A Soil-Gas Emanation Measurement System Used for Radon-222

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ABSTRACT

The transfer of various gases through the soil-atmosphere interface is of interest to agronomists, meteorologists, biologists and many others. Such gases include carbon dioxide, water vapor and radon. A system applicable to measurements of the emanation of gases has been developed and used to investigate the emanation of radon-222. The instrumentation was designed so that the variation of soil gas emanation with time, wind speed, soil moisture, soil type, and geographical location could be studied. The system has measured a net transport through the earth-atmosphere interface of 1.1 ± 0.6 atoms of radon-222 per square centimeter per second (mean of 27 means, eight samples each, $\pm$ standard deviation of 27 means). For a set of eight samples collected on a given site, the coefficient of variation was 0.3 to 0.4. The analysis of a single sample provided the amount of radon-222 collected with an error of about 5 per cent.

The system is completely portable, including the power supply, and can be carried from a vehicle by two men. It was designed to minimize disturbance of the site at all times, providing air flow and pressure within the collecting apparatus comparable to those in nature during sampling. Sampling sites can be chosen as desired, and all equipment can be removed from the site when sampling is completed. During a sampling trip to the Rocky Mountains, the equipment was transported 3000 miles without any breakage. Throughout an experiment on the diurnal variation of radon emanation, two duplicate systems were operated 12 hours each without difficulty.

1. Introduction

The movement of soil-gas through the earth-atmosphere interface has been of interest to agronomists, meteorologists, biologists, water conservationists and perhaps others for many years. Studies of the movement of water vapor from soil into the atmosphere have been conducted extensively, and the movements of other gases such as carbon dioxide and radon-222 also have been investigated. While our research has been concerned with the transport of radon-222 through the interface, the methods and some of the equipment used are applicable in studying the exhalation of other gases. Radon-222 is a radioactive noble gas which is the first decay product of radium-226 in the uranium-238 decay series. The ubiquity of radium-226 in greatly varying amounts is well established, and this isotope and its daughters have been investigated extensively.

Several investigators of radon-222 emanation (Wright and Smith, 1915; Wilkening and Hand, 1960) have used a disc or washer-shaped collecting device with an outside diameter of about 25 cm containing a 1-cm diameter hole at the center. The disk was placed 1 to 2 cm above the soil surface, and the sample was removed through a hose connected to the hole at the center. Atmospheric air swept the soil surface beneath the disc, the radon being collected on adsorbent material for analysis. This use of the disc resulted in a variable flow rate across the soil surface and required determination of the concentration of radon-222 in atmospheric air near the soil surface and making a subsequent correction. The device which we developed for the collection of radon-222 is substantially different and is believed to be an improvement over those previously used. It is with the hope that our experience in this development may be useful to others interested in emanation of gas through the earth-atmosphere interface that this paper is presented.

2. Experimental development

From the beginning of our work we believed that the collection device and system should be designed to maintain conditions at the sampling site which simulated as nearly as possible those existing in the surrounding area during sampling. This requirement was extended to include preservation of antecedent environmental conditions of the sampling site as well. With this general requirement in mind, a device to collect the gas being transferred into the atmosphere from the soil was designed. Also a collection system was developed to separate the captured gas from environmental

