RADON FLUX AT THE EARTH-AIR INTERFACE

by

JOHN E. HAND

Submitted in partial fulfillment of the requirements

for the degree of

MASTER OF SCIENCE IN EARTH SCIENCE

NEW MEXICO INSTITUTE OF MINING AND TECHNOLOGY

1958

TABLE OF CONTENTS

Page

LIST OF FIGURES

LIST OF TABLES

ACKNOWLEDGMENT

ABSTRACT

The purpose of the investigation was to measure the radon content of the atmosphere and of soil gases escaping through the surface of the earth. The method of emanometry was used to accomplish the required analyses. Results have been obtained with an accuracy of $\pm$ 5 percent, giving values comparable to those reported for other parts of the world. The average atmospheric radon concentration at ground level in the Socorro area was found to be $2.3 \cdot 10^{-13}$ curie/liter, and the average flux at the earth-air interface was found to be $9 \cdot 10^{-17}$ curie/cm^2-sec. The atmospheric radon concentration showed a diurnal fluctuation, whereas the radon flux at the earth-air interface appeared to be relatively constant over a diurnal period.

Calculations based on idealized soil and atmospheric conditions indicate that the atmospheric radon content represents an equilibrium between atmospheric radon decay and the radon flux at the earth-air interface. A further calculation indicates that the radium content of the local soil is about $1 \cdot 10^{-12}$ gm Ra/gm soil.

THE RADON FLUX AT THE EARTH-AIR

INTERFACE

INTRODUCTION

Elster and Geitel[1] showed that a negatively charged wire exposed to
the atmosphere for a few hours received a radioactive deposit that was
similar in character to some of the decay products of radium. Positive
proof that this activity was due to radium emanation (hereafter called
radon) was accomplished in 1904 by Bumstead and Wheeler.[2] Further work
by other early investigators provided conclusive evidence that the radon
observed in the free air was due to the diffusion of radon gas from the

1. J. Elster and H. Geitel, "Beschreibung des Verfahrens zur
Gewinning vorübergehend radioaktiver Stoffe aus der atmosphärischen Luft,"
Zeitschrift für Physik, 3, 1902, pp 305-310.

2. H. A. Bumstead and L. P. Wheeler, "On the properties of a
radioactive gas found in the soil and water near New Haven," American
Journal of Science, 17, 1904, pp 77-111.

soil into the atmosphere.[3-5]

An examination of the series of naturally occurring radioactive nuclides reveals the origin of radon (Table 1). Since the radon appears as an intermediate gaseous product, the three natural series need be considered from the radium member only. Expressions for the quantity of the radon daughters present as a function of time are given in Appendix I. It is well established that all soils and rocks contain radium[6] so that one can expect to find radon in nearly all localities. Recent investigations have indicated that the radon content of the air is a balance between the decay of the atmospheric radon and the radon flux at the earth-air interface.[7,8] However, a study of the literature reveals that the data supporting this idea have been gathered from widely scattered localities. In addition,

3. L. B. Smythe, " On the supply of radium emanation from the soil to the atmosphere," Philosophical Magazine, 24, 1912, pp 632-637.

4. E. Rutherford, Radioactive Substances and Their Radiations, Cambridge University Press, Cambridge, 1912, pp 623-637.

5. J. Satterly, "On the amount of radium emanation in the lower regions of the atmosphere and its variation with the weather," Philosophical Magazine, 20, 1910, pp 1-15.

6. J. Faul, Nuclear Geology, John Wiley and Sons, Incorporated, New York, 1954, p 156.

7. J. R. Wright and O. F. Smith, "The variation with meteorological conditions of the amount of radium emanation in the atmosphere, in the soil gas and in the air exhaled from the surface of the ground at Manila," The Physical Review, (2), 5, 1915, pp 459-482.

8. V. F. Hess, "On the radon content of the atmosphere and the radium content of river water," Terrestrial Magnetism and Atmospheric Electricity, 48, 1943, pp 203-206.

Table 1. Pertinent portions of the natural radioactive series.

Series	Member	Half-life*	Energy (Mev)		
			α	β	γ
Thorium					
	Th X, Ra^{224}	3.64d	5.68		
	Tn, Rn^{220}	54.5s	6.28		
	Th A, Po^{216}	.158s	6.77		
	Th B, Pb^{212}	10.6h		0.36	
	Th C, Bi^{212}	60.5m	6.05		
Uranium					
	Ra^{226}	1600y	4.79		0.19
	Rn^{222}	3.825d	5.49		
	Ra A, Po^{218}	3.05m	5.99		
	Ra B, Pb^{214}	26.8m		0.65	
	Ra C, Bi^{214}	19.7m	5.50	3.15	1.8
	Ra C', Po^{214}	$1.5 \cdot 10^{-4}$s	7.68		
	Ra C'', Tl^{210}	1.32m		1.80	
	Ra D, Pb^{210}	22.2y		0.03	0.05
Actinium					
	Ac X, Ra^{223}	11.2d	6.72		
	An, Rn^{219}	3.92s	6.82		
	Ac A, Po^{215}	$1.8 \cdot 10^{-3}$s	7.36		
	Ac B, Pb^{211}	36.1m		0.5 1.4	0.6
	Ac C, Bi^{211}	2.16m	6.62		

* s = second, m = minute, h = hour, d = day, y = year.

Israel[9] shows that in some instances the data are not reliable.

For the southwestern United States, there is no information available concerning the radon flux, the atmospheric radon content, or the possible relationship between these quantities. Hence, the purpose of this thesis is to investigate the local conditions of atmospheric radon occurrence with respect to the following specific points: (1) The direct measurement of the radon content of the atmosphere at the ground level; (2) the quantitative determination of the radon flux at the earth-air interface, and (3) the relationship of the radon flux at the earth-air interface to the radium content of the soil.

EXPERIMENTAL PROCEDURE

Method. The method employed in this study for the quantitative determination of radon utilizes the property of radon to condense at the temperature of liquid oxygen (-183°C). The radon carried by a measured volume of air is condensed within a coil cooled to this temperature. After a collection is completed, the coil is allowed to warm up, and the radon vaporizes. The radon is then transferred to an ionization chamber where its activity causes a small current that is proportional to the number of radon atoms present. The metallic nature of the radon daughters causes them to remain in the coil. After calibration, the current is taken as a measure of the amount of radon in the air which has passed through

9. H. Israel, "Radioactivity of the atmosphere," Compendium of Meteorology, American Meteorological Society, Boston, Massachusetts, 1951, pp 155-162.

the condensing coil. This process is called the method of emanometry.

The emanometric method of analysis excludes the presence in the samples of Rn^{219} and Rn^{220} because of the short half-lives of those two isotopes (Table 1). Consequently, only the radon from the uranium series is available for measurement.

Equipment and Apparatus. The equipment and apparatus used to measure radon concentrations and the radon flux is illustrated in Figures 1 and 2. In order to collect radon samples from the surface of the soil, a lid-type device, covering an area of 193 cm^2, was placed on the ground, and air was drawn through in the manner indicated. Figure 2 is a schematic diagram of the radon condensing and measuring apparatus. The apparatus consists of the three separate sections indicated by the dotted lines; each section will be described individually:

1. Collection. Samples were collected from the atmosphere by connecting the system inlet to the outside air with Tygon tubing. For the collection of radon escaping from the surface of the earth, a "lid" (Figure 1) was attached to the end of the Tygon tubing and placed on the ground as shown. The collection system consisted of trap 1, the condenser coil, the flowmeter, pump P_2, and the connecting lines and valves. Trap 1, immersed in a mixture of dry-ice and acetone, removed the water vapor from the incoming air. The condensing coil was constructed from 3/8 inch-diameter copper tubing. To provide sufficient surface at low temperature within the coil, the tubing was tightly packed with finely-cut copper turnings. The turnings were 1 mil thick, 10 mils wide, and cut about 1/8 inch long. After the tube was packed with the turnings for a length

