T H E

AMERICAN JOURNAL OF SCIENCE

[F O U R T H S E R I E S .]

ART. XIX.—*The Probable Influence of the Soil on Local Atmospheric Radioactivity*; by JAMES COX SANDERSON, B.A.*

Introduction.

THROUGH the investigations of Elster and Geitel† attention was first called to the fact that the ordinary atmosphere normally contains radioactive constituents which can be collected by suspending a negatively charged wire for some time in the open air. Their experiments also showed that the air of cellars and caves contains a relatively greater proportion of radioactive material than the free air above ground, which suggested the possibility that the radioactive substances in the air might be of subterranean origin. The work of other investigators along the same lines has contributed largely to the extension of our knowledge of the nature and origin of atmospheric radioactivity.

Bumstead‡ collected the active deposit upon a long wire, stretched in the open air and charged to a high negative potential. He was the first to show conclusively that the atmospheric radioactivity was due in part to the presence of radium emanation and its disintegration products, and in part to the emanation and the disintegration products of thorium.

Gockel§ has examined the problem from the meteorological point of view and has found very considerable variations in the atmospheric radioactivity to exist with rising and falling

* Thesis presented to the Faculty of the Graduate School of Yale University, June, 1911, for the degree of Doctor of Philosophy.

† Phys. Zeitschr. ii. 590, 1901.

‡ This Journal, xviii, p. 2, 1904.

§ Phys. Zeitschr., iv, 604, 1902-3; v, 591, 1904; ix, 304, 1908; ix, 907, 1908.

AM. JOUR. SCI.—FOURTH SERIES, VOL. XXXII, No. 189.—SEPTEMBER, 1911.

barometer, varying direction and velocity of winds, etc. The radium emanation is evidently the one which is affected by such changes in atmospheric conditions, since it has a comparatively long half-value period, can be carried great distances and diffuses to great heights before completely disintegrating. The thorium emanation, on the other hand, with its comparatively short life, will only be found near the surface of the earth. Dadourian* and W. Wilson† used the method of the negatively charged wire to determine the ratio of the amounts of radium and thorium emanations in the atmosphere. Kurz,‡ using much the same method, obtained accurate curves, as well as some quantitative data, for the active deposits.

Since the emanations present in the atmosphere must have their origin in the soils and rocks of the earth's surface, a study of the radioactive properties of the underground air might be expected to furnish valuable data on the nature and origin of atmospheric radioactivity.

Ebert,§ Dadourian|| and Blanc¶ have investigated the radioactivity of the underground air. In each instance the active deposit was collected upon a negatively charged electrode over which underground air was drawn, or which was allowed to stand in a vessel filled with underground air. Blanc placed a large vessel, opened at the bottom, upon the ground and collected, upon a negatively charged electrode, the active deposit, produced by the emanation diffusing into it. He plotted the decay curve of this active deposit and, from the curve, deduced the amount of thorium active deposit initially present. He then allowed thorium emanation to diffuse into the same vessel from a tube containing a known amount of thorium hydroxide mixed with sand. On comparing the amounts thus obtained, he concludes that there must be at least 1.45×10^{-3} gram of pure thorium per gram of vegetal earth in Rome.

In one of the earlier experiments of Elster and Geitel,** the underground air was drawn continuously into the electroscope for an hour. The rise of the activity, due to the production of active deposit, was measured and, when this had reached a maximum, the current of air was stopped and the decay of the activity followed for many hours. They afterward devised and employed the method of collecting the active deposit on a negatively charged electrode which is subsequently placed in the ionization vessel. An examination of the literature of the subject has failed to disclose any other instance where the

* This Journal, xxv, 335, 1908.

† Phil. Mag., xvii, 321, 1909.

‡ Phys. Zeitschr., ix, 177, 1908.

§ Phys. Zeitschr., iv, 162, 1902-3.

|| This Journal, xix, 16, 1905.

¶ Phys. Zeitschr., ix, 294, 1908; Phil. Mag., xviii, 146, 1909.

** Phys. Zeitschr., iii, 574, 1901-2.

radioactive properties of the underground air have been studied by drawing a current of air directly through an electroscope.

The object of the present investigation was to determine, by an elaboration of the early experiment of Elster and Geitel just cited, the character and amount of the gaseous radioactive constituents of the soil.

