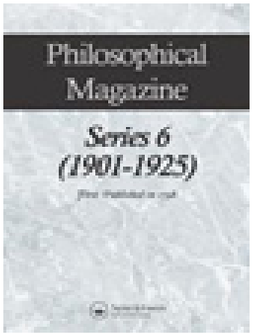

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XII. On the radioactive matter present in the atmosphere

A.S. Eve M.A.

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energy per cubic cm. of vibrations of frequencies between k and $k+dk$, is

$$\frac{RT}{\pi^2 V^3} k^2 dk,$$

or, in terms of the wave-length λ in free æther, the energy of wave-lengths between λ and $\lambda+d\lambda$ is

$$8\pi RT\lambda^{-4}d\lambda \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

This is one-eighth of the amount found by Lord Rayleigh ('Nature,' May 16th), but agrees exactly with that given by Planck (Drude's *Annalen*, iv. p. 553) for large values of λ. It seems to me that Lord Rayleigh has introduced an unnecessary factor 8 by counting negative as well as positive values of his integers ξ, η, ζ. From formula (7), it follows that the total energy of radiation at temperature T is

$$8\pi RT\int_{\lambda_0}^{\infty} \lambda^{-4}d\lambda + \int_0^{\lambda_0} f(\lambda, T, t)d\lambda, \quad . \quad . \quad . \quad (8)$$

in which λ_0 is the shortest wave-length for which the vibrations may be supposed to possess their full energy, and the second integral represents the energy of waves of wave-length less than λ_0, the energy of radiation of these waves being a function not only of T and λ, but also of t, the time which has elapsed since the closing in of the æther. Formula (8) does not, of course, claim to express the partition of energy in the radiation emitted by a hot solid: it is the radiation when a mass of gas has been shut up for time t in a perfectly reflecting enclosure. And the formula applies only to the continuous spectrum of the gas produced by molecular motions; no account is taken of the line spectrum, produced, so far as we know, by atomic vibrations.

XII. *On the Radioactive Matter present in the Atmosphere.*

By A. S. Eve, *M.A., McGill University, Montreal* *.

THE presence of radium in the earth, and of the emanation of radium in the atmosphere, has been well established. Exact measurements of the amount of the radioactive substance in the air are, however, needed. Professor Rutherford was kind enough to propose to the writer some methods and experiments by which to estimate the quantities present, and to throw further light on their effects.

The main objects of these investigations were:—

1. To estimate the amount of radioactive matter present in

* Communicated by Professor E. Rutherford, F.R.S.

a known volume of the atmosphere, measured in terms of the mass of radium required to maintain the supply constant.

2. To ascertain if the natural ionization of the air can be entirely attributed to the radioactive matter present in the atmosphere.

3. To determine the rate of formation of ions due to the active matter in the air.

4. To find the distance from which active matter can be collected on a wire maintained at a high negative potential.

I. *On the Amount of Radioactive Matter present in the Atmosphere.*

If a wire is raised to a high negative potential for two or three hours in the open air, it is known that it collects active matter from the air. The rate of decay of the activity of the deposit thus obtained approximates closely to the rate of decay of the matter similarly collected from the emanation of radium.

It is not necessary to repeat here the summary of evidence collected by Professor Rutherford in the last chapter of 'Radioactivity.' It appears certain that the radium in the earth gives rise to radium-emanation in the atmosphere, and that the emanation in turn disintegrates successively into the three products of rapid decay, radium A, B, C. Bumstead has accurately compared the decay of the active deposit obtained from the air of Newhaven, Connecticut, and has shown that it must be ascribed to radium. He has also observed in the same locality the presence of thorium emanation. It may be of interest to compare the rate of decay of the active deposit obtained from the emanation of radium, as given by Professor Rutherford in his Bakerian Lecture *, with the rate of decay of the active deposit derived from the atmosphere in Montreal, as determined by the present writer

These results are shown in Table I. and in the curve (fig. 1).

It is possible that some of the observed difference is due to the presence in the air of active matter from radioactive elements other than radium, such as thorium, but in any case the difference is not large, and it may be partly experimental.

In order to measure the quantity of emanation present in a given volume of the atmosphere, a simple method of comparison was employed. A negatively-charged wire was

* Phil. Trans. Roy. Soc. ser. A, vol. cciv. pp. 169–219.

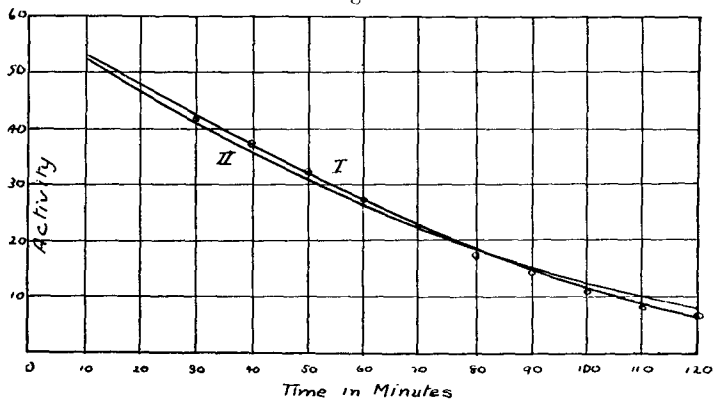
used to collect the active deposit from a known volume of air, and the amount was measured by an electroscope. A known weight of pure radium bromide in solution was then

TABLE I.