To system
inlet

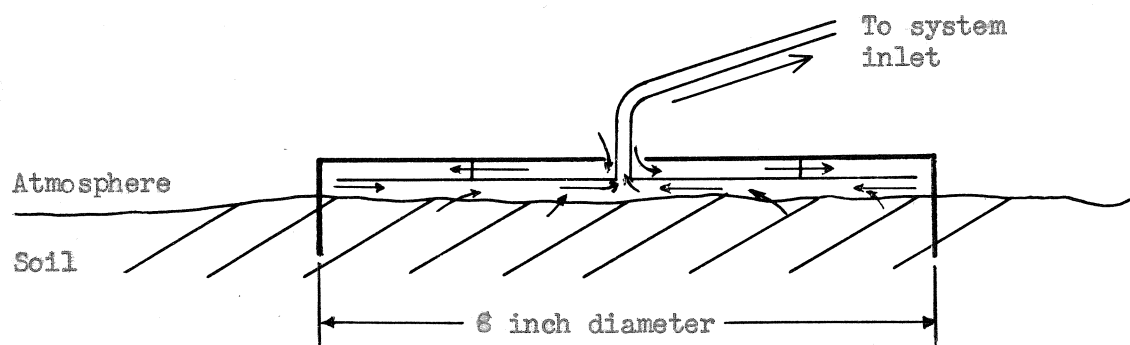Atmosphere

Soil

← 6 inch diameter →

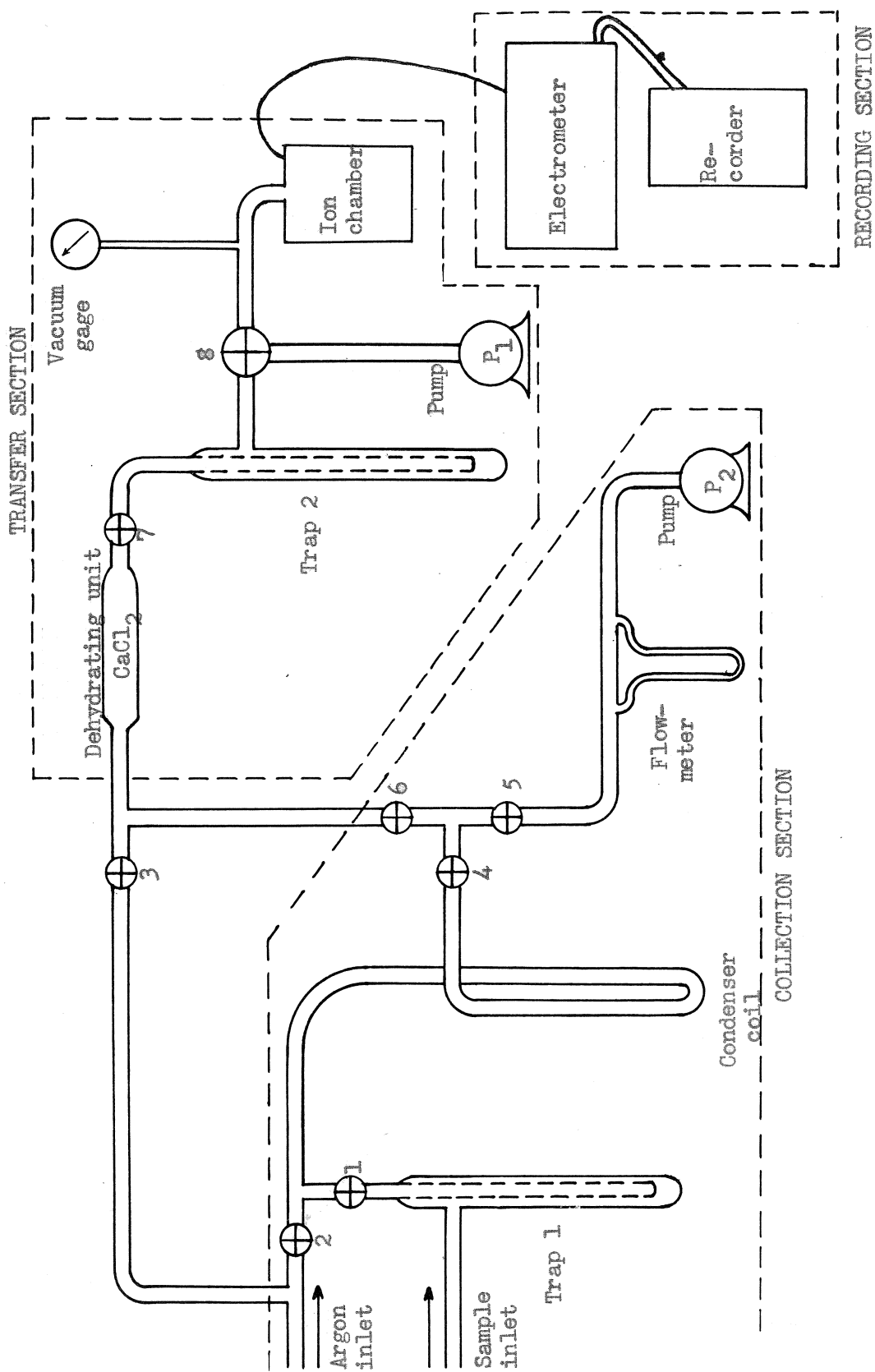Figure 1. Collection device for exhalation radon.

Figure 2. Radon collection apparatus

of 36 inches, it was bent to form a coil which would fit inside a 3 inch-diameter Dewar flask. This gave, essentially, a porous copper plug in which the radon condensed. The flowmeter was constructed from 5 mm Pyrex tubing, bent in the U-shape shown (Figure 3), and attached at each end to a 3-inch length of 1 mm capillary tubing which formed a bypass around the U-tube. To insure a low evaporation rate of the indicating liquid within the U-tube, the tube was filled with Octoil S. The flowmeter was calibrated for different rates of air flow by means of a Flowrator*. Pump P_2, used to pull the air through the collection system, was an old Cenco, Hyvac vacuum pump. Although the pump would no longer give a vacuum better than 10 cm Hg, it did provide a strong steady flow of air through the system.

2. Transfer. When the collection of a sample was completed, the condensed radon was transferred to the ionization chamber through a dehydrating unit and through trap 2. The dehydrating unit was filled with $CaCl_2$; the Pyrex trap was immersed in liquid oxygen. The purpose of both of these units was to insure the complete removal of water vapor and other contaminants from the gases entering the ionization chamber. Experiment showed that radon did not condense within the Pyrex trap. (Prior to the construction of the copper condensing coil, attempts to condense the radon in a Pyrex trap at the temperature of liquid oxygen were unsuccessful). The ionization chamber was built from an 8-inch length of 6 inch-diameter brass tubing, giving a volume of about 1.6 liters. The central electrode

* Flowrator is the trademark of the rate-of-flow indicator manufactured by Fischer & Porter Company, Hatboro, Pennsylvania.

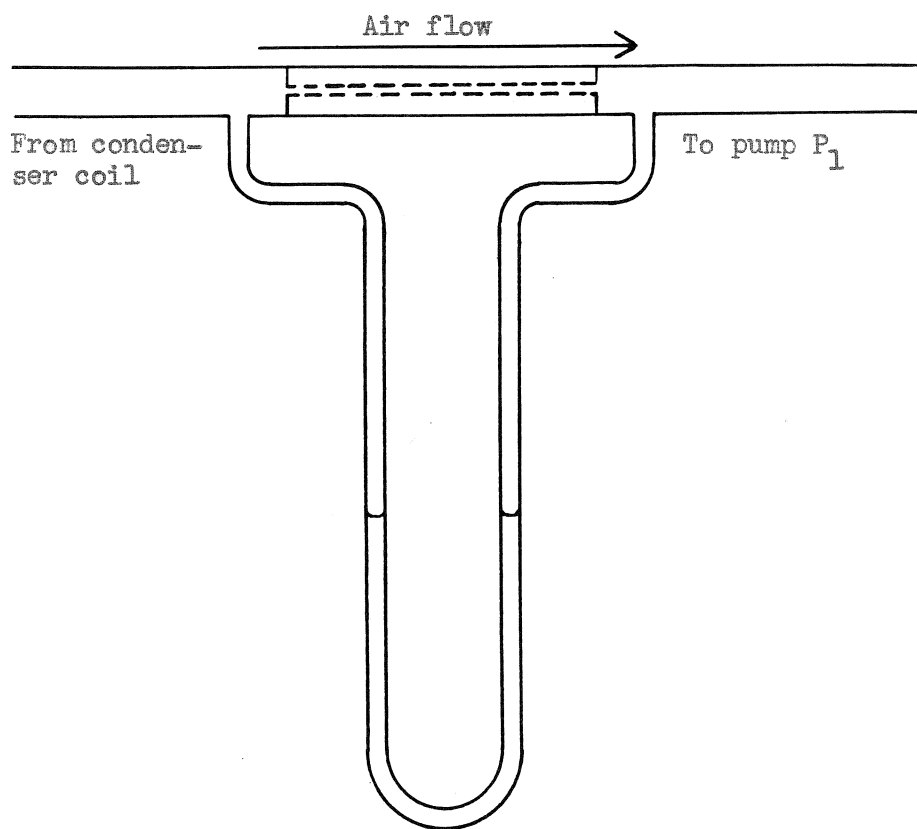Air flow

From conden-
ser coil

To pump P$_1$

Figure 3. Flowmeter.

was constructed from 3/8 inch-diameter brass rod. Stupakoff glass-to-metal seals supported the electrode at both ends. The potential applied to the chamber was 90 volts; the wall of the chamber was negative with respect to the central electrode.

      3. Measurement and recording. The ion-chamber current was measured with a Beckman Ultrohmeter, a direct-current amplifier type of electrometer. An Esterline-Angus recorder was used on the output of the electrometer to give a continuous graph of the radon activity. The lower limit of detection of the electrometer was $4 \cdot 10^{-15}$ amp above background current. The combined leakage current of the chamber and electrometer input was $1.6 \cdot 10^{-14}$ amp. With an argon filling of one atmosphere in the ion chamber, the normal background current plus leakage current was $(2.8 \pm 0.4) \cdot 10^{-14}$ amp.

    Calibration. Before experimental data obtained with the apparatus can be interpreted, the radon concentration of the air entering the condensing coil must be related to the ion current observed when the condensed radon is transferred to the ion chamber. This relationship is influenced by the collection efficiency of the condensing coil, by losses incurred while transferring the radon from the coil to the ion chamber, and by the ion chamber characteristics. To avoid the difficulties involved in applying individual corrections to the data for these factors, the overall efficiency of the apparatus was determined by making ion-current measurements with known quantities of radon.

    The calibration was determined by means of a standard solution containing $1.00 \cdot 10^{-7}$ gm radium obtained from the United States Bureau of

Standards. To facilitate handling the standard solution, a boiling-flask and reflux condenser were assembled (Figure 4) and attached to the system inlet. Once it was ascertained that the system was vacuum tight, the reflux condenser assembly was removed and the vial containing the standard was broken within the boiling-flask. Enough distilled water was added to the standard to give a total volume of about 250 ml solution. The reflux condenser was again attached to the system inlet, and the calibration performed in the following manner:

The solution was gently boiled in the flask at the same time that argon was admitted through a Pyrex tube into the lowest part of the boiling solution. The argon acted as a carrier gas, transporting the radon through the reflux condenser to the system inlet, through trap 1, and into the condensing coil. The regulator valve on the argon cylinder was adjusted to give the desired flow through the coil as indicated by the flowmeter. For this part of the experiment, pump P_2 was disconnected from the system, and the outlet side of the flowmeter was vented to the atmosphere by rubber tubing.