The general scheme of the investigation was as follows :

A current of underground air was drawn from a small iron vessel, buried about 120 centimeters beneath the surface of the earth under the basement floor of the Physical Laboratory, and was conducted through the ionization chamber of a gold leaf electroscope. Immediately following the introduction of the air there was a rise in the activity to about eighteen times the natural leak of the electroscope. This was due to radioactive emanations in the underground air. The activity continued to rise, at first rapidly, and then more slowly, for about three hours, as the result of the production of the radium active deposit. The air current was continued and a very gradual rise in the activity was noted for about three days, when it reached a maximum. If, at this stage, the flow of air was suddenly stopped, a sharp falling off of the activity was noticed which continued for five or six minutes and then reached a minimum. This part of the activity fell to half value in about one minute and was easily identified as thorium emanation.

A relation was found between the amount of thorium emanation set free per cubic centimeter of the soil and the corresponding activity as measured in the electroscope. It was thus possible to compare the amount of thorium emanation set free per cubic centimeter of the soil with that liberated by a known amount of thorium.

The case of the radium emanation is more simple; since the half value period of radium emanation is comparatively long, it suffers no appreciable loss of activity in the time taken to reach the electroscope. It is therefore only necessary to standardize the electroscope by introducing the radium emanation in equilibrium with a known amount of radium and determining the rate of leak after the emanation has stood for three hours.

Some additional experiments were made on a number of soils, sands and minerals, with a view to comparing their emanating power.

Apparatus.

The electroscope consisted of two cylindrical metal vessels, 30 centimeters high and 13 centimeters in diameter, placed one above the other. The lower vessel, or ionization chamber, was provided with a central electrode which extended, through

sealing wax insulation and guard ring, into the upper vessel, and served to support the gold leaf. The guard ring was kept charged to —300 volts. The walls of both vessels were grounded, while the gold leaf and central electrode were charged negatively. The source of potential was a battery of small storage cells. The position of the gold leaf was read by means of a telemicroscope through mica windows in the case of the upper vessel. A small tube near the bottom of the ionization vessel admitted the current of air, which was drawn out through a similar tube near the top.

To get the underground air, several bricks were removed from the cellar floor and a deep hole dug in the sandy soil beneath. An iron cylinder, 13 centimeters in diameter and 30 centimeters high, with many holes drilled in its top, bottom and walls, was fitted with a brass tube, 130 centimeters long and 0.4 centimeter in diameter, and, after being wrapped in a covering of cheese cloth, was buried deep in the hole, so that, when the earth had been filled in, only two or three centimeters of the tube protruded vertically above the surface. Connection was made with the inlet of the ionization vessel by a rubber tube and a glass bulb containing glass wool, to act as a dust and ion filter. The outlet of the ionization vessel was connected with a filter pump through rubber and lead tubing. Suitable pinchcocks were placed at the outlet and inlet of the ionization vessel.

Radioactivity of the Underground Air.

After the normal air leak of the electroscope had been determined, the current of underground air was started and readings were taken every ten or fifteen minutes, over thirty divisions of the scale, for an hour and forty-five minutes. As will be seen later, these readings, when plotted, fitted almost perfectly the curve showing the increase of the amount of ionization due to α -particles from radium A and C. The current of air was maintained for about forty-four hours, when the active deposit of thorium was fifteen-sixteenths of its equilibrium value and practically the maximum activity was attained. At this point, the current of air was stopped exactly on the minute, and readings taken every minute for the next four minutes. This was repeated a number of times. As will be shown later, these readings approximated closely to the decay curve of thorium emanation. Finally the underground air connection was removed and the emanations swept out by drawing the air from the room through the electroscope. Readings were taken, at first every few minutes and then at longer intervals, which, when plotted, showed the dis-

FIG 1.

- I. Current of underground air flowing.
- II. Current of underground air stopped.
- III. Underground air swept out.

integration of radium A and C, and finally, of thorium B and C. Figure 1 shows graphically the growth and decay of the activity due to the presence of radium and thorium emanations in the underground air. The ordinates are activity in scale divisions per minute and the abscissas represent time; for convenience, the scale of the abscissas is different in different parts of the curve, as indicated.

