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To cite this article: A.S. Eve M.A. (1907) LXXII. On the amount of radium emanation in the atmosphere near the earth's surface , Philosophical Magazine Series 6, 14:84, 724-733, DOI: [10.1080/14786440709463735](https://doi.org/10.1080/14786440709463735)

To link to this article: <http://dx.doi.org/10.1080/14786440709463735>



Published online: 16 Apr 2009.



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LXXII. *On the Amount of Radium Emanation in the Atmosphere near the Earth's Surface.* By A. S. EVE, M.A.*

IT has been proved by Elster and Geitel that a negatively-charged wire, exposed for a few hours in the air, receives a radioactive deposit similar in character to the quick-changing products of radium. Radium is known to be widely distributed among the constituents of the earth's crust. Moreover, there is evidence that the emanation from the radium in the earth escapes into the atmosphere. From the emanation arise the active deposits of radium in the atmosphere. The radioactive changes in the air and soil account for the ionization of the air ; and important electric and meteorological effects result, the characters of which are at present imperfectly understood.

It is, then, important to form an estimate of the amount of emanation in the atmosphere, expressed in terms of the quantity of radium required to keep the supply constant. Throughout this paper the cubic metre, and one-billionth (10^{-12}) gram of pure radium will be adopted as units of measurement. By "pure" radium is meant that which generates 110 gram-calories per hour per gram of radium.

The first attempt to measure the amount of radium emanation in the atmosphere was made by the present writer †, by an indirect method, in the following manner :—On the ground in the Campus of McGill University was placed a large zinc cylinder, of known volume, with closed ends. Along the axis was a wire, charged negatively to 10,000 volts, on which was collected the active deposit from the air in the cylinder. The activity of this deposit was measured by a gold-leaf electroscope, which was calibrated by means of the active deposit collected on a wire from the emanation obtained from a standard solution of radium of known strength. By this comparative method it was found that a cubic kilometre of air contained the active deposit which could be obtained from the emanation arising from $\cdot 14$ gram of pure radium bromide. In other words, one cubic metre of air near the earth's surface, at the place of measurement, appeared to contain the emanation from 82×10^{-12} grams of pure radium. But there was no definite evidence of the existence of the emanation ; its presence was inferred, not proved. There was also an objection to the method. The active deposit collected on a charged wire fluctuates largely in magnitude ;

* Communicated by the Author. A preliminary note on this paper appeared as a Bulletin of the Royal Society of Canada, 21st June, 1907 (Ottawa).

† Phil. Mag. July 1905.

the maximum is 16 times the minimum, and the quantity depends in a subtle manner on a variety of meteorological conditions. This difficulty was to a great extent overcome by taking the average of a number of determinations.

In the autumn of 1906 Professor Rutherford found that charcoal, prepared from coconuts, entirely absorbed the radioactive emanations, provided they were passed very slowly through the charcoal. He was good enough to assign to me the work of using his method in the determination of the amount of emanation in the atmosphere.

Air was therefore drawn through glass tubes, containing charcoal, for various times and at various speeds. The tubes were heated by Bunsen flames, and the gases were collected over water and introduced into an electroscope, which had previously been exhausted by a water-pump.

Difficulties at once arose. In order to absorb the emanation completely, it is necessary that the velocity of the air-current through the charcoal should be small, certainly not exceeding 1 c.c. per second. At this rate, it would take a little more than 11 days to pass one cubic metre through the charcoal; and it will be remembered that radium emanation decays to half-value in a little less than four days. It was soon found that the proportion of emanation absorbed was a function of the speed. The greater the speed, the less percentage of a given quantity of emanation was absorbed. It was found best to run the air through the charcoal at a fair speed, thus absorbing only a fraction of the emanation passing through the charcoal. Air was then bubbled through standard solutions of radium, carrying the resulting emanation through the tubes containing charcoal. These were heated, and the gases driven off were collected, and introduced into the electroscope as before. It will be noted that the method is a comparative one, and that the conditions are identical, except that in one case there is the unknown amount of emanation in the atmosphere, and in the other case the known amount of emanation extracted from the radium solution by bubbling air through it.