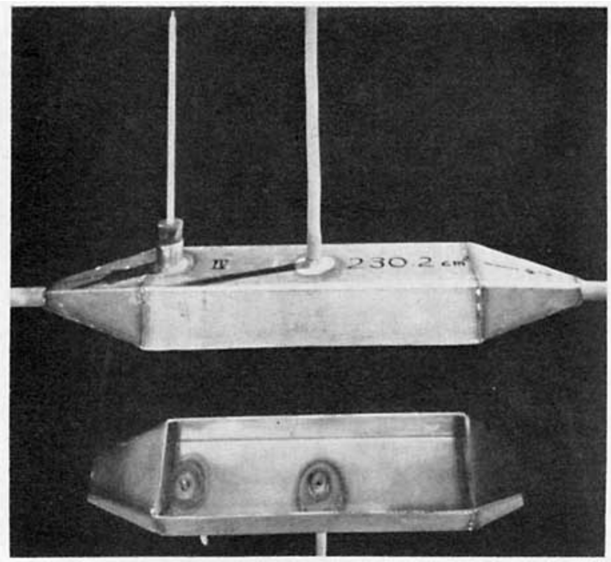

FIG. 1. Collector for sampling radon-222 emanation at the earth-atmosphere interface. Opening in collector is shown by reflection.

air for transportation to the laboratory where samples were processed.

The collecting device was designed to fulfill several requirements. First, minimal disturbance to the soil during the placement and operation of the collector was found to be indispensable. Also, the collector was to be constructed so that it could be sealed to the soil surface, thus prohibiting both loss of gases from the collector and the admission of external air, the radon-222 content of which varies. Since we were attempting to perform measurements in a natural environment, flow within the collector was to resemble air movements in the free atmosphere. While providing such natural flow, the configuration of the collector had to be such that emanation could be reported in units of flux density (i.e., quantity of gas emanating per unit area per unit time); thus, the area of soil being sampled had to be well defined by the collector. Finally, the device had to be durable, inexpensive, and easily fabricated.

The collector developed is shown in Fig. 1, in which the open bottom of the collector is visible by reflection. This opening circumscribed an area of soil approximately 30.5 cm by 7.6 cm. Early models of the collector had sharpened edges about 2.5 cm deep which were pushed into the soil to seal the periphery of the collector to the soil. As undisturbed surface soils in central Illinois are mostly devoid of rocks and gravel, the seal was satisfactory. However, the edges cut slots into the the earth, permitting soil gases having relatively high radon contents to leak into the collector. Such a disturbance resulted in large variation in measurements made only a few centimeters apart. Removing the knife edges from the collectors and sealing them to the soil with caulking compound lowered the variation to a satisfactory level and still afforded an acceptable seal. (With the knife edge seal, the standard deviation for eight samples taken close together and within a one hour interval was as high as 100 per cent of the mean, while with the caulking material seal, a 30 per cent value was attainable on the same sampling sites.) The collector was designed as a straight tunnel in order to permit a reasonably constant flow across the entire surface being sampled.

After selection of a reasonably flat surface the collector was placed with the open face adjacent to the soil and with minimum disturbance to the soil. As part of the consistent effort to maintain pre-existing conditions at the site, the collecting device was carefully placed over the sampling area while the collecting system was in operation, thus ensuring that emanating radon could not accumulate within the system before the beginning of sampling. During the interval of sample collection, the device was not disturbed. At all times, then, operating procedures were such that flow across the soil surface simulated that existing in the unobstructed surrounding environment.

The collector was fabricated from 16-gage sheet stainless steel which was formed and welded. This material provided sufficient stiffness for dimensional stability and, at the same time, satisfied the cost, durability, and ease of fabrication requirements.

The collection system to be used with the collector described above was designed to fulfill the criteria for sampling in the following respects. Sites where samples were to be taken were to be undisturbed for several days before sampling, prohibiting the establishment of fixed installations which would interfere with rainfall, solar heating, and the turbulent structure of winds over the sites. The collection system also had to duplicate as nearly as possible the meteorological conditions over the area being sampled, and trouble-free operation of the system was requisite for the successful accomplishment of uninterrupted sampling. These requirements were met by the use of a completely portable system by means of which flow within the collector could be adjusted to best simulate that in the free atmosphere.