From the accepted values of λ for radium A and C, a curve was plotted showing the increase with time of the ionization due to the α -rays from radium A and C. The number of ions produced by an α -particle of each kind was taken as proportional to its range. The ordinates were percentages of the total activity. Assuming that the curve passed through the point obtained empirically at 105 minutes, it was successively made to include five or six of the preceding points and in each case the value of the total activity due to radium A and C deduced. The mean of these values was taken, and from this mean the value of the ordinate when $t = 0$ was obtained. This represented the combined activities of radium and thorium emanation.

The part of the curve showing the disintegration of thorium emanation is plotted from the accepted value of λ for that substance. The observed values are in good agreement with those calculated; readings had to be taken at time intervals so short that certain irregularities in the motion of the gold leaf became very apparent and were an obvious source of error.

In plotting the rise of the activity of radium A and C, the active deposit of thorium was neglected, because the first disintegration product of thorium emanation is rayless. In the last part of the curve, however, thorium A, B and C are in equilibrium with the emanation, and in the curve for the decay of the activity due to radium A and C, their contribution to the total ionization had to be taken into consideration. The initial values of the activity, in scale divisions per minute, for the active deposits of thorium and radium, were deduced as follows:

At 35 minutes and 95 minutes after the emanations had been swept out, the total activity was 1.65 and 0.90 scale divisions per minute respectively. At 35 minutes the radium A and C was 46.7 per cent of its initial value; the thorium B and C, 96.4 per cent. At 95 minutes the values were 14.5 per cent and 90.5 per cent, respectively. If x be the initial value of the radium A and C, and y the initial value of the thorium B and C, we have: $.467x + .964y = 1.65$ and $.145x + .905y = 0.90$ which give, $x = 0.64$ and $y = 2.22$ scale divisions per minute. During the disintegration of radium A and C, the curve for thorium B and C is practically a straight line and

is so drawn in the figure. The decay curve for radium A and C is superposed on this straight line to show the combined activities.

The values in the last part of the curve correspond to a half-value period for thorium active deposit of about 12 hours, which is in sufficiently good agreement with the accepted value, namely 10.6 hours.

Points surrounded by circles are observed points; those with double circles are mean values of two or more readings; a point in a square is one which was extrapolated from the curve.

Standardization of the Thorium Emanation.

In order to compare the thorium emanation in the underground air with that produced by a known amount of thorium, it was necessary to obtain a relation between the amount of emanation produced per cubic centimeter of the earth or sand and the corresponding rate of leak of the electroscope. The calculation was simplified by substituting for the mercury flask and connecting tube a brass tube, 140 centimeters long and with an internal diameter of about 0.4 centimeter. This tube was provided with a sharp, conical head of iron, 3 centimeters long, with a shank which fitted snugly in the tube and a shoulder which rested firmly against its end. The tube was driven into the earth with a heavy mallet until only a centimeter or so remained above ground. It was then withdrawn a short distance and the conical head driven out with a blow from a long iron rod passed into the tube. The ionization chamber of the electroscope was then connected to the brass tube by a length of rubber tubing and observations were made as follows:

The filter pump was started (running about 3 liters per minute) and the current of air continued for several minutes until the ionization vessel was filled with underground air. The pump was then shut off, and the vessel with the underground air allowed to stand for half an hour, when the supply of air was renewed by running the pump for another short interval. This operation was repeated every half hour. At the end of about three hours the radium A, B and C had come into equilibrium with the emanation and the radium part of the observed activity had attained a constant value.

With the pump running, a series of readings was then made which gave the combined activities of radium emanation, radium A, B, C, and thorium emanation. The pump was stopped and five or six minutes allowed for the thorium emanation to decay. A series of readings then gave the constant radium part of the activity alone. The difference

between the activity with the pump running and the activity when the air was at rest was due to the thorium emanation.

It is easily seen that all the thorium emanation is drawn from

a sphere whose radius $R = \sqrt[3]{\frac{3qT}{4\pi P}} = 23.5$ centimeters, where

q = cubic centimeters per second drawn by the pump (50); T , the number of seconds required for thorium emanation to decay practically completely (420); and P , the volume of air per cubic centimeter of the earth or sand (.366). This is the radius of the sphere from which all the air is drawn in seven minutes and any emanation originating beyond this sphere will entirely decay before reaching the electroscope. Since the tube was driven into the soil a distance equal to about six times the radius of this sphere, no variation in the concentration of thorium emanation was to be expected. The radium emanation, on the other hand, with its longer half-value period, was drawn from a sphere of relatively enormous size, but, in standardizing the radium, the underground air current was never running for lengths of time aggregating to more than twenty minutes and in this time all the air was drawn from a sphere of only about 32 centimeters radius.