Before each calibration run, the accumulated radon in the reflux condenser system was removed by the following procedure: Valve 1 at the inlet of the apparatus was closed and the vent on the reflux condenser (Figure 4) was opened. The solution was boiled and argon was bubbled through, flushing the radon from the boiling-flask and reflux condenser into the atmosphere. When all of the accumulated radon had been removed, the reflux condenser vent was closed, valve 1 was opened, and a collection was made. With this method, only the radon that was generated

System inlet
connection

Vent

To atmosphere

Cooling water outlet

Reflux condenser

Argon
inlet

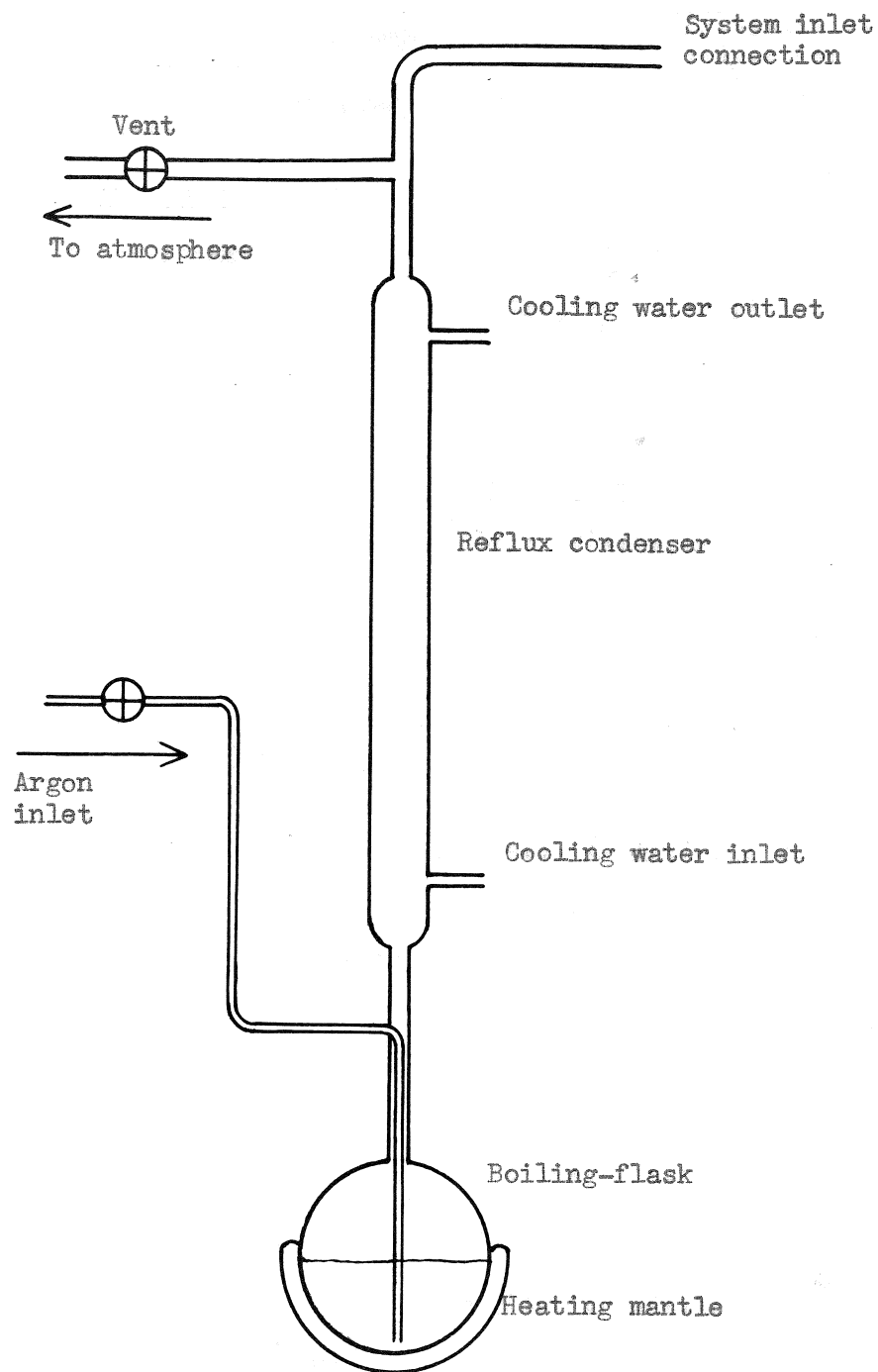Cooling water inlet

Boiling-flask

Heating mantle

Figure 4. Reflux condenser and boiling-flask assembly.

during the time of collection was deposited. Since the total radium content of the solution is known, the production rate of the radon, n_o, can be calculated:

$$n_o = \lambda_{Ra} N_{Ra}$$
$$n_o = 3.60 \cdot 10^3 \frac{\text{radon atoms}}{\text{sec}} \tag{1}$$

where λ_{Ra} is the radium decay constant and N_{Ra} is the number of radium atoms present. Because the time interval during which the radon atoms are in transit between the boiling-flask and the condensing coil is small, it is assumed that the generation rate, n_o, is equal to the rate of arrival of the radon in the condenser coil. For a collection efficiency of 100 percent, n_o is also the rate of condensation of the radon in the coil. Consequently, the number of radon atoms present in the condenser coil after a collection time T, is given by:

$$N(T) = n_o \int_0^T e^{-\lambda t} dt$$
$$N(T) = \frac{n_o}{\lambda}(1 - e^{-\lambda T}), \tag{2}$$

where λ is the decay constant of radon. When the collection is stopped, the radon is transferred to the ion chamber, and the current generated within the chamber is recorded. (In practice, the average transfer time was two minutes. Comparison of this time with the radon half-life, 3.825 days, reveals that the radon lost by decay during the time from the end of the collection until the start of the measurement is negligible). The ratio of N(T) to the observed ion current then provides the overall calibration of the apparatus. The validity of this calibration depends

upon the assumption that the overall efficiency of the system is 100 percent. To verify this assumption, the ratio is taken of the ion-chamber current obtained from a sample of known radon content to the current calculated for this sample from the energy-loss by ionization in the chamber. This calculation is based on the fact that each alpha particle from radon has an energy of $5.49 \cdot 10^6$ ev and that an average energy of 33 ev is required for one ionization process. Therefore, if we have $N(T)$ radon atoms, the disintegration rate is $\lambda N(T)$ particles/sec; hence the current, I, is

$$I \text{ (amp)} = \lambda N(T) \frac{\text{particles}}{\text{sec}} \cdot \frac{5.49 \cdot 10^6 \text{ev}}{\text{particle}} \cdot \frac{1 \text{ charge}}{33 \text{ev}} \cdot \frac{1.6 \cdot 10^{-19} \text{ coul}}{\text{charge}}$$

$$I \text{ (amp)} = 5.5 \cdot 10^{-20} N(T). \tag{3}$$

If $N(T)$ in this equation is converted to curies of radon, an initial ion current of $9.7 \cdot 10^{-4}$ amp can be expected provided that the ion chamber contains 1 curie of radon. (In order to exclude contributions to the current from the decay of the radon daughters, only the initial value of the ion current was used in the calibration and experimental data taken).

In the first experiments performed to determine the efficiency of the system, the condenser coil was made from 1/4 inch-diameter copper tubing which was closely wound to a diameter of 1.25 inches for a length of 11 inches. The total length of the tubing immersed in the liquid oxygen was about 11 feet. Data were taken for various air flow rates and collection times. At a flow rate of 1 liter per minute and a collection time of 1 hour, the efficiency of the system was found to be only 7.7 percent.

Attempts to improve the efficiency of this condenser coil and to use it for collecting atmospheric radon samples proved to be unsuccessful. The porous copper plug condenser, described previously, was then constructed. After this condenser had been installed in the system, data were again taken with the standard radium solution providing the radon. At all flow rates up to 1.5 liters/minute, for 1 hour collection periods, the efficiency of the system was essentially 100 percent. Typical results are listed in Table 2. On the basis of these data, a flow rate of 1.5 liters per minute and a collection period of 1 hour were taken as standard parameters for sample collections.

Table 2. Overall efficiency measurements.

Flow rate	Collection time	Calculated ion current	Measured ion current	Overall efficiency
ltrs/min	hours	10^{-14} amp	10^{-14} amp	percent
1.5	0.50	36	36	100
1.5	0.75	54	54	100
1.5	1.00	72	72	100
1.5	1.00	72	72	100

The calibration factor for the apparatus may now be calculated. For each collection of Table 2, the value for the calculated ion current is converted into the equivalent quantity of radon. The ratio of the measured value of the ion current to the calculated quantity of radon then provides the calibration factor for each collection. Since the system

is shown to be essentially 100 percent efficient, the above ratios give
identical values for each case. It was found that:

10^{-14} amp corresponds to $10.1 \cdot 10^{-12}$ curie of radon.   (4)

The calibration is expressed in this manner because a current unit of
10^{-14} amp proved to be both convenient and practical.

The largest error in the results involves the determination of the
initial value of the ion current. This uncertainty results from two
causes: the inability to fill the ion chamber instantaneously, and the
statistical fluctuations in the initial radon decay rate. In order to
minimize these effects and to increase the accuracy of the results, a
scheme was devised whereby the initial value of the ion current was
obtained by averaging the recorded current over the first 15 minutes of
operation. To obtain the standard deviation of a continuous observation
of a statistical source, the method of Schiff and Evans was followed.[10]
Appendix II contains a description of the averaging scheme used to find
the initial value of the ion current, and an error analysis that gives
the standard deviation of this value. It is shown that for an initial
ion current of 10^{-14} amp averaged over a continuous observation period
of 15 minutes, the standard deviation is ± 5.4 percent. (The standard
deviation decreases for higher values of the initial current). The
accuracy of the results of this investigation is limited to ± 5 percent

10. L. I. Schiff and R. D. Evans, "Statistical analysis of the
counting rate meter," Review of Scientific Instruments, 7, 1936,
pp 456-462.

in order to allow for a possible inefficiency of the "lid" during sample collections.