The portable system designed included an alternating current motor-generator; a plywood box containing a pump, tubing, valves, Dewar flasks, radon traps, and associated chemicals; a box for transporting shredded dry ice; and one containing miscellaneous small supplies. The system is represented schematically in Fig. 2 and pictorially in Fig. 3. As mentioned above, the system was set in operation before placement of the collector. During the period between placing the collector and beginning the sample collection (2 or 3 minutes), the pump was run and the flow rate adjusted to give proper flow within the collector. Radon swept from the collector in this interval was removed from the closed-loop system by a "flushing" trap, a standard radon trap. At the initiation of sampling, the three-way valve was rotated 90 degrees to start the collection

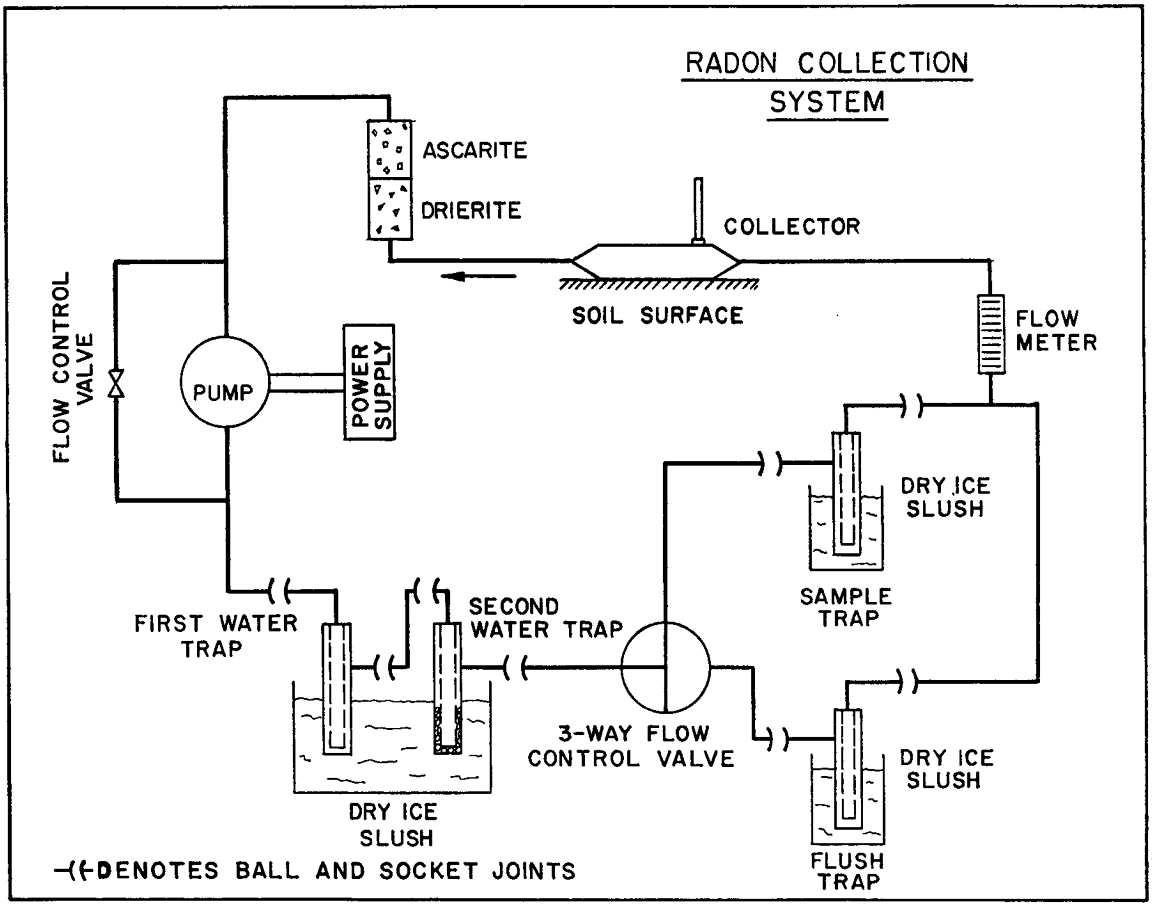

Fig. 2. Schematic representation of the system for collecting radon-222 emanating at the earth-atmosphere interface.

