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RADEX: AN ACTIVE MONITOR FOR CONTINUOUS MEASUREMENT
OF ^{222}Rn FLUX IN SOIL

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May 1984

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ABSTRACT

A radon exhalation monitor, RADEX, was developed at the Environmental Measurements Laboratory. RADEX is an active radon monitor that continuously samples radon gas emanating from the soil. A hemispherical accumulator is placed on the soil and radon gas is drawn into the RADEX counter at a low, controlled flow rate. As ^{222}Rn decays, the resulting radon daughters are focused by an electrostatic field, deposited directly onto a semiconductor detector, and counted. RADEX's ability to detect rapid changes in radon exhalation is made possible by analyzing RaA alphas with fast decay times. The pulses produced by the radon daughters are integrated hourly and are recorded. This system can operate unattended for 1 week, at which time a desiccant column must be changed. Thus, RADEX allows one to observe and to record continuously the radon flux from the soil.

INTRODUCTION

To accurately measure radon flux from the earth requires a system that will not disturb the natural terrain. Unfortunately, at present no system is available that totally meets this requirement. To date, there are two approaches available. The first is a loosely-coupled arrangement, i.e., when the collecting device is lightly sitting on the soil, which has the advantage of minimal disturbance to the natural terrain. However, it suffers from serious dilution problems when the ambient air mixes with the radon gas emanating from the soil. This results in a lower and unpredictably variable measurement of the actual radon flux being released from the ground. The second is a tightly coupled arrangement, which involves sampling on or slightly into the soil and, as such, affects the natural interaction that occurs at the soil/air interface. Isolated from the atmosphere and acting like a blanket, the sampling area becomes immune to normal environmental phenomena such as wind, rain, snow and temperature variations. Thus, because the normal interaction between the emanated radon and the surrounding air is inhibited, the measurement is often biased towards the higher values. However, since the tightly-coupled measurements are immune to the variability encountered with the loosely-coupled sampling, the former determinations yield a more accurate assessment of the actual exhalation rate of the radon gas.

We have used the later method to develop a continuous monitoring instrument, the RADEX, for determining radon flux. The RADEX monitoring system is discussed in detail in this report.

INSTRUMENTATION

Sampling methods presently used for measuring radon exhalation from the earth are generally based on passive detection devices, such as: track etch film, charcoal canisters, and accumulators (Fleischer, 1980; Countess, 1976; Kraner et al., 1964). For the design of RADEX, an active accumulator is used for measuring the radon flux. Figure 1 shows the hemispherical accumulator, which has a diameter of 23 cm and a volume of 7 L. The accumulator is placed onto a rim that has been anchored into the ground to a depth of at least 1 to 2 cm. Attached to the accumulator is a length of Tygon tubing through which air and the radon gas is removed at a constant flow rate. The combination of a cold trap (1.8 L in volume) and a filter tube (Balston) will remove most of the moisture from the sample. At this point in the loop, the pump is located. A continuous and constant flow rate of $200 \text{ mL} \cdot \text{min}^{-1}$ is regulated by a pressure switch and associated circuitry. Figure 2 shows the pump with its control circuit which is described in detail in EML-389 (Latner et al., 1981). After the pump, a desiccant column removes the remaining moisture from the sample before it enters the counting chamber. In addition, a 7.6 cm diameter glass fiber filter is used to remove the radon daughters and, thus, only radon enters the counting chamber.

The counting chamber (shown in Figure 3) is cylindrical in shape, 24.5 cm in diameter, 21.3 cm in height and has a volume of 10 L. The aluminum counting chamber is lined with Teflon, which has aluminum foil attached to the inner surface. The foil is insulated from the cylinder housing, allowing a high potential to be applied to the foil without electrical breakdown occurring. A positive 3500 V is applied to the foil, establishing a static electric field. The electric field acts to focus and direct the newly formed radon daughters towards the detector (Wrenn et al., 1975; Negro and Watnick, 1978). The ionized molecules then deposit directly onto the active area of the detector. A Surface Barrier Solid State Detector, 300 cm² in area and ring mounted, is used in the RADEX monitor. The detector is mounted in a Teflon holder, which insulates it from the chamber's electric field potential. An 'O' ring is used to hermetically seal this assembly to the counting chamber and, thus, insure proper sample flow from input to output ports. The ring mounted detector, by virtue of its minimum mass housing, maximizes the ratio of active detector area to inactive area, thus, offering the greatest possible sensitivity.