After some trials with various electroscopes, a very satisfactory one was made as described in a previous paper*. A filter flask was silvered inside; a good earth-connexion was made from the silver coating; a rubber stopper sealed the top, and held the wire, sulphur-bead insulation, and light Dutch-metal leaf. Some of the silvering was scraped away, and the leaf was read by a microscope with a graduated eye-

* *Phil. Mag.* August 1907.

piece. Rubber tubing connected the side opening of the filter flask to a bulb containing phosphoric pentoxide ; and between this and a three-way tap was a capillary tube, useful in preventing any violent inrush of air, which would destroy the gold-leaf. The three-way tap led to the water-pump, and to the vessel containing the gas to be introduced into the electroscope.

The first series of observations made with a single tube of charcoal gave uncertain results, and some of the charcoal contained a small quantity of radium. Fresh coconuts were procured, and charcoal was prepared free from radium. The whole of the work was done in the Chemistry Building, which has been kept free from contamination by strong radioactive preparations. The air tested was obtained from outside, drawn through clean glass tubes passing through a hole in the window-frame. A manometer was used to test the velocity of the air-currents employed. This was calibrated by the help of an air-tight iron boiler, one-tenth of a cubic metre in volume, which could be exhausted by the water-pump.

Three glass tubes, each 1·2 cm. in diameter, containing 7 grams of charcoal, were placed in parallel, so that the air-current was divided. A comparison was made between the emanation obtained from the tubes (1) after the air-current had *run* for the number of days specified, (2) after the tubes had *rested*. The experiments were made in February and March, under winter conditions, in Canada. The results are given in scale-divisions of the observing microscope per minute. The natural leak of the electroscope was, and still is, ·065 division a minute.

No. of days.	Run.	Rest.
8	·147	...
7	·10
4	·136	...
3	·095
3	·118	...
3	·085
3	·10	...
3	·084
4	·11	...
3	·083

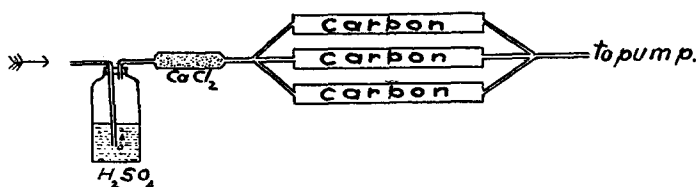
The average speed of the air-current was about 3·7 cm./sec. A solution containing 10^{-10} gram of radium was then placed in a flask, and the emanation from it carried for three days at the same speed as before through the carbon tubes. These,

when heated, gave emanation measuring .22 division a minute, of which .11 was due to the natural leak and the emanation in the air drawn through. Thus, 0.95 cb.m. of air contained the emanation from $.025 \times 10^{-10} / .11$ gram of radium, or 1 cubic metre from about 24×10^{-12} gram of radium. My result by the active deposit method was $3\frac{1}{2}$ times as great as this.

It will be seen that the results in the above table for a "run" were always greater than for a "rest," proving the existence of the emanation in the atmosphere; but the difference between them on which the result depends is about .025—a small quantity, about half the magnitude of the natural leak. It was, therefore, desirable to work on a larger scale.

Three iron pipes were prepared, 37 cms. long and 4 cms. in diameter. Each contained 220 grams of finely divided charcoal. Air from outside the building was drawn by a

Fig. 1.



water-pump through H_2SO_4 and $CaCl_2$, and was passed through the three iron tubes arranged in parallel. By this means the speed could be increased threefold, and yet the volume and cross-section of the absorbing charcoal was sufficiently large for good results. A convenient speed was found to be .09 cb.m. per hour, about 8 c.c. per second through each tube, or 8 cb.m. altogether in 3.7 days.

