Soil concentrations of "emanating radium-226" and the emanation of radon-222 from soils and plants

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ABSTRACT

The emanation of radon-222 from soils has been studied. Factors investigated which affect the rate of emanation include wind speeds across the surface, atmospheric turbulence, soil moisture, soil type and location. Values of emanation and concomitant variation in these factors are reported.

An investigation of radon-222 release from leaf surfaces has been initiated; preliminary results of this study are reported.

Measured values of radium-226 in the soil capable of giving rise to diffusing radon were found to vary 70-fold in different geological regions but were uniform in Champaign County, Illinois, the content being $(0.64 \pm 0.09) \times 10^{-12}$ grams of "emanating radium-226" per gram of soil. The emanation rate of radon-222 from soils into the atmosphere varied 1000-fold among geographical regions but was shown to be uniform in Champaign County, averaging $(140 \pm 73) \times 10^{-18}$ curies per square centimeter per second.

The findings of the research indicate that radon-222 may be used successfully as a cost-free tracer in studies of vertical diffusion in Champaign County, Illinois.

1. Introduction

Israel (1951) discussed "the use of radioactive emanations as an indicator of *austausch* movements" and listed earlier studies of atmospheric concentrations and soil emanations of naturally occurring radioactive materials. A number of investigations of emanation of radioactive gases from the soil, including those reported herein, have been made since 1951. When considering the use of radioactive emanations from the soil as a tracer material, one should heed the advice of Jacobi & Andre (1963) that any study of vertical diffusivity using a single vertical array of sampling inlets requires an evaluation of the horizontal distribution of the material to be used as a tracer. A horizontally isotropic distribution is desirable, for under such a circumstance, horizontal advection of the tracer is of no consequence, and tracer concentrations in the atmosphere are functions only of altitude, source strength, and vertical diffusion properties, primarily turbulence. A study of vertical diffusivity using natural radioactive emanations from the soil as a tracer has the advantages that the materials are chemically and biologically inert, do not change phase, as does water, and can be measured in trace amounts with required precision. The similarity between radioactive emanations from soils and the "area source" problem in the field of air pollution is apparent.

Observations of radon-222 concentrations at several elevations for periods of approximately 24 hours have been reported by Moses, Stehney & Lucas (1960). More recent studies at Argonne National Laboratory continue to demonstrate (see Figure 1) the responsiveness of this material to atmospheric variables. It is apparent that having such data on the time rate of change of quantities of material in the atmosphere and knowing the flux of that material at the lower boundary, the flux of materials at several elevations could be determined. Determination of flux densities of materials at various elevations are used in investigations of vertical diffusion in the atmosphere.

Data for 8 groups of radon concentration measurements of the kind shown at the bottom of Figure 1 have been combined to provide

[1] Presently at the University of California at Berkeley.