Let n be the amount of thorium emanation in equilibrium per cubic centimeter of the earth. Then $\frac{n}{P}$ is the amount of

emanation per cubic centimeter of underground air, and this will be its concentration whether at rest or in motion, until it leaves the earth to enter the tube. If k_1 is the total volume of the brass tube and all connections, up to the ionization chamber,

$\frac{k_1}{q}$ is the time taken to reach the ionization chamber and the

concentration will there have the value $\frac{n}{P} e^{-\frac{\lambda k_1}{q}}$, where λ is

the decay constant for thorium emanation. The time taken to

pass through the ionization chamber is $\frac{k_2}{q}$, where k_2 is the

volume between the inlet and outlet. The activity measured is proportional to the mean concentration of the emanation in its passage or

$$N \propto \frac{1}{T} \int_0^T e^{-\lambda t} dt = \frac{1}{\lambda T} (1 - e^{-\lambda T}) = \frac{q}{\lambda k_2} (1 - e^{-\frac{\lambda k_2}{q}})$$

Hence the concentration

$$(I.) \quad N = \frac{nq}{P\lambda k_2} e^{-\frac{\lambda k_1}{q}} (1 - e^{-\frac{\lambda k_2}{q}})$$

where $k_1 = 101.5$ and $k_2 = 2750$.

To test the accuracy of the measurements involved in this equation, a series of readings was made with the pump running at different speeds; i. e. for different values of q . By taking the mean of several measurements, the activity when $q = 34.5$ (pump fully on) was found to be 2.06 scale divisions per minute.

FIG. 2.

The corresponding value given by the equation was $1.64 n$. This ratio was used to determine the ordinates of equation I, which was plotted for values of q from zero to 5 liters per minute. The other observed values of the activity corresponding to different values of q were then plotted directly and it will be observed that they fit the curve very closely. (Curve I, figure 2.) Since $1.64 n$ gave a rate of 2.06 scale divisions per minute, one scale division per minute must indicate a mean concentration in the electroscope of $0.80 n$ per cubic centimeter.

The Thorium Standard.

In selecting a standard for comparison with the amount of thorium emanation obtained with the underground air, a number of difficulties are apparent. No one as yet has devised a

method by which either all or a definite fraction of the thorium emanation in equilibrium with a known amount of a thorium salt may be obtained with certainty. It may reasonably be assumed, however, that if the thorium salt could be obtained in the form of a sufficiently thin coating on the surface of some neutral material, all of the emanation produced within it would escape. For this reason the thorium solution was thoroughly mixed with a considerable quantity of neutral sea sand so that each grain was covered with a very thin film of radioactive material.

Another point which must be taken into consideration is the requirement that all the disintegration products of thorium, preceding the emanation, shall be present in equilibrium amounts. In the present experiment this was accomplished by taking a solution of a thorium mineral containing a known proportion of thorium.

The mineral chosen was thorite, containing 52 per cent of ThO_2 in the form of a silicate. When this mineral was dissolved in hydrochloric acid, diluted and mixed with a granular neutral material, a thin gelatinous film of free silicic acid, containing the ThCl_4 , was formed on each grain, and it was thought that such a film would possess a very great emanating power, an assumption which was amply justified by comparison with thorium in other forms. By adopting as a standard the most emanating form of thorium which could be found, it was assured that the results obtained for the thorium content of various materials were very nearly minima. The standard was prepared by dissolving 0.1 gram of thorite in a little hydrochloric acid. It was then diluted and intimately mixed with 884 grams of white beach sand, which had been previously tested and found free from any measurable radioactivity. This mixture was thoroughly dried and then moistened with 12 cubic centimeters of water. The standard thus prepared was placed in a vertical tin cylinder, about 35 centimeters high, closed at its lower end, except for a brass connecting tube, and provided with a cover having a small inlet hole in its center. About a centimeter from the bottom was a platform of wire gauze on which was placed a disk of cotton. The air was sucked in at the top, through the sand, out at the bottom and thence, through a rubber connecting tube, to the ionization chamber.