When the current of air had been flowing for a definite time, usually 3.7 days, the iron cylinders were removed and heated in succession to a dull red heat over a combustion furnace, and the expelled gases were collected in large vessels over water.

The 660 grams of carbon gave off 15,000 c.c. of gases, and these were drawn in 18,000 seconds through three small glass tubes in series, each containing 7 grams of charcoal. The small tubes were heated over a Bunsen burner, and the gases driven off were collected over water and introduced into the electroscope. Two or three hours later, the rate of leak of the gold leaf was determined. A complete observation occupied about nine hours.

An example of the method followed is here given :—

	Division per minute.
A. Natural leak of the electroscope	·065
B. Obtained from tubes after 3·7 days' rest	·25
C. Obtained from tubes after 3·7 days' run	·65
C—B. Due to emanation in 8 cb.m. of air	·40

A standard solution, containing 10^{-9} gram of radium, was then placed in a flask, and air was bubbled through the solution, through drying-tubes, and through the three iron pipes, at the same rate and for the same time as before.

	Division per minute.
D. Due to the standard solution and to all causes under heading C	2·1
D—C. Due to 10^{-9} gram radium	1·45

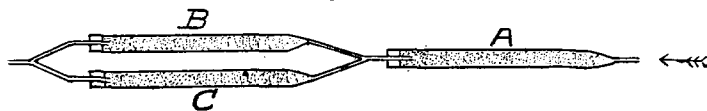
Hence 10^{-9} gram of radium gave 1·45 D/m, and 8 cb.m. of air gave ·40 D/m. Therefore, 1 cb.m. of air contained the emanation from $\frac{40}{145} \times \frac{10^{-9}}{8}$ or 35×10^{-12} gram of radium.

This was the largest value obtained. The smallest was about half this; and the mean value was 27×10^{-12} gram.

This method was laborious, a complete observation requiring nearly 4 days for pulling the air through the pipes, and 9 hours for obtaining the concentrated gases and introducing them into the electroscope. These experiments were made in the month of May.

A third method was now adopted. Glass tubes were taken, 25 cms. long and 2 cms. in diameter; each was filled with 50 grams of freshly prepared charcoal. They were arranged

Fig. 2.



in the manner shown in fig. 2. The air passed through A, and then divided into two currents passing through B and C respectively. It is obvious that the speed through B, or C, is half that through A; and it is known that the absorption is a function of the speed. Each tube was heated by two Bunsen burners held by hand, and the process of heating was continued until the gases practically ceased to come from the charcoal. The air in the tube was then blown out, and also

collected over water. The gases thus obtained from one tube were nearly sufficient to fill the electroscope. After several trials, a uniform speed of 6.7 c.c. per second, and a period of 2.7 days were adopted ; so that the total amount of air drawn through tube A was 1.56 cb.m. The amount of emanation collected in tube A was usually about 20 per cent. greater than the amount in B or C. The total amounts obtained in the three tubes added together are given in the following table :—

Date.	Divisions per minute.	
	Run.	Rest.
July 10	251
" 13	331	...
" 16	221
" 19	383	...
" 25	335	...
" 29	243
Aug. 1	390	...
" 3	233
" 6	373	...
" 9	360	...

Thus the mean of the "runs" was .362 and of the "rests" .237, with a difference of .125 D/m. The fluctuations in value of the middle column are in excess of experimental errors, and indicate that the amount of emanation in the air at Montreal in July was varying between maximum and minimum values in the ratio of 7 to 4, approximately. This variation is small compared with that observed in the active deposit collected from the atmosphere, for which the corresponding ratio is 16 to 1. This wider divergence is probably due, not to a change in the amount present, but to the meteorological conditions existing during the time of collection.

The charcoal in the tubes was next calibrated with standard solutions, three in number, of strengths 10^{-10} , 1.57×10^{-9} , and 1.57×10^{-7} . The method was the same as that described above.

Strong Solution. 1.57×10^{-7} gram.