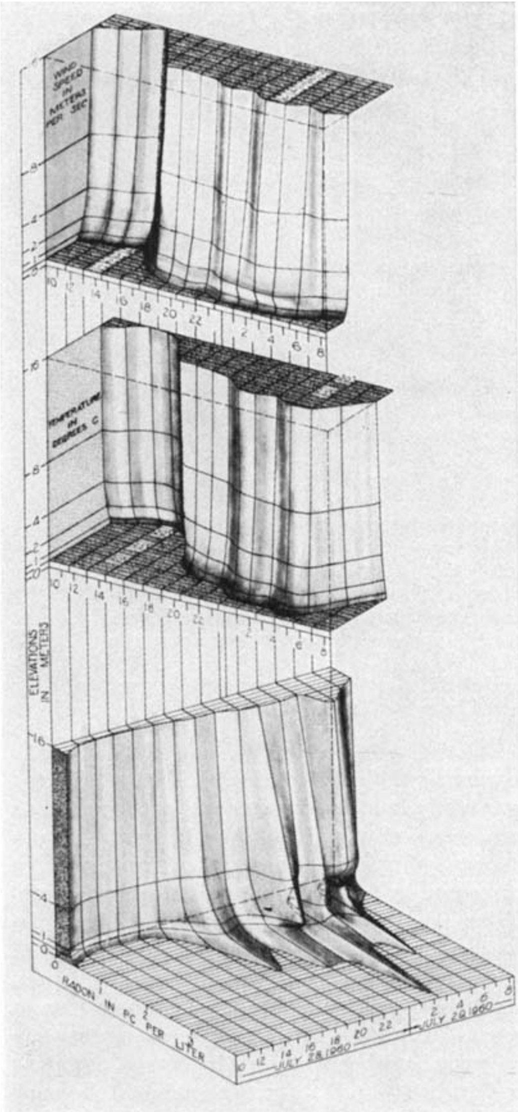

FIG. 1. Concurrent radon concentrations, temperature, and wind speeds plotted with elevations and time at Argonne National Laboratory under the following conditions: good visibility and convective type clouds characterized the day while the night was without clouds and clear until after midnight when ground fog and fog developed until daybreak.

average values shown in Figure 2. While such limited data does not provide a radon climatology for the observation site at Argonne National Laboratory in the State of Illinois, U.S.A., it provides an indication of general features of radon behavior in the atmosphere. These include: (1) highest concentrations near the earth, the source, (2) maximum concentrations during the night and minimum concentrations during the day, (3) inverse fumigation just after sunrise, (4) late morning radon concentration profiles showing weak vertical concentration gradients or evidence of good mixing, and (5) early evening concentrations increasing more rapidly near the ground than above as shown by the slanting isopleths in the right portion of the figure. The measurements which provide the basis for these figures also gave indication of the desirability of further investigation of the utility of radon as a tracer material in studies of the atmosphere.

The primary purpose of the research reported herein was to determine the uniformity of emanation of radon-222 from soils in Champaign County, Illinois, in order to judge its suitability for use as a tracer material for studies of vertical diffusivity. This study has had three areas of effort: (1) soil "emanating radium-226" concentrations, defined as the quantity of that material, the parent of radon-222, sufficiently near the surface of soil particles that it is capable of releasing radon into the pore spaces between the particles. That portion released which is held within crystal structures is not of interest; (2) the emanation of radon-222 from soils, its variation with location, soil type, wind speed, turbulence, soil moisture, pressure, etc., and (3) the release to the atmosphere of radon-222 by leaves of plants.

2. Experimental methods

While daughters of uranium-238, uranium-235, and thorium-232 contribute to the atmosphere's natural radioactivity, our subject for study was radon-222. A number of processes are available for determining the quantity of this material present at a location; we used the method described by LUCAS (1957). This method effectively excludes other radionuclides normally present in the atmosphere and is not dependent upon the state of equilibrium between radon-222 and its daughters.

Briefly the analytical method included collection of the radon by adsorption on activated coconut charcoal chilled in a dry ice solution. The charcoal trap containing the sample was brought to the laboratory where the radon-222 was desorbed by heating the charcoal to 300C.

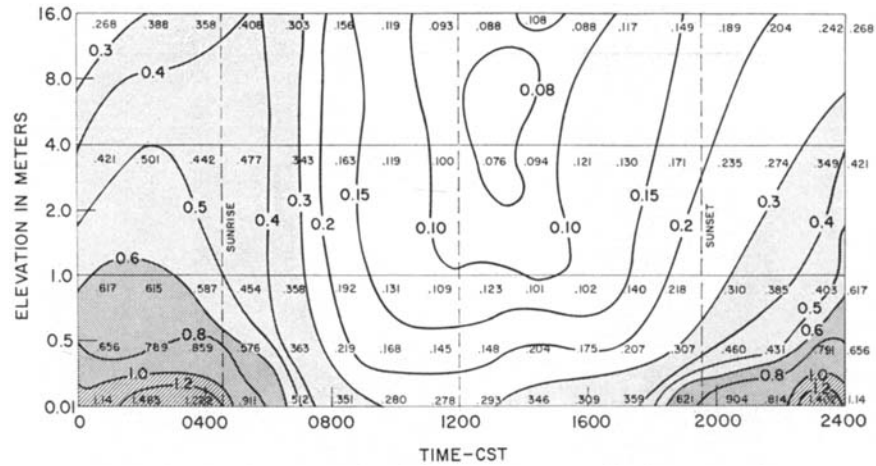

FIG. 2. Radon-222 concentration (in 10^{-12} curies liter^{-1}) variation with elevation and time at Argonne National Laboratory; average values for eight 24-hour periods.