Decay of Excited Activity.

Time in minutes.	Radium Emanation. Rutherford.	Atmospheric deposit. Eve.	Difference.
10	52	52	.....
20	45·4	48	+2·6
30	40·4	42·5	+2·1
40	35·6	37·5	+1·9
50	30·4	32·5	+2·1
60	25·4	27·8	+2·4
70	.....	22·8	.....
80	17·4	18·5	+1·1
90	.....	14·5	.....
100	11·6	11·0	− ·6
110	.....	8·0	.....
120	7·6	6·6	−1·0

Fig. 1.



taken, and the resulting emanation was collected and transferred to another closed vessel; the excited activity was again obtained on a negatively-charged wire and measured by the same electroscope. In this way the emanation in the air was measured in terms of the amount of radium which would be required to produce it. In all cases the charged wire was exposed for almost three hours, so that practically

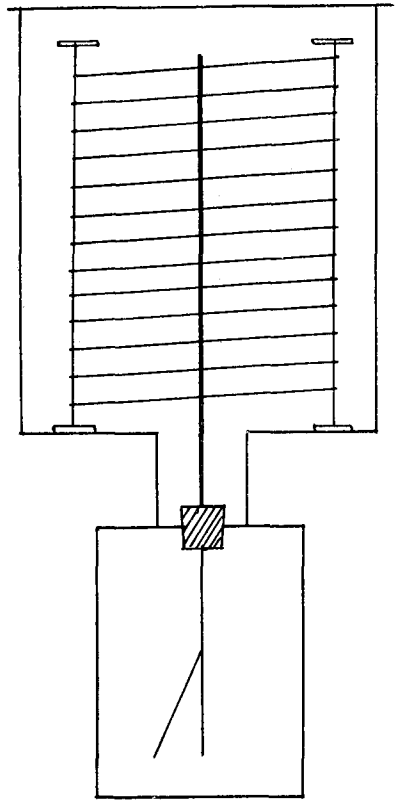
a maximum amount of activity was obtained. A Wimshurst influence-machine, driven by a small electric motor, was used, and the potential of the wire varied from about −9000 to −11,000 volts. It was necessary to work with a large vessel in a building into which no radioactive matter had been introduced. The conditions were satisfied by the Engineering Building at McGill University, which is situated at a considerable distance from the Physics Building. In the hydraulic laboratory there is a large water-tank 808 cms. high, and 152 cms. square, with a total volume of 18·7 cubic metres. The sides are made of iron 3·5 cms. thick. The tank is filled from time to time with water supplied by the City of Montreal, and derived from the rivers St. Lawrence and Ottawa. The river water is practically free from radio-active matter, nor could any appreciable activity be discovered in the mud and slime deposit left at the bottom. The tank was placed at the writer's disposal by the courtesy of the authorities of the Engineering Building, and a series of experiments were made, extending from November to April. In all cases the activity was measured ten minutes after the Wimshurst machine was stopped. For the sake of simplicity the activity is expressed in terms of the scale-divisions of the reading-microscope used to observe the fall of the gold-leaf of the electroscope. It may be noted that one scale-division was about equal to a fall of 3·6 volts, and that the capacity of the system as shown in fig. 2 was 3·5 E.S. units. The wire, after removal from the tank, was rapidly coiled on a metal reel, somewhat similar to the outer part of a large fishing-reel, and was then placed in a zinc cylinder connected to earth. Along the axis of the cylinder was a brass rod, insulated by a sulphur support, and directly connected with the gold-leaf system of an electroscope placed beneath the zinc cylinder.

The fall of potential per minute, due to the active matter collected on a wire 8 metres long placed in the iron tank, ten minutes after removal, corresponded to 3·6 scale-divisions. This is the mean of the results of a large number of observations, taken on different days, and the readings varied from 2·8 to 4·3. Thus the active matter collected from 18·7 cubic metres of air caused a fall of potential measured by 3·6 scale-divisions per minute.