The theoretical considerations governing this case are much the same as before. When the underground air was drawn directly into the brass tube, it retained its equilibrium concentration up to the instant of entering the tube. In the case of the cylinder, a horizontal layer of air enters the sand practically devoid of thorium emanation and passes through it in a time

$\frac{k_s}{q}$, where k_s is the volume of air in the interstices of the

sand. Each layer will therefore acquire a concentration $\frac{n}{P} (1 - e^{-\frac{\lambda k_3}{q}})$ before leaving the sand, where n is the emanation per cubic centimeter of sand and P the volume of air in one cubic centimeter of sand. The terms relating to the decay in transit and the mean value in the ionization chamber will keep the same form. Hence

$$(II.) \quad N = \frac{nq}{\lambda P k_2} (1 - e^{-\frac{\lambda k_2}{q}}) e^{-\frac{\lambda k_1}{q}} (1 - e^{-\frac{\lambda k_3}{q}})$$

where $k_1 = 347$, $k_2 = 2750$, $k_3 = 201$, $P = .366$.

With the pump drawing 3.03 liters per minute the rate of the electroscope was 6.865 divisions per minute; for the same value of q , equation II, gives 0.0886 n . This ratio was used to determine the ordinates, and equation II was plotted (v. curve II, figure 2). Two other observed values of the activity, for different values of q , fell near enough this curve to show the correctness of the assumptions.

Since 6.865 scale divisions per minute correspond to a concentration of 0.0886 n , we see at once that one scale division corresponds to a concentration of 0.0129 n . But n is the emanation in equilibrium with

$\frac{0.52 \times 0.1}{545}$ gram of $\text{ThO}_2 = 9.45 \times 10^{-5}$

gram ThO_2 , and hence one scale division per minute corresponds to a concentration equal to the emanation in equilibrium with 1.22×10^{-6} gram of ThO_2 or 1.07×10^{-7} gram of pure thorium under the conditions specified.

Other Materials.

A larger cylinder, made from a section of galvanized iron pipe, with a diameter of 17.6 centimeters, a height of 71 centimeters, and a capacity of about 15 liters, was provided with a gauze screen and cotton pad, as before, and used to test the thorium content of other materials.

The earth under the cellar floor consists of about 85 per cent disintegrated red sandstone, none of which disintegrated in situ, but which was carried by streams from a melting glacier. The grains were worn and rounded in transit. The remaining 15 per cent consists of fragments of trap rock and various crystalline gneisses and schists. The sample designated "Sandstone II" was taken from some excavations about a mile from the physical laboratory. This earth has the same constituents as the above, but disintegrated where it was found and was not carried by glacial streams.

A sample, called "surface soil," was of the same general nature as the earth under the laboratory floor, but contained a considerable proportion of loam. It was a specimen of the soil in the rear of the Laboratory, and was removed from just beneath the surface.

The results of the measurements of the different specimens were of the same general character, although, as might be expected, the earth taken from near the surface was less active than the material lying deeper. This is probably due to the leaching action of the surface water.

A test on white beach-sand showed no activity. This was significant and made it seem probable that the activity of soil is due for the most part to the material covering the separate grains and not to mineral constituents of the grains themselves. To test this point some cellar earth was heated with fairly strong hydrochloric acid, filtered and washed. The sand thus obtained was thoroughly dried and placed in the larger cylinder, which it half filled. As will be seen from the table, fully half of the activity was removed by this process. This specimen is called "treated earth" in the table.

A solution was prepared by dissolving 0.8135 gram of thorinite, containing 78 per cent ThO_2 , in a little concentrated nitric acid with gentle heating, evaporating to dryness, dissolving and diluting to 50 cubic centimeters with distilled water. 10 cubic centimeters of this solution was diluted still further and mixed with 884 grams of neutral beach sand in the same manner as has been described in the case of thorite. As is shown by the results given in the table, the emanating power of the thorium nitrate thus prepared is noticeably less than that of the thorium chloride contained in a gelatinous film of silicic acid.