The tubes gave

A.	B.	C.	Total.
57	22	25	104 divisions a minute.

In this case about 20 per cent. of the possible maximum was absorbed and given off by heating. It is noteworthy

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that the first tube A retained more than B and C, although the current through A was twice as fast as through B or C. All the emanation was not driven off by the first heating; at the second heating, two days later, the amounts obtained were

A.	B.	C.	Total.
7·3	3·7	4·4	15·4

The amount of emanation used in this case was large, and would at a maximum have caused the gold-leaf to move at a rate measurable by about 1000 divisions a minute. But in the 3·7 days only half this amount would be generated. It took six heatings and several days' rest to free the charcoal from this heavy dose of emanation.

Medium Solution. $1·57 \times 10^{-9}$.

The amounts obtained were

A.	B.	C.	Total.
·53	·40	·41	1·34

from which must be deducted ·36 due to the emanation in the air pulled through. Therefore $1·57 \times 10^{-9}$ gram caused 0·98 D/m under the conditions of the experiment. This is, again, about 20 per cent. of the possible maximum.

The amounts caught by the three tubes were nearly equal, and in a number of experiments with solutions of this strength I found that the leading tube A usually stopped but little more than B or C, about 25 to 30 per cent.

Weak Solution. 10^{-10} .

The effect was in this case small and difficult to measure, about ·06 division a minute.

These three calibrations are in fair agreement and give, per 10^{-10} gram,

·066 D/m for the strong solution,	
·062 " " medium "	
·06 " " weak "	

The mean value is ·063 D/m per 10^{-10} gram of radium.

But it will be remembered that 1·56 cb.m. of air gave ·125 D/m.

Hence 1 cb.m. of air contains the emanation from $\frac{·125}{1·56} \times \frac{10^{-10}}{·063}$ gram, or from 127×10^{-12} gram of radium.

Summary.—The amount of emanation per 1 cb.m. of air

near the earth's surface at Montreal has been determined four times :—

	Grams of radium.
In 1906, by the active deposit method	82×10^{-12}
In February and March 1907, with small tubes of charcoal (21 grams)	24×10^{-12}
In May 1907, with 660 grams of charcoal .	35×10^{-12}
In July and August 1907, with 150 grams of charcoal	127×10^{-12}

These figures, whilst of the same order, are not in satisfactory agreement. I have great confidence in the last large result : every point has been carefully verified, and several months of practice permit of a remarkable degree of accuracy in the measurement of these small quantities—a degree of accuracy which can only be realized by those who have done practical work of this kind. It is, of course, possible that the amount of emanation in the atmosphere may vary in different seasons of the year. During July the weather was moderately warm (65° – 70° F.), with S. and W. winds and sun. Every day or two there were thunderstorms with rain. The heavy rain may have forced emanation from the ground in larger quantities than in winter and spring. The active deposits on wires during July were not abnormal. The ionization was not large ; for I found $n_{+}=425$, $n_{-}=-830$ on a hazy day, $n_{+}=400$, $n_{-}=450$ on a remarkably clear day. It is my intention to continue to measure the emanation in the atmosphere for some months to come, in order to ascertain if there is a variation between summer and winter values.

A few notes are added which may save trouble to those who are making experiments similar to these. The emanation is given off from the charcoal in the latter part of the heating ; so that it is necessary to heat thoroughly. With emanation from 10^{-9} or 10^{-10} gram of radium about 97 per cent. of the total emanation absorbed by the charcoal may be driven off by careful heating with two Bunsen burners. With stronger solutions, of the order 10^{-7} gram, only 80 to 85 per cent. can be driven off in this manner by the first heating. Charcoal containing emanation can be partially de-emanated by passing a strong current of air through it. A general principle will be found to hold good in this case, as in most others, that whatever is easily absorbed is easily extracted, and conversely.

I venture to recommend the last-described method for the measurement of emanation in the atmosphere. It is well to

use three tubes and to add the results obtained, as it tends to minimise the effect of small errors.