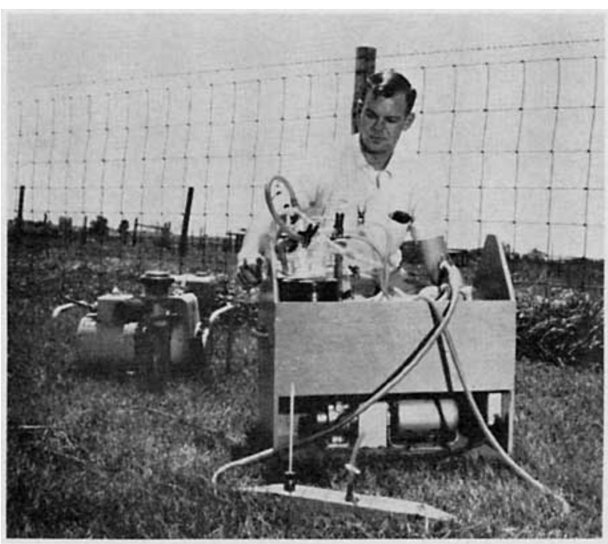

Fig. 3. Portable system for collecting emanating radon-222 in use in the field.

of emanating radon in the sample trap. At the end of the sampling period (usually 300 seconds), a reverse turn of the three-way valve terminated the sample. At all times, then, flow within the collector was continuous.

While it was intuitively reasoned that conditions within the collection device should simulate natural air movements and gustiness, there existed a need to determine the relationship between wind speed across the surface of the soil and the emanation measured. First, we determined the relation between flow rate within the collection system and wind speed within the collecting device measured at 1.25 cm above the soil using an anemometer operating on the directly-heated thermopile principle. A variety of flow rates were used while the concomitant emanation was collected and determined. A graph of the results is shown in Fig. 4 in which the linear relationship, at the wind speeds studied, is apparent. Knowledge that there is a dependency of emanation upon wind speed across the soil surface led to the conclusion that wind speeds existing just above the soil in the surrounding area should be measured and duplicated as closely as possible within the collector during sampling.

Average wind speeds in the free atmosphere were measured at 1.25 cm above ground level using the hot-wire anemometer previously mentioned. Average speeds of 0.03 meter per second and 0.08 meter per second were measured for an "average" day and a "windy" day, respectively. The results of the testing showed that a flow rate of 4 liters per minute in the system would produce within the collecting device a measured velocity of 0.045 meter per second at 1.25 cm above the ground, a speed approximating those occurring naturally most of the time. It should be noted on Fig. 4 that the average for 216 samples taken at the flow rate of 0.045 meter per second does not fall on the dotted line. Also, the emanation measurements reported by Evans, Kraner, and Schroeder (1962) were taken with a wind speed of zero and are not greatly different from our mean of 216 samples. It is concluded that for the very low wind speeds ordinarily observed between the grass blades at 1.25 cm above the ground, there is no demonstrable variation of emanation with variation in wind speed. The left end of the curve in the figure should become approximately horizontal at the very low wind speeds which normally prevail.

Gustiness patterns were observed on the wind speed meter with the anemometer probe at 1.25 cm elevation in the free atmosphere and with the probe at the same elevation inside the collector with a flow rate of 4 liters per minute. The range and frequency of pulsations appeared similar in both cases.

