197	0.356	0.271	0.325	0.354	0.307	0.374	
1.5-3		0.298	0.278	0.262	0.283	0.344	0.300	0,322	
	Avg.	0.262	0.339	0.284	0.300	0.36	0.317	0.318	0.311
Control									
3.0-0							0.10		
1.5-0							0.00		
1.5-0							0.001		

Note: The background gamma values used in the calculations, 23.5 and 20.75 uR/h, are based on data from inverted H-chambers placed directly on the phosphogypsum. These chambesr were sealed in Rn resistant mylar bags. The H-chambers inside the plastic basins were also inverted to minimize condensation on the electrets.

Table 3. Summary of Radon Surface Flux Measurements Made on the Ona Phosphogypsum Flux Trays Using Passive Electret Ion Chamber Techniques,Oct.96-Mar.97.

PG Tray	Passivr Surface Flux Modified EIC H- Chambers (bare)							Passive Surface Flux Modified EIC H-Chambers (collars)					
Depth in.	10/1-2/96 pCi $m^{-2}s^{-1}$	10/21-23/96 pCi $m^{-2}s^{-1}$	1/13-14/97 pCi $m^{-2}s^{-1}$	2/7-10/97 pCi $m^{-2}s^{-1}$	2/12-14/97 pCi $m^{-2}s^{-1}$	3/25-26/97 pCi $m^{-2}s^{-1}$	Over All Avg.	10/1-2/96 pCi $m^{-2}s^{-1}$	1/13-14/97 pCi $m^{-2}s^{-1}$	2/7-10/97 pCi $m^{-2}s^{-1}$	2/12-14/97 pCi $m^{-2}s^{-1}$	3/25-26/97 pCi $m^{-2}s^{-1}$	Over All Avg
3.0-1	0.655	0.396	0.501	0.528	0.599	0.547		0.771	0.694	0.907	0.649	0.893	
3.0-2	0.530	0.403	0.453	0.472	0.525	0.542		0.866	0.717	0.745	0.714	0.730	
3.0-3	0.620	0.515		0.522	0.533	0.482		0.824	0.756	0.697	0.896	0.587	
Avg.	0.602	0.438	0.477	0.507	0.552	0.524	0.517	0.820	0.722	0.782	0.753	0.737	0.763
1.5-1	0.247	0.278	0.458	0.2735	0.294	0.452		0.373	0.506	0.682	0.458	0.438	
1.5-2	0.325	0.300		0.359	0.423	0.382		0.403				0.409	
1.5-3	0.282	0.266		0.305	0.366	0.387		0.442				0.437	
Avg.	0.285	0.281	0.458	0.313	0.361	0.407	0.351	0.406	0.506	0.682	0.458	0.428	0.496
Control													
1.5-0		0.011											
1.5-0		0.015											