Identification. In order to remove any doubt that the activity detected was due to radon, the build-up curves of the activity from the samples were compared with the theoretical build-up curve for radon and its daughters as given by Israel.[11] (Figure 5). After secular equilibrium of the activity was reached, as indicated by the recorded current, the decay curves of the collected samples were compared with the theoretical decay curve of radon. (Figure 6). The close fit of the activity data of the collected samples with the theoretical curves (solid lines) of Figures 5 and 6 identifies the samples as radon.

RESULTS

Atmospheric radon samples were taken through Tygon tubing with the open end of the tubing placed 0.8 meter above the ground level. Measurements of the radon flux were taken with the "lid" placed on the ground just south of Weir Hall on the campus of New Mexico Institute of Mining and Technology. Approximately six weeks before collections were to be made, a small area of the soil surface at this location was loosened to a depth of about three inches. All coarse stones and pebbles were removed in an attempt to make the first few inches of the soil homogeneous. The surface then remained undisturbed until the collection "lid" was placed in

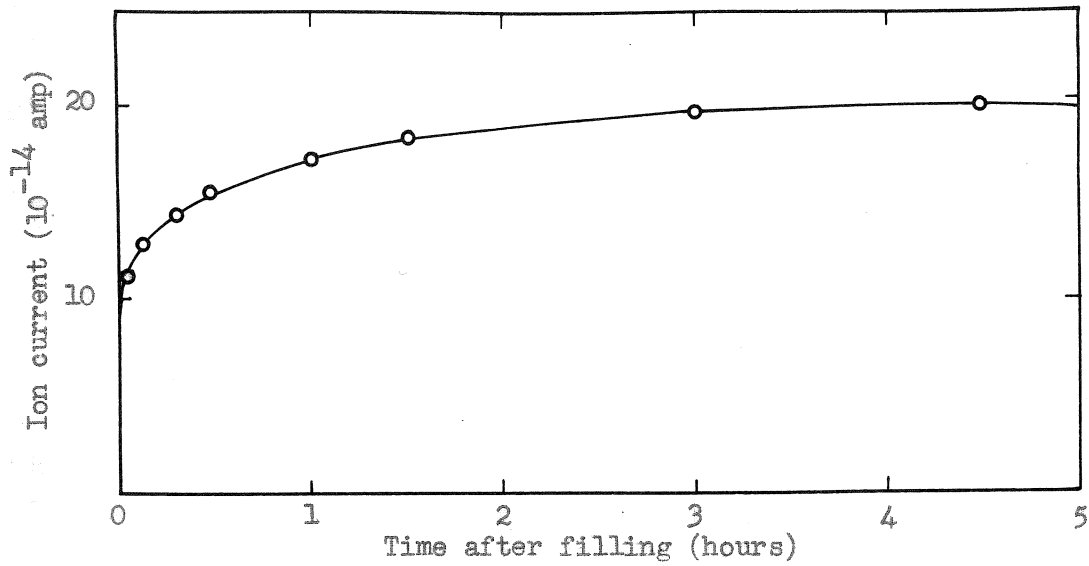11. H. Israel, op. cit., p 156.

Figure 5. Comparison of experimental data with the theoretical activity of radon and its daughters.
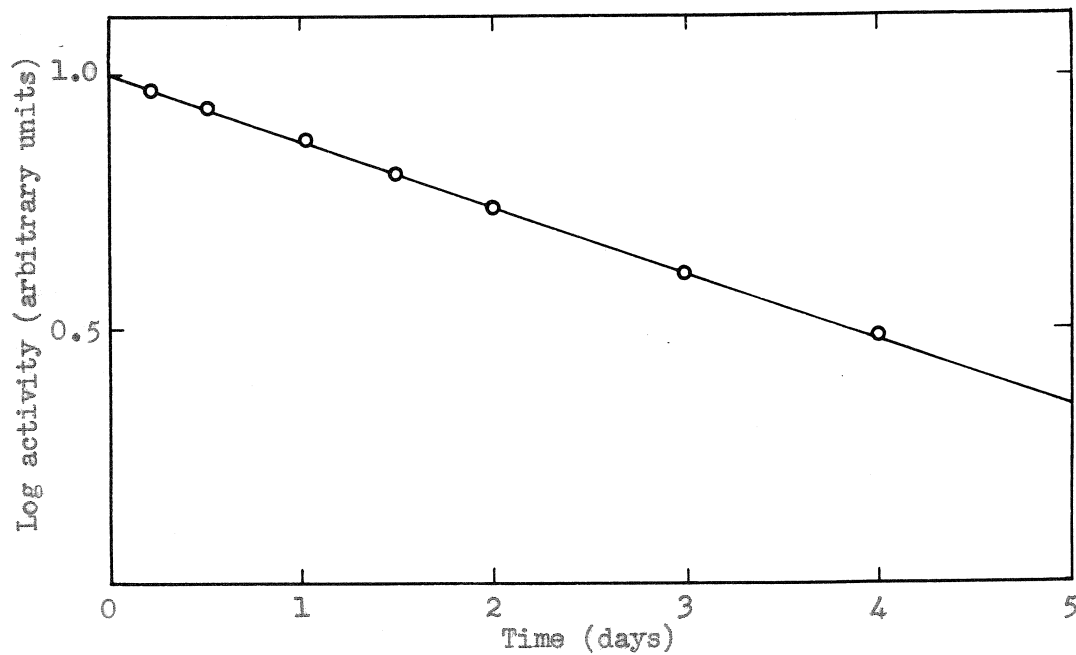

Figure 6. Comparison of experimental data with the theoretical radon decay curve.

position.

Information supplied by Dr. R. H. Weber of the New Mexico State
Bureau of Mines and Mineral Resources indicates that the soil of the
campus area is derived mostly from Tertiary rhyolitic and latitic detritus
that has been transported eastward from the Socorro Peak area. Small
quantities of andesitic constituents may be present; the basaltic content
is negligible. In general, the equivalent uranium content varies from
about .001 percent for the basalts up to about .004 percent for the rhyo-
lites.

Data. With these sampling and environmental conditions, the atmos-
pheric and exhalation, or flux, data of Table 3 were obtained. Figure 7
is a plot of the atmospheric radon for Saturday, December 28, 1957. An
early morning maximum is clearly shown. The weather over that week-end
was warm and windy through the day, becoming calm and cold at night. A
strong column of smoke north of town indicated the presence of an atmos-
pheric inversion layer extending to perhaps 1000 feet above the terrain.
During the following week, several inches of snow fell, and no data were
taken. The ground was open through January 10 and 11, 1958, and radon
measurements were again made. The data taken during that time are plotted
in Figures 8 and 9. The data cover a period of 24 hours for both the
atmospheric and exhalation radon.

Discussion. The general purpose of the investigation on January 10
and January 11 was (1) to confirm the early morning atmospheric radon
peak; (2) to examine the behavior of the radon flux over a 24 hour period;
and (3) to calibrate the continuous atmospheric radon monitor by operating

Table 3. Experimental atmospheric radon and flux data.

Date	Type of collection	Mean time	Total radon collected	Exhalation or flux	Atmospheric concentration
		MST	10^{-12} curie	$10^{-18} \dfrac{\text{curie}}{\text{cm}^2\text{-sec}}$	$10^{-13} \dfrac{\text{curie}}{\text{liter}}$
Dec. 28 1957	Atmosphere	0215	20		2.3
		0331	28		3.1
		0454	32		3.5
		0603	43		4.8
		0714	37		4.1
		1225	24		2.7
	Exhalation	0839	124	126	
		0955	66*	49	
		1110	101	105	
Dec. 29 1957	Atmosphere	0921	49		5.4
	Exhalation	1031	117	100	
		1209	183**		
Jan. 10 1958	Atmosphere	1947	34		3.8
Jan. 11		2213	53		5.9
		0035	59		6.6
		0429	57		6.4
		0650	55		6.1
		0910	53		5.9
		1530	6.1		0.68
Jan. 10	Exhalation	2102	101	81	
		2324	107	74	
		0541	103	69	
		0800	109	81	
		1020	87	77	
		1421	57	73	
		1755	59	74	

*The "lid" was moved to a hard, compacted surface for this collection.