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 bottles were constructed of kovar steel shells covered at one end by quartz glass windows and lined with silver-activated zinc sulfide scintillation material. After transfer, the sealed bottle was allowed to stand about two hours during which short-lived radon decay products approached equilibrium with the parent radon-222. 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 in determining the amount of radon collected.

Emanation from the soil was collected by a system described by PEARSON, RIMBEY & JONES (1965). The collection system consisted of a sealed circulating pump, carbon dioxide and water traps, and sample traps which removed the radon from circulating air. The collection device used was a rectangular stainless steel tube with one open face which was placed over the site to be sampled. One who endeavors to measure interface phenomena must recognize the hazard that the sampling method may so disturb the environment within which the measurement is to be made that results obtained are meaningless. Therefore, we simulated natural conditions as closely as possible within our collector, used a portable system so that the sampling site could be in equilibrium with natural conditions before initiation of sampling, and began sampling within 1 or 2 minutes of placing our collector.

"Emanating radium-226" of soils was determined by placing about 5 g of dried soil in a flask with a weak acid solution. The flask was then flushed with radon-free air, sealed, and left undisturbed for about one week for radon-222 accumulation from its parent, radium-226. After standing, the radon was transferred to charcoal and the analysis accomplished as previously described.

3. "Emanating radium-226" measurements

In the presentation of a summary of our research, it seems logical to present first a summary of measurements made of quantities of "emanating radium-226" in a variety of soils (see Table 1). In this table soils are classified, roughly, as those containing clays or fine materials, those which are predominantly sandy in texture, and those obtained at locations where uranium-238 and/or radium-226 are present in commercially important concentrations regardless of soil texture. The first three soil types listed, Flanagan, Catlin, and Drummer, cover approximately 85 % of Champaign County, Illinois, and are not statistically significantly different from each other (at the 90 % confidence

TABLE 1. *Radioactivity of soils (in 10^{-12} g of "emanating radium-226" per g of dry soil).*

Location	"Mixed Clay" Soil	"Sandy" Soil	Mining Operation
1. Agronomy Plots, Urbana, Illionis			
Flanagan type (mean of 20 samples)	0.679		
Catlin type (mean of 15 samples)	0.633		
Drummer type (mean of 15 samples)	0.592		
(mean of 50 samples)	0.64		
2. Argonne National Lab., Argonne, Ill. (mean of 3 samples)	0.690		
3. Eddyville, Illinois	0.510		
4. Metropolis, Illinois	0.516		
5. Scottsville, Kentucky	0.576		
6. Sylacauga, Alabama	0.794		
7. Albany, Georgia (red clay type)	0.390		
8. Leesburg, Florida		0.232	
9. Galveston, Texas (beach sand)		0.147	
10. Texas City, Texas (Zircon sand piled on beach in 1963)		0.623	
11. Rice University "Commons", Houston, Texas	0.460		
12. Atwood, Kansas	0.484		
13. San Luis Valley, Colorado	0.342		
14. Central City, Colorado (mining area) Wood Mine (ore and tailings, respectively)			14.3 & 0.45
Xenotine from Spring Gulch			5.26
Yttrium outcropping			2.87
15. Blow sand over Todilito limestone, Grants, New Mexico			10.4
16. Socorro, New Mexico, NMIMT			
Weir Hall		0.244	
Workman Mine (floor)	0.414		
17. Iceland (six locations)	0.172		

level) with regard to "emanating radium-226" content.

It is interesting to observe that the soils with a sandy texture have a smaller "emanating radium-226" content than do those in the first column, the clay type, except for the Zircon sand piled on the beach at Galveston, Texas. This latter material was used as a source of radioactivity during a cross-calibration experiment conducted in conjunction with the First International Symposium on the Natural Radiation Environment at Rice University, Houston, Texas, in April, 1963. It is believed that the other sandy soils exhibited lower "emanating radium-226" concentrations than the clay type soils because sandy soil particles have a smaller surface to volume ratio. Thus, a radon atom produced in the interior of a sandy soil particle has less chance of escaping into soil pore spaces, the measured "emanating radium-226" being therefore reduced.