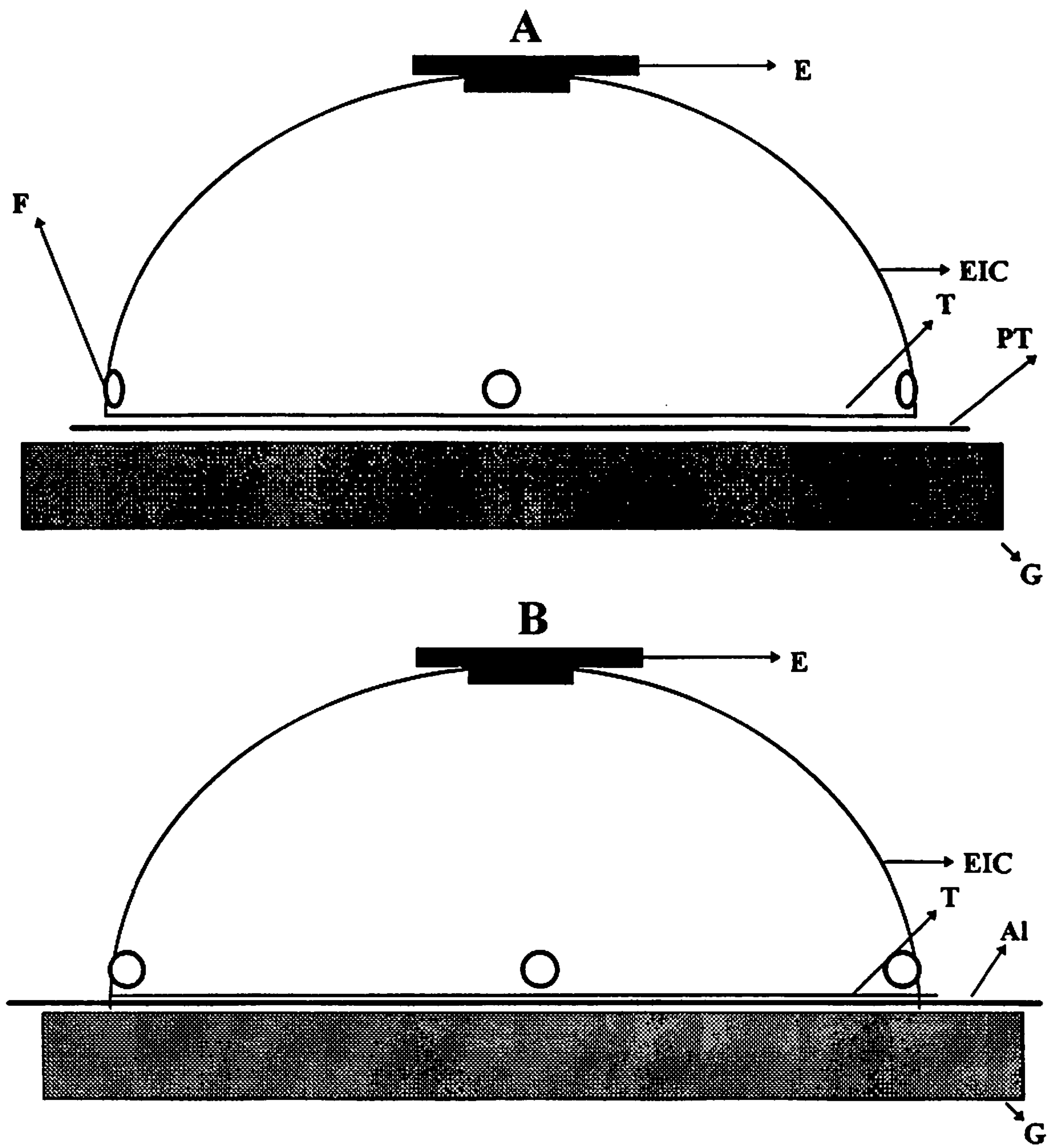
Note : All measurements made with modified H-chambers and with ST electrets
Collars refers to stainless steel collars attached to the base of the chamber;

Table 4. Comparision of Electret Ion Chamber (EIC) and Large Area Activated Charcoal Canisters (LAACC) using Phosphogypsum Test Trays as the Radon Flux Source.

Comparasion	Rn Flux,pCi m-2 s-1 LAACC*	EIC	Ratio EIC /LAACC
EIC Flow Through			
Simultaneous, 2/18-20/97			
1.5 -in	0.34	0.32	0.94
3.0 -in	None	None	---
Over All, 10 05/96-3/26/97			
1.5 -in	0.34(2)	0.31(7)	0.91
3.0 -in	0.69(1)	0.74(6)	1.06
			0.98
EIC Passive (bare)			
Simultaneous,10/21-22/96			
1.5 -in	0.35	0.28	0.81
3.0 -in	0.69	0.44	0.63
			0.72
Over All, 10 05/96-3/26/97			
1.5 -in	0.34	0.35	1.02
3.0 -in	0.69	0.52	0.75
			0.88
EIC Passive (collar)			
Simultaneous --none	----	---	----
Over All, 10 05/96-3/26/97			
1.5 -in	0.34(2)	0.50 (5)	1.45
3.0 -in	0.69(1)	0.76 (5)	1.10
			1.27

Notes:

LAACC values are Lab A for 10/96 and average of results reported for Lab A and B for 2/97.
Values in ( ) indicate no. of measurement episodes in overall average.
Each measureement episode usually involved three replicate trays for each source depth.



E : Electret
G : Ground or tailings
PT : Paper Towel to allow radon to pass
T : Tyvek Window
EIC : Electret Ion Chamber
Al : Al sheet to stop radon
F : Filtered openings

NOTE
The Chamber A allows radon
The Chamber B stops radon

Fig.1 Schematic Drawing of the Passive EIC Radon Surface Flux Monitor