Another experiment was made in which a volume of 10 cubic centimeters of the thorianite solution was freely diluted and thoroughly mixed with 15 liters of the cellar earth. The normal activity of the earth was subtracted from the reading obtained, and this gave the activity due to the thorianite alone. The table shows that in this condition the thorium is slightly more emanating than when mixed with sand, but the difference is not so striking as in the case of thorite. In the table this specimen is designated "enriched earth."

Measurements were also made of the amount of thorium emanation escaping from the thorium minerals monazite (5 per cent ThO_2) and thorianite (78 per cent ThO_2). The monazite was in the form of fine sand, and the thorianite in the form of cubical crystals about 3 millimeters on a side. About 500 grams of each were placed in a glass cylinder and held in place by glass wool. Air was drawn through and readings taken as before.

In all the foregoing experiments the relation between the amount of thorium emanation per cubic centimeter and the activity as measured in the electroscope was deduced from equation II by substituting the proper values for the constants k_1 , k_2 , and P .

The experiment designated "surface diffusion" was made by placing a rectangular metal plate of about 2400 square centimeters, one centimeter above a leveled surface of cellar earth and drawing air, into the electroscope, through a tube soldered in the middle of the plate. In this way the air was made to pass slowly over the surface of the soil, which was quite moist. An experiment was also made with rather large fragments of broken red sandstone. The fifteen-liter cylinder was filled with this material and air drawn through it as before. It is obvious that equation II does not apply in either of these cases.

The third column gives the amounts of the thorium in one cubic centimeter of sand, under the specified conditions, which will liberate the same amount of emanation as one cubic centimeter of the material considered. The fourth column gives the amount of thorium present in one cubic centimeter of the material, in those cases where this was known.

Material tested	Activity in scale divisions per minute	Thorium equivalent in grams per c. c.	Am't of Thorium actually present per c. c.
Cellar Earth	2.06	1.35×10^{-6}	
Surface Soil	1.13	1.08×10^{-6}	
Sandstone II	1.41	1.35×10^{-6}	
Sea Sand	0.00	0.00×10^{-6}	
Surface Diffusion	0.20		
Broken Sandstone	0.55		
Thorianite Sand	12.32	1.50×10^{-4}	2.04×10^{-4}
Monazite	6.86	2.04×10^{-4}	1240×10^{-4}
Thorianite	95.2	3.58×10^{-4}	30400×10^{-4}
8 Liters of treated earth	0.50		
8 Liters of cellar earth	1.01		
Enriched earth	6.48	5.15×10^{-6}	7.00×10^{-6}

It will be seen from the table that the thorium in the sand treated with thorianite solution is about four hundred and twenty times as emanating as the thorium in monazite, and about six thousand, one hundred times as emanating as the thorium in thorianite.

The Radium Standard.

The radium emanation obtained on dissolving 0.0907 gram of North Carolina uraninite (containing 76 per cent of uranium) was introduced in the electroscope. After standing for about

three hours, the rate of leak of the electroscope was 110 divisions per minute. Taking 3.4×10^{-7} gram as the amount of radium in equilibrium with one gram of uranium, the amount of radium in the sample was 2.34×10^{-8} gram. Approximately 14 per cent of the radium emanation escapes from the specimen of (powdered) uraninite taken as a standard,* and therefore the emanation introduced into the electroscope was in equilibrium with only 86 per cent of 2.34×10^{-8} gram, or 2.0×10^{-8} gram of radium. One scale division per minute therefore corresponded to the ionization produced by the emanation, radium A, B, and C, in equilibrium with 1.82×10^{-10} gram of radium.

The mean of several measurements of the activity of the radium emanation, in equilibrium with radium A, B, and C, contained in the underground air was five scale divisions per minute. The total amount of radium emanation present in the volume of underground air necessary to fill the electroscope was therefore equal to that in equilibrium with $5 \times 1.82 \times 10^{-10}$ or 9.1×10^{-10} gram of radium. The volume of the ionization chamber of the electroscope was 3,800 cubic centimeters and therefore each cubic centimeter of underground air contained radium emanation equivalent to that in equilibrium with 2.4×10^{-13} gram of radium. The porosity of the earth was 36.6 per cent, that is to say each cubic centimeter of earth contained 0.366 cubic centimeter of air. Therefore the amount of radium emanation produced by one cubic centimeter of earth was equal to 2.4×10^{-13} times 0.366 or 8.9×10^{-14} gram of pure radium.