**This collection covered a two hour period. The purpose was to collect as much radon as possible, so that the activity could be compared to the activity of a radon standard.
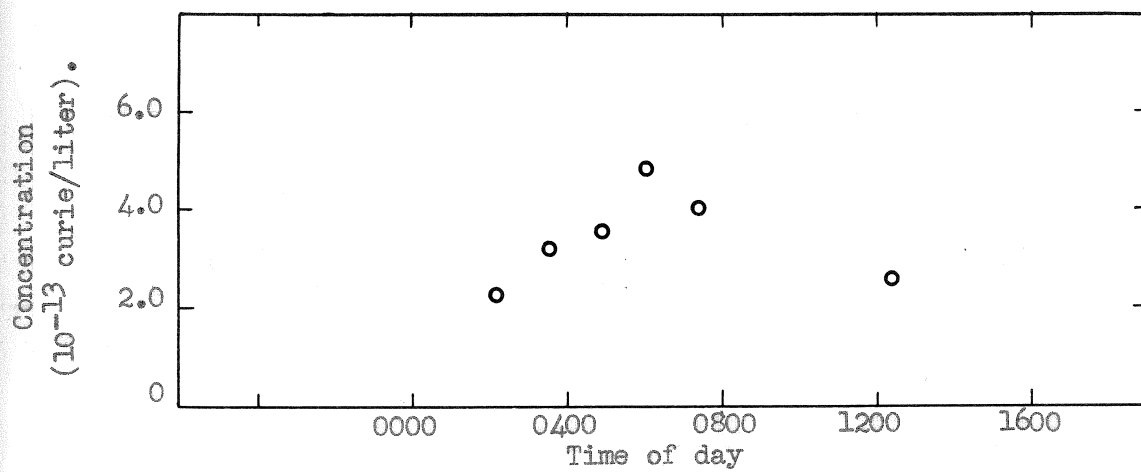
Figure 7. Atmospheric radon concentrations for December 29, 1957.
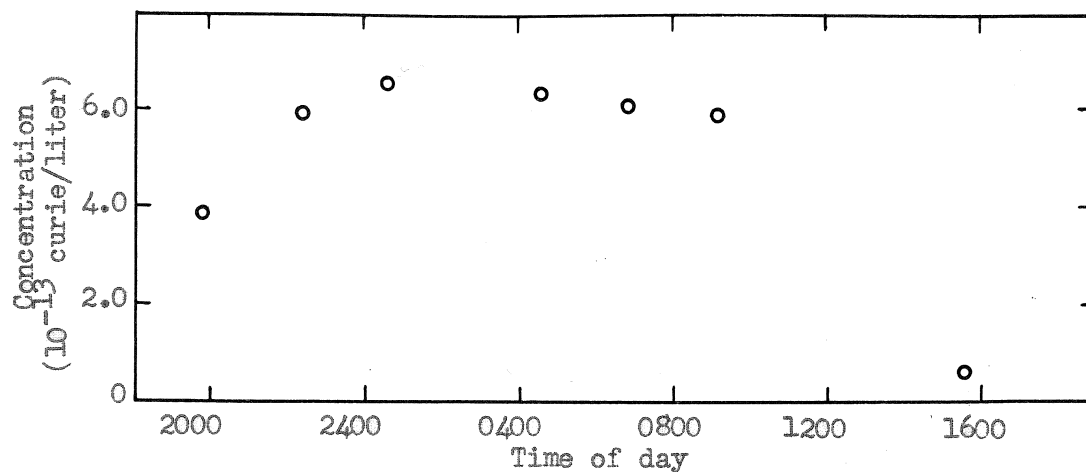
Figure 8. Atmospheric radon concentrations for
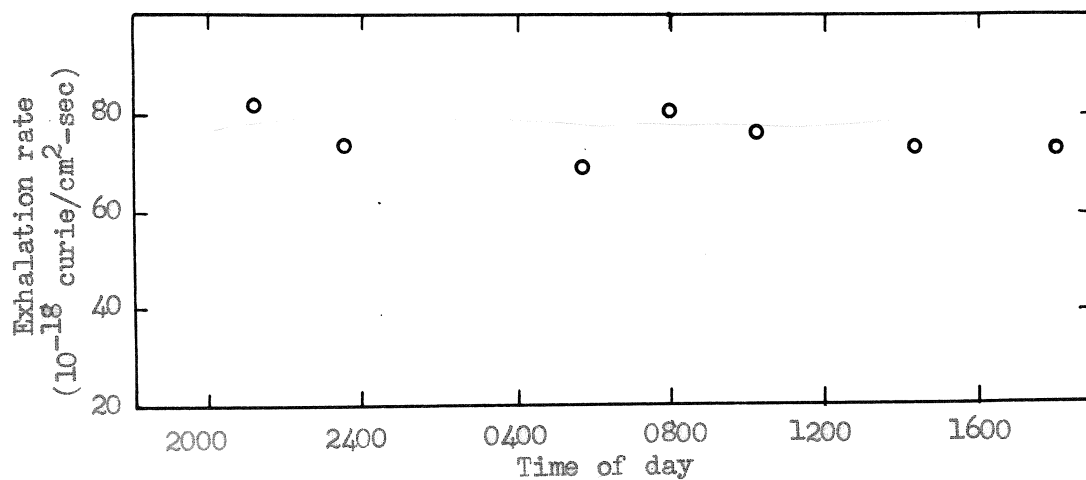January 10 and 11, 1958.

Figure 9. Radon flux at the earth-air interface for
January 10 and 11, 1958.

the monitor and the emanometric apparatus simultaneously.[12] Figure 8 again shows a diurnal variation of the atmospheric radon, with the build-up beginning in the early evening and the peak rapidly disappearing when the inversion was broken in the morning. Slight air motions were detectable beginning about 0200 Saturday. These persisted at intervals until a well-defined cold front with strong winds arrived at 1230.

The exhalation data for January 10 and 11 indicate that the radon flux was relatively constant over a diurnal period, even though high winds persisted throughout the afternoon. The variation in the flux data of December 28 suggests that (1) either there is a noticeable effect of the "lid" geometry, or (2) the first flux value obtained on this date includes some radon that had accumulated in the "lid". It is also possible that both of these effects were present. Comparing the flux values of December 28 and 29 with those of January 10 and 11 suggests that long-period fluctuations of the flux occur.

It has been shown by Wilkening[13] that within any volume element of the atmosphere at the ground level, radioactive equilibrium usually exists between radon and its daughters. Since the emanometric apparatus measures only the radon and the continuous monitor measures only the radon daughters, this result permits the data from the two methods of measurement to be related and gives a calibration factor for the continuous monitor. The calibration was obtained by simultaneous operation of the two units

---

12. M. H. Wilkening, "Monitor for natural radioactivity in the atmosphere," Nucleonics, 10, 1952, pp 36-39.

13. M. H. Wilkening, Private communication.

during the measurements of January 10 and 11. It was found on the average that 100 counts-per-minute on the monitor corresponded to a radon concentration of $2.8 \cdot 10^{-13}$ curie/liter. Wilkening's observations of the ground-level atmospheric radon content with the continuous monitor cover a period of several years. His mean value, when evaluated from the present calibration, indicates that the average concentration of atmospheric radon at Socorro is $2.3 \cdot 10^{-13}$ curie/liter.

The mean value of the radon flux for the Socorro area is taken as $90 \cdot 10^{-18}$ curie/cm^2-sec (Table 3). The local values of the radon flux and the atmospheric radon concentration are listed in Table 4 together with a few representative values of these quantities from the literature.[14] Table 4 shows that both the atmospheric radon concentration, N_0, and the radon flux at the earth-air interface, J, vary considerably over a world-wide scale. The mean value of N_0 given by Israel is the result of some 2200 observations at 19 scattered localities, most of which are in Europe. In the southwestern United States, higher concentrations of atmospheric radon are to be expected than those concentrations found at the island stations and coastal cities listed in Table 4. For Socorro, the average value of the atmospheric concentration is close to Israel's mean value. The wide variations noted in the flux values illustrate the effect of different soil compositions to a greater extent than do the values of N_0 because the flux suffers no dilution from ocean or sea breezes. (Compare N_0 and J for Innsbruck with N_0 and J for Manila).

---

14. H. Israel, *op. cit.*, p 158.

Table 4. Representative atmospheric and exhalation radon measurements.

Name	Place	Mean value

ATMOSPHERIC RADON CONCENTRATION, N_0

		$10^{-13} \frac{curie}{liter}$
Hess	New York, 1942	0.97
Schwalb	Bad Nauheim, 1935	5.85
Illing	Innsbruck, 1933	4.36
Wright and Smith	Manila, 1912-14	0.71
Ashman	Chicago, 1908	0.95
Israel	World-wide mean	2.53
Hand and Wilkening	Socorro, 1958	2.3

RADON EXHALATION, J

		$10^{-18} \frac{curie}{cm^2-sec}$
Smythe	Dublin	74
Wright and Smith	Manila	21
Zupancic	Innsbruck	23
Zeilinger	Innsbruck	50
Kosmath	Leibenau/Graz	40
Israel	Mean value	40
Hand	Socorro	90

Although the mean value of J for Socorro is higher than the given mean value, this difference can be attributed to either a higher-than-normal uranium content of the local soil or to an insufficient quantity of exhalation data.

Correlation of flux and atmospheric radon. In order to correlate the

flux and the atmospheric radon, a simplified description of the migration of radon through the soil, across the earth-air interface, and into the atmosphere will be constructed. The following assumptions are made in order to idealize the soil and atmospheric conditions:

1. Uniform soil composition extending to a depth of several meters.

2. Constant and homogeneous production of radon within any volume element of the soil for a depth of several meters.

3. The net diffusion of radon occurs only in the positive x-direction.

4. Meteorological conditions have no effect on the movement of radon within the soil.

5. Normal eddy diffusion determines the vertical distribution of radon in the atmosphere.

The following symbols are used:

D_- (cm^2/sec)	= Radon diffusion coefficient in the soil.
P (atoms/cm^3-sec)	= Radon production rate in the soil.
D_+ (cm^2/sec)	= Coefficient of eddy diffusion in the atmosphere.
J_+ (atoms/cm^2-sec)	= Net radon flux in the atmosphere.
J_- (atoms/cm^2-sec)	= Net radon flux in the soil.
C (atoms/cm^3)	= Radon concentration in the soil.
C_1 (atoms/cm^3)	= Radon concentration of the soil at depth.
C_0 (atoms/cm^3)	= Radon concentration of the soil at the surface.
N (atoms/cm^3)	= Radon concentration of the atmosphere.
N_0 (atoms/cm^3)	= Radon concentration of the atmosphere at the ground level.

Since the net diffusion is considered to occur only in one direction, the x-coordinate is taken perpendicular to the surface of the earth. The

surface of the earth is then represented by the plane x = 0; the positive
x-direction extends into the atmosphere and the negative x-direction
extends downward into the earth.