Figure 4 shows the circuitry, which can be divided into four main sections. These are: the air flow system, the power supplies, the charge-sensitive preamplifier, and the window counter/comparator circuits.

The signal developed by the surface barrier detector is coupled to a compact charge-sensitive preamplifier designed around a low power, triple operational amplifier chip (L144). This circuitry has been described in detail previously (Watnick and Latner, 1982). An emitter follower acts as a buffer between the preamplifier and an associated multichannel analyzer that processes the analog signals. Typical waveforms for RaA (²¹⁸Po, 6 MeV) and RaC' (²¹⁴Po 7.8 MeV) alpha pulses are shown in Figure 5.

The preamplifier output is also routed to a window counter/comparator circuit. This unique circuit design uses a very efficient arrangement of the four sections of a single chip, low power, quad comparator (L166). The input stage utilizes two of the comparators to form upper- and lower-level discriminators. A RaA (²¹⁸Po) alpha signal will cause the normally 'off' output of the lower-level discriminator to turn on. When this occurs, a ramp voltage is generated at its output at the junction of capacitor (C) and diode (D). For a typical 80 μsec wide input pulse, the ramp voltage will reach a peak amplitude of 1.7 V before the pulse is terminated. At levels above 1 V, the lower-level comparator (L.C.) goes on, and delivers an output pulse which represents an RaA (²¹⁸Po) event. An RaC' (²¹⁴Po) alpha signal activates both the lower- and upper-level discriminators. The upper-level discriminator then places the diode near ground potential. This effectively prevents the ramp from rising (see Figure 5), and the lower-level discriminator from giving an output. The upper-level comparator (U.C.), however, because of its biasing arrangement, now turns on as its input drops to zero. The pulse output from this upper-level comparator represents an RaC' (²¹⁴Po) event. Thus, in addition to the analog signals routed to the multichannel analyzer, digital signals are available for recording RaA (²¹⁸Po) and RaC' (²¹⁴Po) events on automatic data logging equipment such as EML's CLEVER or SMART COUNTERS (Polito and Negro, 1979).

As in the analog circuitry, the digital outputs are buffered with emitter followers using three of the five transistor arrays packaged in a single integrated circuit (CA3046).

Power for the preamplifier and window counter circuits is supplied by a 9 V battery to a group of regulators whose total current drain is 1 mA, allowing the circuits to operate continuously for 2 weeks. The bias supply for the detector consists of two-22 1/2 V miniature batteries that will operate for as long as their shelf life. To allow extended operation, where a.c. power is available, the 9 V battery can be replaced by an external power supply.

Physically, the circuits are mounted on a printed circuit board packaged in a plastic box whose dimensions are 4x10x3 cm. Both the circuit and bias batteries are housed within this enclosure.

The high voltage power supply used to generate the static field is a d.c.-to - d.c. converter type. It supplies an output of 3500 V for an input of 5 V with a 10 mA drain (see Figure 6). Transformer (T1) with transistor (TR1) form a blocking oscillator that allows a voltage stepup to about 900 V. A voltage quadrupler is then used to increase the output to 3500 V d.c. This technique is quite effective in this application because of the insignificant drain on the high voltage power supply. A portion of the output is sampled and appears at one input of IC1, a low current comparator whose reference input is tied to a stable, temperature-compensated voltage. IC2, a CD4007 dual complementary pair, buffers and inverts the comparator output. This signal then controls the base drive signal for TR1, the blocking oscillator, supplying the necessary correction to hold the output constant.

Figure 7 shows the low voltage power supply, which uses micropower d.c. regulators and voltage converters. The input voltage from the nominal 9 V battery is applied to IC1 and IC2. IC1 and IC2 then supply a +5 V output as long as the input battery voltage remains above 5.2 V. In order to achieve a high degree of isolation, one of the +5 V regulator circuits is used exclusively to power the high-voltage circuit. The remaining +5 V regulator powers two monolithic voltage converters, IC3 and IC4. IC3 functions as a voltage doubler, producing the positive 9 V and, in addition, supplies a negative input for IC4. IC4, in turn generates the negative 9 V line. Since the loading of these supplies remains relatively constant, the output voltages are quite stable.