It was decided, as a result of these tests, to adopt a collection flow rate of 4 liters per minute provided this flow could be shown to be satisfactory by a study of its effects on emanation rate over a period of several hours. This was ascertained by placing the collector over a fixed location and sampling continuously for an eight hour period. Samples were collected in a charcoal sampling trap for 55 minutes. Then the flow was diverted into one of the flushing traps for 5 minutes while the sampling trap was removed from the system and replaced with a freshly purged one. This cycle was repeated each hour. Eight-hour tests of this design were carried out on two different days. Data obtained is presented graphically in Fig. 5. While natural conditions prevailing during the two testing periods established substantially different emanation rates, the uniformity of measurements within each test indicates that the removal of radon at the surface while using the adopted flow rate neither caused a depletion of radon from the soil nor permitted it to build up in concentration just below the surface. The former result would be evidenced by a decreasing emanation rate with time and the latter by an increasing rate. Neither situation was observed.

Other precautions against disturbing naturally occurring conditions at the sampling sites were taken also. Operating procedures were such that all equipment was located downwind of the sites, for example. Then, too, the collection system was designed to maintain atmospheric pressure within the collector, accomplished to within 1.6 mm of water. Finally, except for the fact that grease in the three-way valve became more viscid during cold weather, which necessitated its replacement by a system of clamps and hoses, no difficulties with equipment were experienced during the program.

As a further check of the sampling technique the site near Weir Hall, New Mexico Institute of Mining and Technology, where Wilkening and Hand (1960) made measurements of emanation, was visited and sampled for comparison of their results with those of our project, the data for which are given in Table 1 below. The measurements by the two groups of investigators appear to be remarkably consistent. Such consistency is believed to attest to the integrity of both sampling techniques.

TABLE 1. Emanation of radon-222 from soil near Weir Hall, New Mexico Institute of Mining and Technology, Socorro, N. Mex., on 29 July 1963, in 10^{-18} curies cm^{-2} sec^{-1}.

Investigator	Mean	Range: Highest value	Range: Lowest value
Wilkening and Hand	90	126	49
Pearson, Rimbey, and Jones	101.8±30.1 (std. dev.)	135.8	46.3

Radon-222 analysis. The system described above separated radon-222 from atmospheric air by adsorption on charcoal. Investigators who are concerned with the emanation rate of other materials would need to modify the collection system to suit their needs. However, for the benefit of any interested in radon emanation, a brief account will be given of the way the amount of radon collected was determined.

The technique used for measuring quantities of the gas, radon-222, was developed and has been reported by Stehney, Norris, Lucas and Johnston (1955). This method may be described as follows. Air containing a quantity of radon-222 was passed through a dry-ice chilled trap in which the radon was adsorbed on activated coconut charcoal. The charcoal trap containing the sample was brought to the laboratory where the radon sample was desorbed by heating the charcoal to 300 C. The trap was flushed several times, aged helium being used as a carrier, and the radon-helium mixture was pumped into an evacuated counting bottle. The counting bottle, covered at one end by a quartz glass window, had been lined with silver activated zinc sulfide scintillation material. After the transfer the bottle was sealed and allowed to stand about two hours during