Discussion of Results.

The results of experiments conducted by a number of independent investigators have clearly demonstrated that the radioactive properties of the normal atmosphere near the surface of the earth are due to the presence of very small amounts of the emanations of radium and thorium and the disintegration products of these substances. It is necessary to assume that these emanations are formed by thorium and radium compounds present in the solid material constituting the surface of the globe. The radium emanation because of its relatively slow rate of disintegration might be expected to reach the surface by diffusion from considerable depths and to escape from the interior of masses of rock having a slight porosity. In contrast to this, however, the thorium emanation disintegrates so rapidly that the process of diffusion alone is insufficient to bring any appreciable quantities of this to the surface, except from the uppermost layers of a light and porous earthy covering. It is in the

* Boltwood, this Journal, xxv, 281, 1908.

free surface of such a covering that by far the greater proportion of the thorium emanation which ultimately enters the atmosphere must originate, and its dissemination through the lower strata of the air must be greatly facilitated by winds and other air currents.

It is, moreover, evident that the actual quantities of either emanation which can obtain access to the free atmosphere under the most favorable conditions of exposure, etc., will be much less dependent on the actual amounts of thorium and radium present in the surface materials than on the physical and chemical conditions in which these elements exist and which determine their emanating properties. If the thorium and radium are in the form of compounds which permit the escape of a large proportion of the emanation formed within them, their contribution to the local atmospheric radioactivity will be much more pronounced than that of much larger quantities of the parent radioactive elements existing in a non-emanating form.

Sand and earth consist essentially of disintegrated rock which has undergone more or less chemical decomposition. For facilitating the escape of the emanations the advantage derived from the mechanical subdivision alone is apparent. The decrease in the thorium emanating power of a specimen of earth treated with dilute hydrochloric acid, which was observed in the course of the experiments described in this paper, also justifies the assumption that a considerable proportion of the thorium emanation which escapes originates in the superficial coating covering the separate grains of the specimen of sand examined. It is, therefore, highly probable that the thorium in this surface coating exists in a chemical form which readily permits the escape of emanation.

Since by far the greater proportion of the land areas of the globe are covered with soil of some description, it is apparent that the part played by this in contributing to the radioactive constituents of the atmosphere must be much greater and much more important than the part played by the rocks which underlie it. It is, therefore, to be expected that an examination of the radioactive properties of the soil and the underground air in different localities will afford valuable data for explaining the differences in the radioactivity of the atmosphere already observed in certain instances.

As the average of results of measurements conducted during a period of a year at Montreal, Eve* found the amount of radium emanation present per cubic centimeter of atmospheric air to be equivalent to that in equilibrium with 6×10^{-17} gram

* Phil. Mag., xvi, 622, 1908.

of radium. This is one four-thousandth the amount found for underground air here at New Haven. The average of Joly's values* for the thorium content of many common rocks is approximately ten times the value found from the emanating power for the thorium content of the earth at New Haven, while his value for the radium content is about one hundred times that found for New Haven earth by the method employed in this paper.

Summary.

A determination has been made of the amount of radium emanation in the underground air in the neighborhood of the Sloane Physical Laboratory. It was found that one cubic centimeter of underground air contains radium emanation equal to the amount in equilibrium with 2.4×10^{-13} gram of radium. This is equivalent to the production, per cubic centimeter of earth, of radium emanation in equilibrium with 8.9×10^{-14} gram of radium.

A method has been devised by which the amount of thorium emanation present in underground air can be directly measured in an electroscopie and compared with the emanation evolved by a known weight of thorium salt. In this manner it was found that the earth in the neighborhood of the Laboratory emits per cubic centimeter thorium emanation equivalent to that produced by 1.35×10^{-6} gram of thorium under the standard conditions.

The important bearing of the emanating power of the soil on the radioactive properties of the atmospheric air in different localities has been pointed out, and a method has been suggested by which the emanating power of the soil and the radioactive character of the underground air can be determined in any given locality.

In conclusion, I wish to acknowledge my very great indebtedness to Professor B. B. Boltwood, and express my thanks for his assistance and advice.

Sloane Physical Laboratory, May, 1911.

* *Phil. Mag.*, xviii, 140, 1909.