Consider first the distribution of radon in the soil. Within any
volume element, diffusion, production, and decay of the radon occurs
continuously. It is assumed that these processes have been in operation
for a period of time sufficient for steady-state conditions to be estab-
lished. In general, the behavior of the radon within a volume element
can be expressed by

$$\frac{\partial C}{\partial t} = \text{Production} - \text{Leakage} - \text{Decay},$$

and at the steady state,

$$\text{Production} = \text{Leakage} + \text{Decay}. \qquad (5)$$

It is assumed further that Fick's law of diffusion describes the movement
of the radon in both the soil and the atmosphere. In each environment,
the proper diffusion coefficient is applied. In the present notation,
Fick's law for diffusion in the soil becomes:

$$J_- = - D_- \left(\frac{\partial C}{\partial x} \right).$$

The negative sign on the right indicates that the net diffusion is taking
place in a direction opposite to the direction of increasing radon concen-
tration. The production is denoted by P and the decay by λC. The leakage
term is given as the gradient of the flux, $- D_- \left(\frac{\partial^2 C}{\partial x^2} \right)$. Upon substituting
these quantities into equation 5 the steady-state relation becomes:

$$D_-\left(\frac{\partial^2 C}{\partial x^2}\right) + P - \lambda C = 0. \tag{6}$$

Assuming that P and D_- are constant, equation 6 is solved by applying the boundary conditions:

$$\text{as } -x \to \infty, \quad C = C_1$$
$$-x \to 0, \quad C = C_0$$

giving the solution:

$$C = \left(C_0 - \frac{P}{\lambda}\right)e^{\sqrt{\frac{\lambda}{D_-}}x} + \frac{P}{\lambda}, \tag{7}$$

where x takes only negative values. As $x \to -\infty$, $C = C_1 = \frac{P}{\lambda}$, and equation 7 reads, after slight readjustment,

$$C = C_1\left(1 - e^{\sqrt{\frac{\lambda}{D_-}}x}\right) + C_0 e^{\sqrt{\frac{\lambda}{D_-}}x}. \tag{8}$$

Equation 8 describes the distribution of radon within the soil.

Substituting equation 8 into Fick's law and evaluating the flux at the plane $x = 0$, as x approaches this plane from the negative direction, gives:

$$J_- = (D_- \lambda)^{\frac{1}{2}}(C_1 - C_0). \tag{9}$$

In general, C_0 is small compared to C_1, so that C_0 can be neglected. Evaluating the diffusion coefficient of radon in the soil on the basis of experiments reported in the literature,[15-17] gives $D_- = .072 \ cm^2/sec.$ Considering that only 5-10 percent of the radon generated within the soil

escapes into the soil capillaries where diffusion can take place,[18] the radium content of the local soil is obtained (to an accuracy of about 50 percent) from equation 9:

$$\text{Radium content} = 1.1 \cdot 10^{-12} \frac{\text{gm Ra}}{\text{gm soil}},$$

using $90 \cdot 10^{-18}$ curie/cm^2-sec for J.

For the atmospheric radon distribution, $P = 0$. From considerations similar to those used in deriving the soil equation, the steady-state equation for radon in the atmosphere is:

$$\frac{\partial^2 N}{\partial x^2} - \frac{\lambda N}{D_+} = 0. \tag{10}$$

Equation 10 is solved with the following boundary conditions:

$$\text{as } x \to \infty, \quad N \to 0$$

$$x \to 0, \quad N = N_0$$

which gives the vertical distribution of radon in the atmosphere:

$$N = N_0 e^{-\sqrt{\frac{\lambda}{D_+}}\, x}, \tag{11}$$

15. C. M. H. van Bavel, "Gaseous diffusion and porosity in porous media," Soil Science, 73, 1952, pp 73-80

16. W. Jost, Diffusion, Academic Press, Incorporated, New York, 1952, pp 412-415.

17. D. DeVries, "Some remarks on gaseous diffusion in soils," Transactions of the 4th International Congress of Soil Science, 2, 1950, p 41.

18. B. B. Boltwood, "On the radioactivity of uranium minerals," American Journal of Science, 25, 1908, pp 269-298.

where x takes only positive values. Again applying Fick's law of diffusion and evaluating the result at the plane x = 0, as x approaches this plane from the positive direction, one gets:

$$J_+ = (D_+ \lambda)^{\frac{1}{2}} N_o.$$ (12)

Since the radon flux crossing the plane x = 0 must be the same on both positive and negative sides of this plane, the expressions for the flux, J_- and J_+, are equated to give:

$$D^{\frac{1}{2}}(C_1 - C_o) = D_+^{\frac{1}{2}} N_o.$$ (13)

Substituting $D_+ = 7 \cdot 10^4$ cm^2/sec,[19] $D_- = .072$ cm^2/sec, and using the average value of N_o for the Socorro area, equation 13 gives the same result for the radium content of the local soil as did equation 9:

$$\text{Radium content} = 1.1 \cdot 10^{-12} \frac{\text{gm Ra}}{\text{gm soil}}.$$

(Again this figure is accurate only to about 50 percent). The two values for the radium content of the soil show complete agreement, yet are based on two independent measurements. In addition, the value of the radium content agrees closely with the values presented in the literature[20,21]

19. M. H. Wilkening, "Variation of natural radioactivity in the atmosphere with altitude," Transactions of the American Geophysical Union, 37, 1956, pp 177-190.

20. F. E. Sentfle and N. B. Keevil, "Thorium-uranium ratios in the theory of genesis of lead ores," Transactions of the American Geophysical Union, 28, 1947, pp 732-738.

21. H. Faul, op. cit., p 156.

for rock types similar to those of the local area.

Returning to the expression for the radon distribution in the atmosphere (equation 11) the total number of radon atoms present in a column of the atmosphere having a cross-section area of 1 cm^2 is obtained by integrating equation 11 from $x = 0$ to $x = \infty$. Converting N_o for the Socorro area ($2.3 \cdot 10^{-13}$ curie/liter) to radon atoms/cm^3 and using $D_+ = 7 \cdot 10^4$ cm^2/sec, the integration gives:

$$N_{total} = 7.4 \cdot 10^5 \frac{radon\ atoms}{cm^2} .$$

From this quantity of radon, the number of radon atoms lost per unit time due to decay is:

$$\lambda N_{total} = 1.6 \frac{radon\ atoms}{cm^2\text{-}sec} .$$

The quantity of radon entering the atmosphere is found by converting the flux value to radon atoms/cm^2-sec. The rate of entry is:

$$J = 1.6 \frac{radon\ atoms}{cm^2\text{-}sec} ,$$

using $90 \cdot 10^{-18}$ curie/cm^2-sec for J. Thus, within the accuracy of the experiment, the rate of radon entry into the atmosphere balances the rate of radon decay in the atmosphere. Considering that the radon in the atmosphere is in equilibrium with its daughters, the above result implies that the activity in the atmosphere from each of the radon daughters is also 1.6 $\frac{disintegrations}{cm^2\text{-}sec}$.

SUMMARY AND CONCLUSIONS

The presence of radon in the atmosphere has been verified by comparing the time rate of change in the activity of collected samples with that of known radon standards. The average radon concentration of the atmosphere at the ground level has been quantitatively measured by the method of emanometry. This method was also employed to measure the radon flux at the earth-air interface. For the Socorro area, these measurements indicate that the average ground-level atmospheric radon concentration is $2.3 \cdot 10^{-13}$ curie/liter and the average radon flux is $9 \cdot 10^{-17}$ curie/cm^2-sec. These data were obtained with an accuracy of ± 5 percent and compare favorably with the world-wide averages reported in the literature. The atmospheric radon concentration was shown to undergo a diurnal variation, whereas the flux appeared to be relatively constant over a diurnal period.
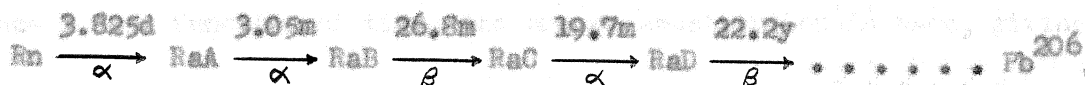
When these data are applied to calculations based on idealized models of the soil and atmosphere, it is found that the radon observed in the atmosphere can be accounted for by the radon flux at the earth-air interface. It is also indicated that the radium content of the local soil is about $1 \cdot 10^{-12}$ gm Ra/gm soil. This result, although containing a large uncertainty, again compares favorably with values found in the literature.

It is suggested that future studies include an investigation of the subsurface radon distribution, since the present equipment and apparatus can be readily adapted to that purpose. The utility of the apparatus can be greatly increased if the collection section is designed as a portable unit.

APPENDIX I

Build-up of activity within the ion chamber

To obtain an insight into the build-up of activity as a result of an initial quantity of radon undergoing radioactive decay, consider a chamber containing an initial quantity of radon, R_o. At time $t = 0$, radon is the only radioactive isotope present in the chamber. As time passes, the following reactions occur within the chamber:

$$Rn \xrightarrow[\alpha]{3.825d} RaA \xrightarrow[\alpha]{3.05m} RaB \xrightarrow[\beta]{26.8m} RaC \xrightarrow[\alpha]{19.7m} RaD \xrightarrow[\beta]{22.2y} \cdots \cdots Pb^{206}.$$

The activity observed in the chamber is due not only to the decay of the radon, but also to the build-up and decay of the daughters RaA, RaB, and RaC. The RaD decays with a half-life of 22.2 years, so that the contributions to the total activity from this daughter (and all succeeding daughters) can be neglected.

Denoting the number of atoms of each of the daughters that contribute to the observed activity by

$$\begin{aligned}
A &= \text{Number of atoms of RaA} \\
B &= \quad " \quad " \quad " \quad " \quad RaB \\
C &= \quad " \quad " \quad " \quad " \quad RaC
\end{aligned}$$

and the number of atoms of radon by R, the following system of equations describing the production and decay of each daughter must be solved:

$$R = R_0 e^{-\lambda_R t} \qquad\qquad 1.$$

$$\frac{dA}{dt} = \lambda_R R - \lambda_A A \qquad\qquad 2.$$

$$\frac{dB}{dt} = \lambda_A A - \lambda_B B \qquad\qquad 3. \qquad\qquad (I)$$

$$\frac{dC}{dt} = \lambda_B B - \lambda_C C \qquad\qquad 4.$$

where λ_R, λ_A, λ_B and λ_C are the decay constants of radon, RaA, RaB, and RaC respectively.