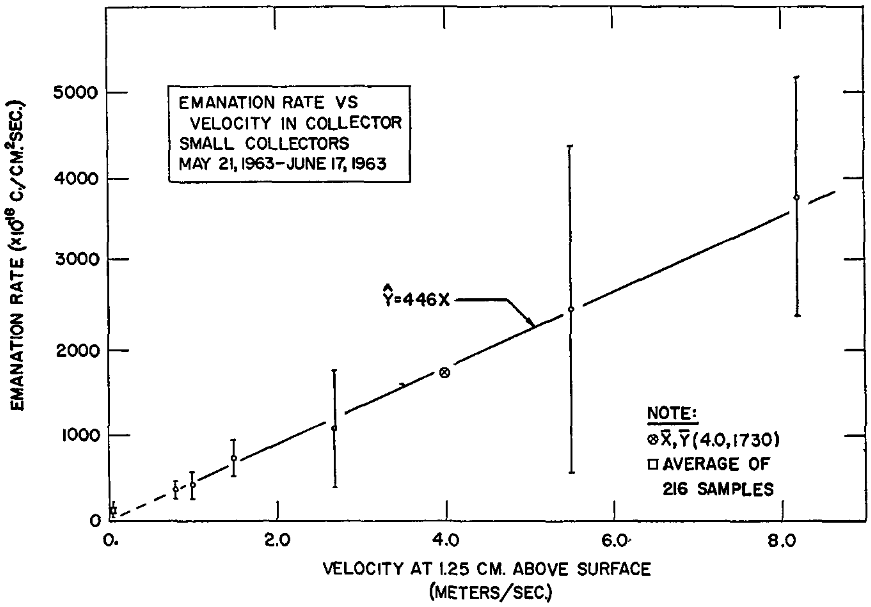

FIG. 4. Emanation rate of radon-222 as a function of wind speed within collector.

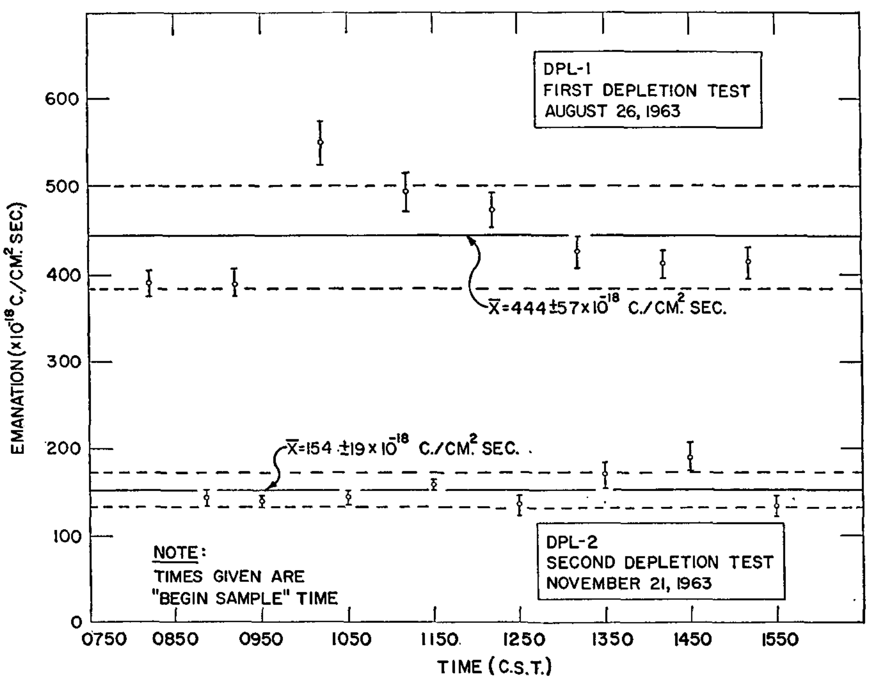

FIG. 5. Results of experiments to determine suitability of adopted flow rate (4 liters per minute) within collector. Upper data from test of 26 August 1963; lower data from that of 21 November 1963. Curves shown are means ± standard deviations for the two tests.

which the short-lived radon decay products established approximate equilibrium with the parent radon. The radon content was determined by placing the counter bottle window in contact with the window of a photomultiplier tube, covering the assembly with a light-tight shield, and counting alpha-scintillations for a measured period of time. Corrections for radon decay during the intervals between collection, transfer, and counting were made. For a typical background counting rate of about 0.1 count per minute, this system has demonstrated an efficiency of 4.85 ± 0.05 (standard deviation) counts per minute per picocurie of radon. The ability of the charcoal to adsorb radon at greater activity and higher flow rates than normally used was tested by placing two charcoal traps in series in the system and measuring the activity retained in each. The average percentage of activity collected in the first trap was 99.38% with a range for four tests of 99.20% to 99.54%.