"Emanating radium-226" contents of some of the soils from the mining areas, as expected, are higher than the others. The samples obtained from Iceland exhibited the lowest activities of all those sampled. Throughout the various geological regions from which samples have been analyzed, then, observed concentrations of "emanating radium-226" varied seventy-fold.

4. Soil emanation measurements

Table 2 presents a summary of a number of measurements of radon-222 emanation from soils. Again Flanagan, Catlin, and Drummer soils were studied most carefully because they represent the most important soil source in our local area. At the New Mexico Institute of Mining and Technology, Socorro, New Mexico, we made measurements of soil emanation at the same site used by WILKENING & HAND (1960); the results of both sets of observations are shown in the table.

Table 3 lists individual measurements of the emanation of radon-222 at two locations where rock with high concentrations of uranium-238 outcrops the surface. The largest value observed near Grants, New Mexico, is approximately 1000 times as large as that observed at many

TABLE 2. *Rates of emanation of radon-222 from soils (in 10^{-18} curies cm^{-2} sec^{-1}).*

Soil type and/or location	No. of samples	Mean ± S.D.
Champaign County, Illinois		
Flanagan	72	111 ± (39)
Catlin	72	157 ± (72)
Drummer	72	151 ± (96)
Flanagan, Catlin Drummer	216	140 ± (73)
Brenton (5·11·64)	8	91 ± (25)
Sidell (5·11·64)	8	174 ± (42)
Thorp (5·15·64)	8	75 ± (18)
Proctor(5·15·64)	8	49 ± (11)
Cullo (5·15·64)	8	53 ± (16)
Argonne National Laboratory, Argonne, Ill.	8	57 ± (5)
NMIMT, Socorro, New Mexico		
Weir Hall (Wilkening and Hand)		90
Weir Hall (Pearson and Jones)	6	102 ± (30)

other locations. The large variation in values obtained was the result of sampling at varying distances from the outcrop over soil or blow sand of different depths.

5. Environmental influences on soil emanation

The primary purpose of our study was to determine the effects of meteorological and soil conditions, time, soil type, and location upon the release of radon-222 from soils in our local area, Champaign County, Illinois. These effects have been reported in PEARSON, RIMBEY & JONES (1965) and in PEARSON & JONES (1965) and are summarized here.

Wind speed. At the very low velocities which normally occur near the soil, emanation is independent of velocity, but at abnormally high velocities a linear relationship with velocity can be observed during a 5-minute sampling period.

Soil moisture. When soils were moistened with approximately 25 mm of water and observed during the following 8 hours of drying, no significant variation of emanation with soil moisture was observed. However, for greater lengths of time, days to weeks, a significant variation of emanation with soil moisture was observed. When the soil was wet (air void fraction[1] about 0.17) the emanation measured was approximately one-third that observed when average soil moisture conditions (air void fraction about 0.40) were prevalent.

Diurnal effects—atmospheric turbulence. Emanation was observed to vary markedly in one 24-hour study. The variation of emanation was attributed primarily to atmospheric turbulence. Thus, near sunrise and in mid-afternoon, when the atmosphere was most turbulent near the soil surface, radon effluxation was highest and most erratic. During the stable nighttime, the flux was relatively constant, increasing slightly. This increase was attributed to restoration of the radon concentration in the top layers of soil which were depleted during the previous daytime period of high emanation.

Seasonal effects. There was a marked decrease of emanation when soil was cold or frozen. However, during a January thaw when approximately 7 cm of soil nearest the surface was not frozen, a very high and erratic emanation was observed.