Starting with 2:

$$\frac{dA}{dt} + \lambda_A A = \lambda_R R.$$

Since R is a function of time, the proper substitution is made, giving:

$$\frac{dA}{dt} + \lambda_A A = \lambda_R R_0 e^{-\lambda_R t}.$$

It can be shown that the solution of this equation is

$$A = \frac{R_0 \lambda_R}{(\lambda_A - \lambda_R)} \left[e^{-\lambda_R t} - e^{-\lambda_A t} \right]. \qquad\qquad (II)$$

The solutions for the abundance of the daughters RaB and RaC have the forms:

$$B = R_0 \left[\frac{\lambda_A \lambda_R\, e^{-\lambda_A t}}{(\lambda_R - \lambda_A)(\lambda_B - \lambda_A)} + \frac{\lambda_A \lambda_R\, e^{-\lambda_R t}}{(\lambda_A - \lambda_R)(\lambda_B - \lambda_R)} + \frac{\lambda_A \lambda_R\, e^{-\lambda_B t}}{(\lambda_R - \lambda_B)(\lambda_A - \lambda_B)} \right] \qquad (III)$$

$$C = R_0 \left[a e^{-\lambda_R t} + b e^{-\lambda_A t} + c e^{-\lambda_B t} + d e^{-\lambda_C t} \right], \qquad\qquad (IV)$$

where

$$a = \frac{\lambda_R \lambda_A \lambda_B}{(\lambda_A - \lambda_R)(\lambda_B - \lambda_R)(\lambda_C - \lambda_R)}$$

$$b = \frac{\lambda_R \lambda_A \lambda_B}{(\lambda_R - \lambda_A)(\lambda_B - \lambda_A)(\lambda_C - \lambda_A)}$$

$$c = \frac{\lambda_R \lambda_A \lambda_B}{(\lambda_R - \lambda_B)(\lambda_A - \lambda_B)(\lambda_C - \lambda_B)}$$

$$d = \frac{\lambda_R \lambda_A \lambda_B}{(\lambda_R - \lambda_C)(\lambda_A - \lambda_C)(\lambda_B - \lambda_C)}$$

Bateman has given the symmetric solution for equations of this type for the nth product.[*]

If the time derivatives of these equations are taken and their sum computed, one obtains the total activity in the chamber as a function of time resulting from an initial quantity of radon, R_0, in the chamber. Israel[**] has computed the value of this activity as a function of time; plotting his data gives the solid curve of Figure 5.

---

[*] H. Bateman, "The solution of a system of differential equations occurring in the theory of radioactive transformations," _Proceedings of the Cambridge Philosophical Society_, 15, 1910, pp 423-427.

[**] H. Israel, _op. cit._, p 156.

APPENDIX II

Determination of the initial ion current and statistical error analysis.

The largest uncertainty in the experiment arises in the determination of the initial ion current. Since only the quantity of radon admitted into the ion chamber is of interest, the initial current must be determined in such a manner that the current generated by the decay of the first radon daughter (3.05 minute half-life) is excluded. The chamber filling time was usually about one minute, which meant that the initial value of the current could not definitely be discerned. To overcome this difficulty and to increase the accuracy of the results, the following scheme was used: Equations I, II, III, and IV of Appendix I show that the activity of radon and its daughters at any given time is the product of an appropriate constant and the initial activity of the radon alone. Since the ion current is proportional to the activity, the current at any given time will also be the product of some appropriate factor and the initial current value. Therefore, if these multiplying factors are evaluated for various times from the above equations, any arbitrary value for the initial ion current can be chosen and the corresponding current-versus-time curve constructed. Using appropriate values for the initial ion currents, build-up curves of the activity were plotted on Esterline-Angus recorder chart paper and templates of the curves were cut from stiff

cardboard. These templates then were matched to the activity curves of the samples over the first 15 minutes of the recorded current. Using the template that gave the best fit to the average current over this period of time, the initial current value was obtained by extrapolating the average current back to the mean filling time of the chamber. This gave, in effect, a time unit of 15 minutes upon which to base the value of the initial current.

To determine the statistical error present in the data, the method outlined by Schiff and Evans was followed.[*] The activity of the collected radon samples was determined by integration of the individual pulses within the ion chamber; in order to analyze the statistics involved, the parameters of the electrometer input circuit had to be determined (Figure 10). The resistance, R, was set by the range switch on the electrometer box; R was 10^{11} ohms for all data taken. The total capacitance of the circuit was measured between the outer wall and the central electrode of the ion chamber. This measurement was made with a Beco impedance bridge, with an oscilloscope used as an external detector to indicate the null point of the bridge. Several measurements indicated that the total capacitance of the input circuit was 60 mmf. For purposes of the error analysis calculations, the system is represented by a condenser shunted with a resistor, the combination having a time constant of 6 seconds. The arrangement is such that a charge q is placed on the condenser each time a pulse is

---

* L. I. Schiff and R. D. Evans, "Statistical analysis of the counting rate meter," Review of Scientific Instruments, 7, 1936, pp 456-462.
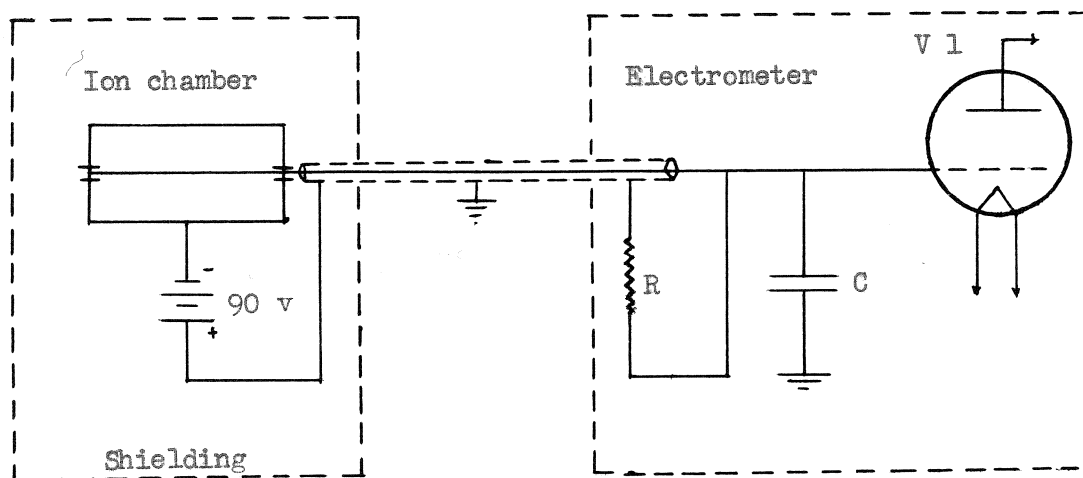
Figure 10. Electrometer input circuit.

received by the system.

It is first assumed that the average number of pulses received per unit time has the constant value x. The pulses are considered to arise at random from a source of constant strength; thus they are distributed in time according to Poisson's law.[*] The number expected during the time interval t to t + dt is

$$xdt,$$

and the expected charge increment of the condenser is

$$q(xdt).$$

If the condenser charge is read at some later time, t_o, it is found that the charge has decayed to

$$qxe^{-(t_o - t)/RC}dt.$$

Assuming that the system has been operating for a period of time, long when compared to RC ($\equiv$ 6 sec), the expected value of the charge, Q, on the condenser at the time of the reading, t_o, is given by integrating over the time of operation up to t_o. This means that the lower limit of the integration is essentially negative infinity; thus

$$Q = \int_{-\infty}^{t_o} qxe^{-(t_o - t)/RC}dt = qxRC \tag{I}$$

To find the standard deviation of the reading, the fact is used that for

---

* F. Rasetti, Elements of Nuclear Physics, Prentice Hall, Incorporated, New York, 1936, pp 32-35.

randomly occurring events obeying Poisson's distribution, the standard deviation of N observed events is $N^{\frac{1}{2}}$. For the present case, the standard deviation of the number of pulses arriving in the time interval dt is

$$(xdt)^{\frac{1}{2}},$$

and the standard deviation of the charge placed on the condenser for this interval is

$$q(xdt)^{\frac{1}{2}}.$$

The contribution of this deviation to the charge Q on the condenser at time t_o is

$$q(xdt)^{\frac{1}{2}}e^{-(t_o- t)/RC}.$$

Considering that the time intervals, dt, are independent of each other, the contribution from each is independent of the others over the total time considered. To obtain the variance of the entire reading, the squares of the individual contributions are integrated over the time period under consideration. The variance of the total condenser charge, Q, at time t_o is then:

$$(\delta Q)^2 = \int_{-\infty}^{t_o} q^2xe^{-2(t_o- t)/RC}dt,$$

giving:

$$(\delta Q)^2 = \frac{q^2xRC}{2}. \tag{II}$$

The standard deviation is:

$$S. D. = \delta Q = q \left[\frac{xRC}{2} \right]^{\frac{1}{2}}$$

and the fractional standard deviation of the reading is:

$$\sigma_1 = \frac{\delta Q}{Q} = \frac{q(xRC/2)^{\frac{1}{2}}}{qxRC}$$

$$\sigma_1 = (2xRC)^{-\frac{1}{2}} \qquad\qquad (III)$$

This is the standard deviation of a single observation. The effect of the averaging period on the standard deviation must next be determined. Schiff and Evans develop the expression for the standard deviation of continuous observations by first considering the statistical fluctuations for n observations taken at times of ΘRC apart. ($\Theta = 1, 2, 3, \ldots k$). For the fractional standard deviation of the mean to be a minimum, it is shown that n must become large as Θ becomes small, such that $n\Theta = $ constant and the condition

$$n\Theta RC = T$$

is satisfied, where T is the time available for the n observations. Considering continuous observations, the above conditions imply that:

$$n \to \infty, \text{ and } \Theta \longrightarrow 0$$

subject to the restriction that

$$n\Theta = \frac{T}{RC}.$$

The fractional standard deviation of the mean then is shown to be

$$\sigma_T = \frac{(1 - 2T/RC)^{\frac{1}{2}}}{(1 - T/RC)} \cdot \sigma_1 \qquad \text{(IV)}$$

where T = averaging period and σ_1 is the standard deviation of a single observation.