3. Results of emanation measurements

Using the equipment and methods described above, 216 measurements of emanation were made in Champaign County, Illinois. These samples were taken at nine locations, each including three soil types which cover 85 per cent of the county. The sites were located two each in the four cardinal directions, one at 6.5 km and one at 13 km from the central site, and emanation was investigated for uniformity among sites and soil types. The emanation measured was $140 \pm 73 \times 10^{-18}$ curies per square centimeter per second (standard deviation). On an atomic basis this is equivalent to 1.1 ± 0.6 atoms per square centimeter per second. The standard deviation from the mean rate measured throughout the county was somewhat greater than that measured on the agronomy plots of the University of Illinois because of greater variation of surface roughness and vegetative cover at the former sites. In some locations, quantities of decaying materials from previous years made difficult the location of the soil surface. While the emanation in Champaign County was shown to be essentially uniform (90 per cent confidence), emanation rates three orders of magnitude higher were found in the uranium mining country near Grants, New Mexico, demonstrating the utility of the equipment over a wide range of emanation levels.

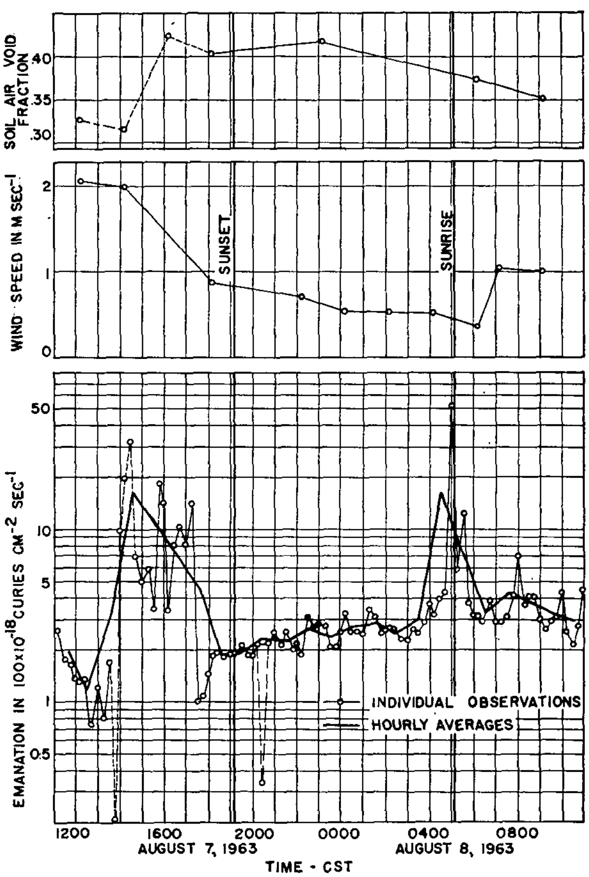

FIG. 6. Results of measurements of diurnal variation of radon-222 emanation and concomitant variations in wind speed at 0.5 m elevation and in soil air void fraction.

In an experiment to determine the diurnal variation of radon-222 emanation, a sample was taken every 15 minutes for 24 consecutive hours, a total of 96 samples being collected, each on a different plot. The plots were confined to an area approximately 3 m wide by 12 m long to ensure relative homogeneity over the surface sampled. In addition to the emanation determinations, meteorological and soil conditions were recorded and noted for the testing interval. Among these were cloudiness, wind speed and direction, wet and dry bulb temperature, barometric pressure, and soil moisture content within the top 5 cm of soil. The site for the 24-hour test was a micrometeorological plot maintained by the Department of Agronomy of the University of Illinois, Urbana. It was grass covered, had been mown regularly, and was well suited for such a test because of uniformity of drainage, soil type, and surface. The soil was classified as a Flanagan type, a well-drained soil with a little more than 1 m of loess above glacial till. The loess is a wind-deposited soil composed of thoroughly mixed, finely divided clays and silts from Iowa, Nebraska, Kansas and other western states.