Location and soil type effects. A study of 9 sites within a 13 km radius and over the three most common local soil types revealed, at the 90 % confidence level, no differences in emanation due to location or soil type. As soils in the prevailing upwind direction, for hundreds of kilometers, have the same geologic history, it is believed that the local site is suitable for studies of vertical diffusivity using radon-222 as a tracer material. However, emanation varied

[1] The term "air void fraction" is used to indicate that fraction of a soil volume occupied by soil gases.

TABLE 3. *Rates of emanation of radon-222 from U^{238} bearing rock outcrops over varying depths of soil or blow sand (in 10^{-18} curies cm^{-2} sec^{-1}).*

Location...	Central City, Colorado	Grants, New Mexico
Sample No.		
1	1.358	1.598
2	3.154	21.277
3	2.280	21.349
4	2.444	127.136
5	991	24.932
6	563	43.205
7	2.870	48.649
8	—	13.997
Mean & S.D.	1.951 ± 987	37.768 ± 26.862

TABLE 4. *Preliminary tests of emanation of radon 222 from leaves of plants, April 21, 1965.*

Flux density in 10^{-18} curies cm^{-2} sec^{-1}. Total emanation in 10^{-12} curies. Plants and pots were located in Bldg. No. 60 greenhouse at Argonne National Laboratory and had been growing since about January 1, 1965.

Location	Type of Measurement	Tobacco 20 min sample	Tobacco 1 hr. sample	Cast bean 1 hr. sample
Top of leaf	Flux density	(13.9 ± 2.1)	(7.53 ± 2.56)	(6.22 ± 1.08)
Top of leaf	Total emanation	(0.267 ± 0.040)	(0.437 ± 0.090)	(0.359 ± 0.062)
Bottom of leaf	Flux density	(28.0 ± 4.8)	(9.86 ± 1.14)	(8.56 ± 1.43)
Bottom of leaf	Total emanation	(0.540 ± 0.093)	(0.579 ± 0.66)	(0.494 ± 0.083)
Soil in pot (20 min. sample)	Flux density	(67.3 ± 7.4)		(76.5 ± 9.7)

1000-fold among geographical regions with widely differing geophysical histories.

Barometric pressure, atmospheric temperature, and relative humidity. Barometric pressure, atmospheric temperature and relative humidity may have produced some response, too, but the data are too limited and the effects too small to permit any definitive statements.

6. Leaf emanation measurements

The 20C water–air phase distribution ratio of 0.25 for gaseous radon (FAUL, 1954) and the 100 to 1000 times greater concentration of radon in soil air as compared to the atmosphere above the soil suggested that soil water would be relatively rich in dissolved radon. From these two conditions the hypothesis may be forwarded that plant roots acquiring the radon-222-rich soil water subsequently transport both water and dissolved radon to above-ground plant parts where both are released to the atmosphere. An investigation of such phenomena is being accomplished in cooperation with Dr. Richard R. Dedolph, Division of Biology and Medicine at Argonne National Laboratory, Argonne, Illinois.

Our first experiment was initiated in the winter of 1964–1965 using greenhouse-grown tobacco (*Nicotiana tabacum* L., cv. Maryland Mammoth) and castor bean plants (*Ricinus communis* L.). Plants were grown in potting soil (1:1:1 loam, peat, sand) in glazed 115 liter earthenware crocks. When the plants were in the early flowering stage, leaf radon emission was assayed with a system similar to that used for soil radon emanation, but with a collector 2.5 cm × 7.5 cm in size. This collection system was not optimal for sample collection from leaves because water vapor, carbon dioxide, and light were excluded from the leaf surface during sample collection. Each of these exclusions is capable of modifying water evaporation from leaf surfaces by inducing changes in stomatal openings.

Despite the cited inadequacies of the sampling system for measurement of leaf surface emanations which render the absolute quantitative value of these data questionable, the data (see Table 4) clearly show that measurable quantities of radon-222 are released from leaf surfaces. The marked decrease in rate of radon emission with length of sampling period, which was particularly evident on the lower leaf surfaces of tobacco, indicates that the inadequacies of the sampling system employed materially decreased stomatal openings and transpiration rates. Differences between the upper and lower leaf surfaces, when the shorter sampling time was employed, are wholly consonant with stomatal distributions on these surfaces (MILLER, 1938).