The present interest lies in the standard deviation of the initial activity in the ion chamber. Since a current of 10^{-14} amp corresponds to an activity of .376 $\frac{\text{disintegrations}}{\text{sec}}$, σ_1 can be evaluated for this value of the initial current:

$$\sigma_1 = (2 \times RC)^{-\frac{1}{2}}$$

$$\sigma_1 = \frac{1}{(.376 \cdot 2 \cdot 6)^{\frac{1}{2}}}$$

$$\sigma_1 = .470$$

Evaluating equation IV above for this value of σ_1 and an averaging period T, of 15 minutes gives:

$$= \frac{(1 - 2 \cdot 15 \cdot 10)^{\frac{1}{2}}}{(1 - 15 \cdot 10)}(.470)$$

$$= (.115)(.470) = .054$$

or expressed as a percentage error, this becomes:

$$\text{Percentage error} = 5.4\%.$$

The computation of the error based on an initial current of 10^{-14} amp

represents the worst possible case, for in all but one of the observations of atmospheric and exhalation radon taken during this investigation the initial current was larger than 10^{-14} amp. The above error figure of 5.4 percent, consequently, is an over-estimate of the uncertainty in the results.

BIBLIOGRAPHY

BOOKS

J. Paul, _Nuclear Geology_, John Wiley and Sons, Incorporated, New York, 1954.

R. Geiger, _The Climate Near the Ground_, Harvard University Press, Cambridge, Massachusetts, 1950.

W. Jost, _Diffusion_, Academic Press, Incorporated, New York, 1952.

K. Rankama and T. G. Sahama, _Geochemistry_, The University of Chicago Press, Chicago, 1950.

F. Rasetti, _Elements of Nuclear Physics_, Prentice Hall, Incorporated, New York, 1936

E. Rutherford, _Radioactive Substances and Their Radiations_, Cambridge University Press, Cambridge, 1912.

E. Rutherford, _Radiations From Radioactive Substances_, Cambridge University Press, Cambridge, 1930.

W. Schmidt, _Eddy Diffusion in the Free Air and Related Phenomena_, Hamburg, H. Grand, 1925.

PERIODICALS

G. C. Ashman, "A quantitative determination of the radium emanation in the atmosphere," _American Journal of Science_, 26, 1908, pp 119-129.

H. Bateman, "The solution of a system of differential equations occurring in the theory of radioactive transformations," _Proceedings of the Cambridge Philosophical Society_, 15, 1910, pp 423-427.

C. M. H. van Bavel, "Gaseous diffusion and porosity in porous media," _Soil Science_, 73, 1952, pp 73-80.

B. B. Boltwood, "On the radioactivity of uranium minerals," American Journal of Science, 25, 1908, pp 269-298.

R. A. Boyle, "The volatilization of radium emanation at low temperatures," Philosophical Magazine, 20, 1910, pp 867-875.

H. A. Bumstead and L. P. Wheeler, "On the properties of a radioactive gas found in the soil and water near New Haven," American Journal of Science, 17, 1904, pp 77-111.

J. Elster and H. Geitel, "Beschreibung des Verfahrens zur gewinning vertübergehend radioaktiver Stoffe aus der atmosphärischen Luft," Zeitschrift für Physik, 3, 1902, pp 305-310.

A. S. Eve, "On the amount of radium emanation in the atmosphere near the earth's surface," Philosophical Magazine, 10, 1905, pp 98-107.

V. F. Hess, "On the radon content of the atmosphere and the radium content of river water," Terrestrial Magnetism and Atmospheric Electricity, 48, 1943, pp 203-206.

H. Israel, "Radioactivity of the atmosphere," Compendium of Meteorology, American Meteorological Society, Boston, Massachusetts, 1951, pp 155-162.

E. M. Kovach, "An experimental study of the radon-content of soil-gas," Transactions of the American Geophysical Union, 25, 1944, pp 563-571.

J. Satterly, "On the amount of radium emanation in the lower regions of the atmosphere and its variation with the weather," Philosophical Magazine, 20, 1910, pp 1-15.

L. I. Schiff and R. D. Evans, "Statistical analysis of the counting rate meter," Review of Scientific Instruments, 7, 1936, pp 456-462.

F. E. Sentfle and N. B. Keevil, "Thorium-uranium ratios in the theory of genesis of lead ores," Transactions of the American Geophysical Union, 28, 1947, pp 732-738.

L. B. Smythe, "On the supply of radium emanation from the soil to the atmosphere," Philosophical Magazine, 24, 1912, pp 632-637.

D. A. DeVries, "Some remarks on gaseous diffusion in soils," Transactions of the 4th International Congress of Soil Science, 2, 1950, pp 41-45.

M. H. Wilkening, "Monitor for natural radioactivity in the atmosphere," Nucleonics, 10, 1952, pp 36-39.

M. H. Wilkening, "Variation of natural radioactivity in the atmosphere with altitude," Transactions of the American Geophysical Union, 37, 1956, pp 177-190.

J. R. Wright and O. F. Smith, "The variation with meteorological conditions of the amount of radium emanation in the atmosphere, in the soil gas and in the air exhaled from the surface of the ground at Manila," The Physical Review, (2), 5, 1915, pp 459-482.

SUPPLEMENT TO THE THESIS

Operational procedure for the emanometric apparatus.

To illustrate the operational procedure of a radon analysis, reference is made to figure 3 of the thesis. The analysis proceeds according to the following sequence of operations:

1. Switch the spectrometer to 180° of position. This allows time for the tubes to warm up and the circuit to stabilise.

2. Fill trap 1 with a mixture of dry ice and acetone.

3. Immerse the scheelite cell in liquid oxygen and allow time for the scheelite part of the cell to cool; note the temperature of the liquid oxygen.

4. Check the valve settings:
   a. Valves 1 and 4 open;
   b. Valves 2, 3, and 5 closed.

5. Now pump by and extract the air flow through the collecting system, as indicated on the flowmeter, with valve 1.

6. Collect the sample for the desired length of time at the selected flow rate.

7. While the sample is being collected, fill the ion chamber with argon and determine the background current as follows:
   a. Check that valve 5 is closed.
   b. Close valve 4, open valve 3, and turn the three-way valve 2 to the line, the ion chamber and the pump P;
   c. Close the valve 2 so connecting ion chamber and the line.
   d. Switch head 1 to 180° degrees.
   e. Introduce argon to atmospheric line to the ion chamber, disconnecting slowly from the system.
   f. Gradually open valve 2, connecting argon into the ion chamber.
   g. When the zero indicates that the chamber is at

Operational procedure for the emanometric apparatus.

To illustrate the operational procedure of a radon analysis, reference is made to Figure 2 of the thesis. The analysis proceeds according to the following sequence of operations:

1. Switch the electrometer to the "on" position. This allows time for the tubes to warm up and the circuit to stabilize.

2. Fill trap 1 with a mixture of dry ice and acetone.

3. Immerse the condenser coil in liquid oxygen and allow time for the inner part of the tubing to reach the temperature of the liquid oxygen.

4. Check the valve settings:
   a. Valves 1 and 4 open.
   b. Valves 2, 5, and 6 closed.

5. Turn pump P_2 on and adjust the air flow through the collection system, as indicated by the flowmeter, with valve 5.

6. Collect the sample for the desired length of time at the selected flow rate.

7. While the sample is being collected, fill the ion chamber with argon and determine the background current by the following method:
   a. Check that valve 6 is closed.
   b. Close valve 3, open valve 7, and turn the three-way valve 8 to the line, the ion chamber and pump P_1.
   c. Start pump P_1 and evacuate the ion chamber and the line.
   d. Immerse trap 2 in liquid oxygen.
   e. Rotate valve 8 to connect the line to the ion chamber, disconnecting pump P_1 from the system.
   f. Slowly open valve 3, admitting argon into the ion chamber.
   g. When the gage indicates that the chamber is at

atmospheric pressure, rotate valve 8 so that the
ion chamber is isolated from the line and the pump P_1.
   h. Remove the liquid oxygen from trap 2.
   i. Close valves 3 and 7; shut off pump P_1.
   j. Record the background current.

8. After collection is made for the specified time, shut off
pump P_2 and close valves 1 and 5.

9. Following the procedure of 7a, 7b, 7c, and 7d above, evacuate
the transfer system.

10. Remove the liquid oxygen from the condensing coil and allow the
coil to warm up.

11. After reaching a vacuum on the transfer side of the system,
recheck the valve settings:
   a. Valve 3 closed.
   b. Valve 5 closed.
   c. Valve 1 closed.

12. Close valve 7.

13. Rotate valve 8 connecting the line to the ion chamber.

14. Shut off pump P_1.

15. Open valve 2, allowing about 4-5 pounds of argon pressure
on the condensing coil.

16. Open valve 6.

17. Open valve 7. This permits the argon to flow through the
condensing coil, pushing the radon through trap 2 and into
the ion chamber.

18. When the gage indicates atmospheric pressure, rotate valve 8,
isolating the ion chamber from the entire system. Close valve 2.

19. While the ion current is being recorded, close valve 6, open
valve 1, surround the condensing coil with liquid oxygen, and
the system is ready to collect another sample.

This thesis is accepted on behalf of the graduate faculty of the Institute by the following committee:

Hans mu Butler

Richard C. Sth.

Clay J Smith

Maurin W Wilkening

Date: _March 12, 1958_