CALIBRATION

Calibration of the RADEX monitor, a necessary step prior to initial use of the instrument, involves setting the window counter discriminator levels, as well as the energy integration regions on the associated multichannel analyzer. The calibration procedure requires that the monitor be placed in an

atmosphere of ^{222}Rn activity maintained at a constant level. We chose 32 pCi/L. At the same time, information on system resolution, stability, and collection efficiency can be obtained. During calibration, a scope display of the preamplifier alpha pulses (Figure 5) shows 2.3 V for RaA (^{218}Po) and 2.9 V for RaC' (^{214}Po).

Discriminator levels can then be set to give appropriate logic outputs for digital counting. These pulses are also routed to the multichannel analyzer, allowing a determination of the resolution of the RADEX monitor. The 6 MeV RaA (^{218}Po) peak shows a resolution of 120 keV FWHM (full width at half-maximum height). This level of resolution allows for ample separation between it and the 7.8 MeV RaC' (^{214}Po) peak, and includes a significant margin to compensate for minor signal drifts that may occur during extended counting periods.

A good approximation of the RADEX monitor's collection efficiency for RaA (^{218}Po) and RaC' (^{214}Po) alphas can be determined using the following equation (Negro and Watnick, 1978), which is valid for constant radon concentrations:

$$C_{\text{Rn}} = \frac{X}{Z'} \frac{0.45}{E V Ff}$$

where

- C_{Rn} = Radon concentration in pCi.L $^{-1}$ (31.5)
- X = Net observed counts
- E = Counting efficiency (0.45)
- V = Decay volume in L (10)
- Ff = Collection efficiency for RaA (^{218}Po) and RaC' (^{214}Po)
- Z' = Integral counting factor. In the steady state it reduces to twice the counting time or $2 \times 33.3 = 66.6$.

The observed counts were averaged for 16 counting periods of 33.3 min each. The following results were obtained:

$$31.5 = \frac{9750.31}{66.6} \frac{0.45}{0.45 \times 10 \times Ff}$$

$$\text{RaA } (^{218}\text{Po}) \text{ and RaC' } (^{214}\text{Po}) \quad Ff = 46.5\%$$

$$31.5 = \frac{4651.75}{33.3} \frac{0.45}{0.45 \times 10 \times Ff}$$

$$\text{RaA } (^{218}\text{Po}) \text{ Ff} = 44.3\%$$

$$31.5 = \frac{5099.94}{33.3} \frac{0.45}{0.45 \times 10 \times \text{Ff}}$$

$$\text{RaC'} (^{214}\text{Po}) \text{ Ff} = 48.6\%$$

Clearly, the collection efficiency of this instrument is high. The small losses can be attributed to the RaA (^{218}Po) ions being deposited onto the dead area and the printed circuit material of the surface barrier detector (Negro and Watnick, 1978). Other losses may be caused by neutral or negatively charged ions that will not be deposited on the detector.

The sensitivity calibration of the RADEX monitor requires a comparison with an independent measurement method, such as the use of charcoal canisters. Pulverized, homogenously mixed soil that had been spiked with various amounts of ^{226}Ra was put into containers. These containers were plastic lined wooden boxes with dimensions of 46 cm on each side by 28 cm high. The hemispherical sampling unit of the RADEX monitor, as well as two charcoal canisters, were placed upon the prepared soil and remained there for periods of time ranging from 24 to 72 hr. The charcoal canisters were then removed, analyzed in a gamma scintillation counter, and a radon flux value was calculated. Because of the uniform nature of the spiked soil bed, the RADEX sampler was exposed to the same flux levels as the charcoal canisters. An accurate comparison can then be made by relating the observed count of the RADEX monitor to the radon flux determined with the charcoal canisters. For improved accuracy, the measurements were repeated many times at different concentrations, and the results averaged. A plot of RADEX cpm vs. radon flux (Figure 8) was made and served as the calibration curve of the RADEX monitor. The sensitivity derived from the calibration curve is 1.0 cpm which is equal to $0.984 \text{ aCi}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$.