Great care was taken to make absolutely certain that the emanation was not derived from radium impurities in the apparatus employed. A very slow current of air through the tubes containing charcoal gave nearly the same result as a "rest" experiment. Charcoal does not absorb well if choked with water-vapour. It seemed sufficient to bubble the air through two flasks of strong sulphuric acid. This acid was always renewed after a calibration experiment, in order to remove any radium which might be carried into the acid from the radioactive solution. I found that water and sulphuric acid absorbed but little of the emanation passing through them during these experiments. In any case, the method is comparative, and a slight absorption cannot, therefore, vitiate my results.

Since Strutt found that one gram of rock, on the average, contained 1.4×10^{-12} gram of radium, and we have seen that 1 cubic metre of the air contains the emanation from about 81×10^{-12} gram, we may conclude that 60 grams of rock would provide the radium emanation in 1 cb.m. of the atmosphere, if all the emanation escaped from the rocks. But Boltwood has shown that only 5 to 10 per cent. of the emanation escapes from a mineral; so that by far the greater part of the emanation in rocks, even near the surface of the earth, must undergo transformation without passing into the air.

Hence, if there enters the air 5 per cent. of the total emanation supplied by the rocks and soils extending to a depth of one or two metres, the supply would be sufficient to account for the emanation in the atmosphere extending to a height of 5 kilometres.

But radium emanation certainly reaches the atmosphere from considerable depths. Dr. Ruttan has kindly collected for me the natural gas and the mineral water from Caledonia Springs. The water, temperature 40° F., contains only 5.6×10^{-12} gram of radium per litre; but 1 cb.m. of the gas contains the emanation from $114,000 \times 10^{-12}$ gram of radium, or about a thousand times as much as a cubic metre of the atmosphere as measured at Montreal. This large output of radium emanation was detected owing to the fact that the gases bubbled through water. In other cases it might escape to no less extent without detection, no water being present.

It may be observed that in the present state of our knowledge the amount of radium in the earth near the

surface; the amount of emanation in the atmosphere; the resulting active deposit; the penetrating radiation due to all these; the ionization in the atmosphere: all are of the correct order of magnitude, so that they may be correlated.

Result.—The emanation in the atmosphere is absorbed by coconut-charcoal: its presence can be proved and its magnitude determined.

Four measurements have been made at Montreal, and the results are given in terms of the amount of radium required to maintain the supply per cubic metre constant. The smallest value obtained was 24×10^{-12} , the largest 127×10^{-12} . The probable average value is 80×10^{-12} . The amount of emanation is of the correct order to account for the active deposits of radium C, which may be collected on negatively-charged wires from the atmosphere.

Now that Professor Rutherford has left McGill University, I wish to state my indebtedness to him. If my papers have had any merit, it may be attributed, without exaggeration, to his influence or inspiration, and for these I am grateful.

McGill University, Montreal,
August 1907.

LXXIII. *The Production and Origin of Radium.* By E. RUTHERFORD, F.R.S., *Professor of Physics, University of Manchester**.

§ 1. **T**HE present point of view of regarding radium as a substance which is undergoing slow transformation was first put forward definitely by Rutherford and Soddy in the paper entitled "Radioactive Change" (Phil. Mag. May 1903, p. 590) in the following terms:—"In the case of radium, however, the same amount (viz. about 1 milligram) must be changing per gram per year. The 'life' of the radium cannot in consequence be more than a few thousand years on this minimum estimate, based on the assumption that each particle produces one ray at each change. ... So that it appears certain that the radium present in a mineral has not been in existence as long as the mineral itself, but is being continuously produced by radioactive change."

On this theory, the parent substance which produces radium must always be present in minerals containing radium. Uranium from the first appeared to be the most probable

* Communicated by the Author, having been read before the British Association, Leicester, August 1907. Previous accounts of the results were given in letters to 'Nature,' Jan. 17 and June 6, 1907.