During the summer of 1965 we modified our sampling system so that leaf radon-222 emissions could be determined under more nearly natural conditions. These sampling modifications provided, within the collecting device, nearly normal light intensities as well as movement of air with nearly normal concentrations of water vapor and carbon dioxide. Six measurements of soil emanation and six of release of radon-222 from leaves of field corn were made on August 5th when the corn was beginning to develop tassels. These measurements were made between 1100 and 1600 local standard time. At this time release from leaves was measured at

$(247 \pm 19) \div 10^{-18}$ curies cm^{-2} sec^{-1}, a figure which includes both the top and bottom of the leaf. Concurrently, soil emanation was $(92.1 \pm 30.1) \times 10^{-18}$ curies cm^{-2} sec^{-1}. While the leaf to soil release ratio was over 7 to 1 for this experiment, it should be noted that the soil emanation was somewhat low, probably due to high soil moisture content. However, the leaf contribution is greater than the average emanation for Champaign County, Illinois, 140×10^{-18} curies cm^{-2} sec^{-1}. This, coupled with the fact that the corn leaf area to soil area ratio can reach 3 to 1 emphasizes the importance of considering the radon-222 contribution of plants during daylight hours.

7. Summary and conclusions

1. The ability of soil to produce radon-222 was investigated by measuring its "emanating radium-226" content. While this ability was shown to be uniform in Champaign County, Illinois, the content being $(0.64 \pm 0.09) \times 10^{-12}$ grams per gram of dry soil, a 70-fold range of content was found in different geological regions.

2. The average emanation of radon-222 from soils at the same location was found to be $(140 \pm 73) \times 10^{-18}$ curies cm^{-2} sec^{-1} or 2.5 ± 1.3 atoms cm^{-2} sec^{-1}.

Emanation rates under various conditions were studied with the following conclusions:

A. At wind speeds normally observed near the soil surface, no significant variation was observed, but at abnormally high speeds a linear relationship prevailed during 5 minute sampling periods.

B. Within 8-hour periods of drying no significant variation of emanation was observed. However, when intervals of days or weeks were considered, emanation increased 3-fold as soil changed from a moist to an "average" condition.

C. During periods when the atmosphere was most turbulent, emanation was highest and most erratic. When turbulence decreased, emanation was smaller and less variable.

D. Within our local area, variations in soil type and location were not significant. However, the emanation varied 1000-fold among other geographical regions.

E. During cold weather and frozen soil conditions, emanation was reduced.

3. Emanation of radon-222 from leaves of plants has been collected and measured. The first quantitative measurements made on field corn at the early tasseling stage during early afternoon provided an emanation value of 247×10^{-18} c cm^{-2} sec^{-1} for both upper and lower leaf surfaces.

Acknowledgements

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КОНЦЕНТРАЦИЯ В ПОЧВЕ «ВЫДЕЛЯЮЩЕГО РАДИЯ 226» И ВЫДЕЛЕНИЕ РАДОНА 222 ИЗ ПОЧВЫ И РАСТЕНИЙ

Изучается выделение из почвы радона 222. Исследуются факторы влияющие на величину истечения, включая скорость ветра над поверхностью, атмосферную турбулентность, влажность и тип почвы и место. Представлены количество выделяющегося радона и изменения этой величины, связанное с перечисленными факторами.

Также представлены предварительные результаты начатого исследования выделения радона 222 через листья растений.

Измереное количество радия 226 в почве допускающее увеличение диффузий радона, изменялось в 70 раз в различных геологических областях, но было равномерно в Champaign County, Illinois. В этих областях содержалось $(0{,}64 \pm 0{,}09) \times 10^{-12}$ г «выделяющего радия 226» на грамм почвы. Количество выделяющегося из почвы в атмосферу радона 222 менялось в 1000 раз в различных геологических областях, но в среднем было $(140 \pm 73) \times 10^{-18}$ кюри см$^{-2}$ сек$^{-1}$.

Полученные данные указывают на возможность успешного использования радона 222 в качестве даровых меченых атомов при исследовании вертикальной диффузии в Champaign County, Illinois.