FIELD TESTS

After completing all laboratory tests and calibrations, the RADEX monitor underwent field testing at our remote station at Chester, New Jersey to uncover any problems that might be caused by the effects of temperature, humidity, and fluctuations in line voltage. For these tests, the sampling hemisphere was placed on the soil, while the remainder of the RADEX system was located in a nearby trailer (see Figures 9A and 9B). Tygon tubing was used for sample transport between outside and inside. Figure 10 shows a 10-day plot of hourly readings taken with the RADEX monitor at Chester, New Jersey.

In order to verify the measurement accuracy of the RADEX monitor under field conditions, sampling was performed for 6 hr using an accumulator that operated on a set schedule. Samples from the accumulator were taken by an evacuated gas collecting cylinder after 15 and 45 min, at which time the accumulator was removed from the soil for 15 min. This procedure was repeated four times. Over the same time periods, gas samples were also removed (again using an evacuated cylinder) from the sampling hemisphere of the RADEX monitor. Simultaneously, the RADEX monitor was in operation throughout these tests. Table 1 shows the results of these tests, comparing the RADEX monitor, the gas sample from the monitor and the accumulator. It should be noted that the sample reaching the RADEX detector and that which is drawn from the sample head are identical and lend themselves to a valid intercalibration of the response of the detector system. However, comparisons between these and the accumulator are less certain. The accumulator is obviously not sampling the same area as the RADEX, and may be exposed to different activity levels. In addition, the accumulator is a passive device, while the RADEX is active. Thus, while the results of the accumulator measurements are included in Table 1, further experiments are necessary to establish the relationship between these two sampling methods. With this aside, agreement is good except for those periods during or following a rainfall where the RADEX reads higher. This difference arises from the fact that the hemispheric sampler always remains on the ground and prevents the soil beneath it from getting wet, and the other sampling methods used do allow the soil to get wet. The diffusion rate of radon gas is different for wet and dry soil.

CONCLUSION

The RADEX monitor has been in continuous operation for a period of over 6 months, with tests made on a regular basis. It has shown itself to be a useful, accurate and reliable instrument that can be used to examine rapid changes of radon exhalation from the earth and to make continuous measurements over any repeated time periods.

As a cautionary note, it is possible that the measurements may be biased towards a higher value because of two factors. The first is the small negative pressure applied to the area being sampled. The second is the effect of the tested area being covered by the hemispherical sampler and, thus, being isolated from the normally occurring climatic variations. Future plans have been formulated to further study and improve the RADEX monitor. The effect of air flow rate on exhalation factor will be investigated.

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TABLE 1

CHESTER, NJ INTERCOMPARISON FLUX MEASUREMENTS ($\mu\text{Ci}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$)

Date	Weather Condition	RADEX Monitor	Gas Sample from RADEX Monitor	Accumulator
6/29/83	Clear	180.23 $\pm$ 20.4	189 $\pm$ 43	N.D.
7/11/83	Clear	65.58 $\pm$ 4.04	63 $\pm$ 7.9	67.6 $\pm$ 16.1
7/13/83	Clear	43.31 $\pm$ 4.88	53.6 $\pm$ 13	N.D.
7/25/83	Rain	99.9 $\pm$ 1.03	99.6 $\pm$ 31.3	75.24 $\pm$ 48.63
7/27/83	Clear	90.69 $\pm$ 3.3	85 $\pm$ 10.2	63.1 $\pm$ 15.7
8/17/83	Clear	147 $\pm$ 10.55	134.8 $\pm$ 49.75	149.2 $\pm$ 60.2
8/31/83	Rain	386 $\pm$ 11.23	417 $\pm$ 49	131 $\pm$ 52

N.D. = samples lost in preparation.

Figure 1. RADEX monitor.

Figure 2. Constant flow pump system.

Figure 3. Detector chamber.

Figure 4. Circuit diagram.

Figure 5a. Analog signal for RaA and RaC'.

Figure 5b. Ramp voltage waveforms.

Figure 6. High voltage circuit diagram.

Figure 7. Low voltage circuit diagram.

Figure 8. Calibration curve of the RADEX monitor.

Figure 9a. RADEX accumulator at Chester, NJ site.

Figure 9b. RADEX monitor in trailer at Chester, NJ site.

Figure 10. Ten day plot of the RADEX monitor at Chester, NJ site.