Table 2 presents hourly mean values of radon-222 flux densities and associated soil and meteorological conditions. These data and the individual observations of radon emanation are shown graphically in Fig. 6. Although quantitative comparisons are not justified by a single sequence of measurements, several qualitative observations are afforded. It appears that the rate of radon emanation is not dependent upon the absolute value of wind speeds, but rather upon atmospheric turbulence. Thus, during the afternoon there is a period of rather high and variable emanation and a similar

Table 2. Radon-222 flux and meteorological conditions. 7–8 August 1963.

Date	Time hour beginning (CST)	Flux density hourly mean 10^{-18} c cm^{-2} sec^{-1}	Soil condition			Meteorological conditions: Wind						
			Porosity	Air void fraction	Moisture (% dry wt)	Dir (° azimuth)	Speed (m sec^{-1})	Std. dev. (m sec^{-1})	Temp. DB (°C)	Humidity R.H. (%)	Sky cloudiness (%)	Press (in. Hg)
7 Aug.	1115	187										29.36
	1215	117	0.56	0.33	0.17	360	2.07	±0.48	32.0	45	10	29.35
	1315	319										29.34
	1415	1616	0.55	0.31	0.17	360	1.97	±0.45	31.4	48	0	29.32
	1515	1075										29.32
	1615	786	0.59	0.42	0.13							29.31
	1715	446										29.30
	1815	189	0.59	0.40	0.14	22	0.82	±0.25	28.0	61		29.30
	1915	194							25.5	69		29.31
	2015	232										29.32
	2115	225										29.33
	2215	267				45	0.70	±0.15	21.2	83	0	29.33
	2315	238	0.60	0.42	0.14							29.33
8 Aug.	0015	275				45	0.60	±0.19	17.0	96	0	29.33
	0115	292							18.2	91		29.33
	0215	253				68	0.53	±0.05	17.0	93	5	29.32
	0315	299							15.3	99		29.32
	0415	1632				45	0.53	est.	15.8	96	5	29.32
	0515	634										29.33
	0615	327	0.58	0.38	0.14	180	0.37	±0.12	19.7	93	45	29.33
	0715	433				225	1.07	±0.19	24.6	71	100	29.34
	0815	372										29.33
	0915	328	0.54	0.35	0.13	225	1.00	±0.36	26.8	73	100	29.32
	1015	302										29.31

but shorter period near sunrise. During the stable night-time period, the flux was relatively constant, increasing slightly. The increase probably is attributable to a gradual restoration of the radon concentration in the top layers of the soil which were depleted during the previous daytime period of turbulence. These statements are compatible with the findings reported by Evans, Kraner and Schroeder (1962). Furthermore, changes in soil pore space available for gases probably contributed to over-all effects. Barometric pressure, atmospheric temperature, relative humidity, and wind direction changes may have produced some response, too, but the data are too scanty to permit any definitive statements. Even in an isolated experiment such as that just discussed, however, it is evident that the diurnal variation of radon emanation is a rather complicated function of many interrelated parameters. The exhalation of other gases would most likely exhibit complex dependencies also.

4. Summary and conclusions

A device and a system for the collection of the emanation of a soil-gas, radon-222, has been designed, constructed and tested. It was designed to provide conditions within the collecting device which simulate those of the surrounding unobstructed atmosphere. Disturbance of the sampling site prior to the collection of the emanation was avoided. Dependence of emanation upon wind velocity across the soil surface has been demonstrated. Long sampling periods with this system give evidence neither of accumulation of abnormally high concentrations of gas in surface soil nor its depletion due to the presence of the collecting device.

The system has been used for a comparison study of emanation in a 26 km by 26 km pattern over soils with a common geologic history. No significant variation of emanation with location or soil type was found at the 90 per cent confidence level. A study of the diurnal variables such as wind speed and atmospheric stability was observed.

The equipment described here appears to be entirely satisfactory for the collection of radon-222 diffusing through the earth-atmosphere interface. With carefully considered modification it should be adaptable for the collection of other materials rising from the surface of the earth.

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