A relatively small, air-tight, zinc cylinder 154 cms. high and 25 cms. in diameter was then taken, and the emanation derived from 2×10^{-4} mgs. of pure radium bromide was introduced into it. The volume of the cylinder was 76,000 c. cms., and the emanation was distributed through the

contained air by means of convection currents caused by heating the outside of the cylinder. After charging for three hours the wire was removed and wound on the reel,

Fig. 2.



and then placed as before in the zinc cylinder above the electroscope. The activity, ten minutes after the charge stopped, caused a fall of potential of 7·8 scale-divisions per minute. The solution of radium bromide, initially freed from emanation by passing a current of air through it, had been allowed to stand for fifteen hours in a closed vessel before introducing it into the zinc cylinder. Since the constant of change, λ, for radium emanation is about ·18, it can readily be shown that only one-tenth of the full supply of the emanation had been obtained from the radium bromide. Thus the total active deposit, which could be obtained from 2×10^{-4} mgs. of radium bromide in radioactive equilibrium,

corresponds to 78 scale-divisions ; and from 1 gramme of radium bromide to $3{\cdot}9 \times 10^8$ scale-divisions.

Hence we can determine the amount of radium bromide required to maintain the supply of emanation contained in 1 cubic kilometre of air at Montreal. For the emanation in the iron tank, whose volume is 18·7 cubic metres, caused a fall of potential corresponding to 3·6 scale-divisions, and 1 gramme of radium in radioactive equilibrium would supply emanation corresponding to $3{\cdot}9 \times 10^8$ scale-divisions ; therefore 1 cubic kilometre of the air contains emanation supplied by ·49 gramme of pure radium bromide.

Thus, making certain assumptions, we can form a rough estimate of the probable amount of emanation in the atmosphere. We can first of all calculate the amount of emanation, supposed equal in quantity per cubic metre to that in the air at Montreal, distributed in a spherical shell around the earth 1 kilometre high. Since the surface of the earth is 5×10^8 square kilometres, the amount in this shell corresponds to the emanation released from $2{\cdot}5 \times 10^8$ grammes of radium bromide in radioactive equilibrium.

Several observers have shown that the excited activity at high altitudes is equal to, if not greater than, that on the plains ; and it thus seems probable that this distribution of emanation might extend for at least 10 kilometres. In that case, the emanation in a shell 10 kilometres high must be released from $2{\cdot}5 \times 10^9$ grammes, or 2460 tons.

Now, three-quarters of the surface of the earth is covered with water, and if we further suppose that the emanation arises from the land alone, we obtain one-quarter of the above value, or 610 tons.

This is an estimate of the total amount of radium required to furnish the emanation in the atmosphere over the land of the earth's surface. It is probable that a vastly larger amount exists in the earth, since the greater part of the emanation would be transformed without ever issuing from the earth's surface.

An objection may be raised to the above estimate, inasmuch as the measurements were taken in the building, and not out-of-doors. It was found that air driven from the room through the tank did not affect the result, and the production of ions was proceeding at a very slow rate, as will be seen in Section III., so that there was no reason to suppose that the conditions within the building were different from those outside in respect to the amount of radioactive matter present in the air.

Nevertheless, it was decided to make some measurements

out-of-doors, and for this purpose a large hollow zinc cylinder was used, 610 cms. long and 77 cms. in diameter, having a volume of 2·8 cubic metres, or about one-seventh of that of the large iron water-tank. This cylinder was placed on the College Campus, at some distance from the Physics Building. The active deposit was collected as before on a negatively-charged wire along the axis of the cylinder. When the ends of the cylinder were left open, the amount of observed activity showed the usual variations, and was affected by the velocity of the wind, humidity, and the atmospheric conditions. When the ends were closed, fairly steady results were obtained.

A comparison of the results is given in the following table :—

TABLE II.

	Total activity in scale-divisions.	Activity per cubic metre in scale-divisions.
Zinc cylinder out-of-doors :		
(1) Ends open	·34	·12
(2) Ends closed	·15	·054
Zinc cylinder in the Engineering Building	·50	·18
Iron tank in the Engineering Building	3·6	·19

The ratio of the volumes of the tank to the cylinder is 6·7 : 1, and the ratio of the activities is 24 : 1, so that the excited activity collected in a vessel out-of-doors was between one-third and one-quarter of that collected within the closed tank in the Engineering Building. In order to see whether the effect was apparent or real, the wire cylinder was put in the Engineering Building, not far from the water-tank, and the excited activity was then exactly proportional to that obtained in the large tank. No satisfactory explanation of the difference between the values in the building and out-of-doors has at present been found, and the point will receive further investigation. Radioactive matter has not been introduced into the Engineering Building, and it will be shown later that the rate of production of ions, q, was as low as any observed.

If we take the values obtained in the cylinder out-of-doors, we must reduce the estimate of the radium bromide required to supply the emanation, in a shell 10 kilometres high over the land-surface of the earth, from 610 to 170 tons.

II. *On the Collecting Distance of a Negatively-charged Wire.*

An estimate was next obtained of the approximate distance from which excited activity can be drawn from the air to a wire raised to a potential of −10,000 volts. On a still day the amount collected on 6 metres of wire was almost the same whether the wire was suspended at some distance from the ground, or was placed within the zinc cylinder with open ends. In the cylinder the potential gradient was steeper, but the wire was to some extent screened from slight air-currents. The collecting distance was therefore approximately equal to the radius of the cylinder, or to 40 cm.

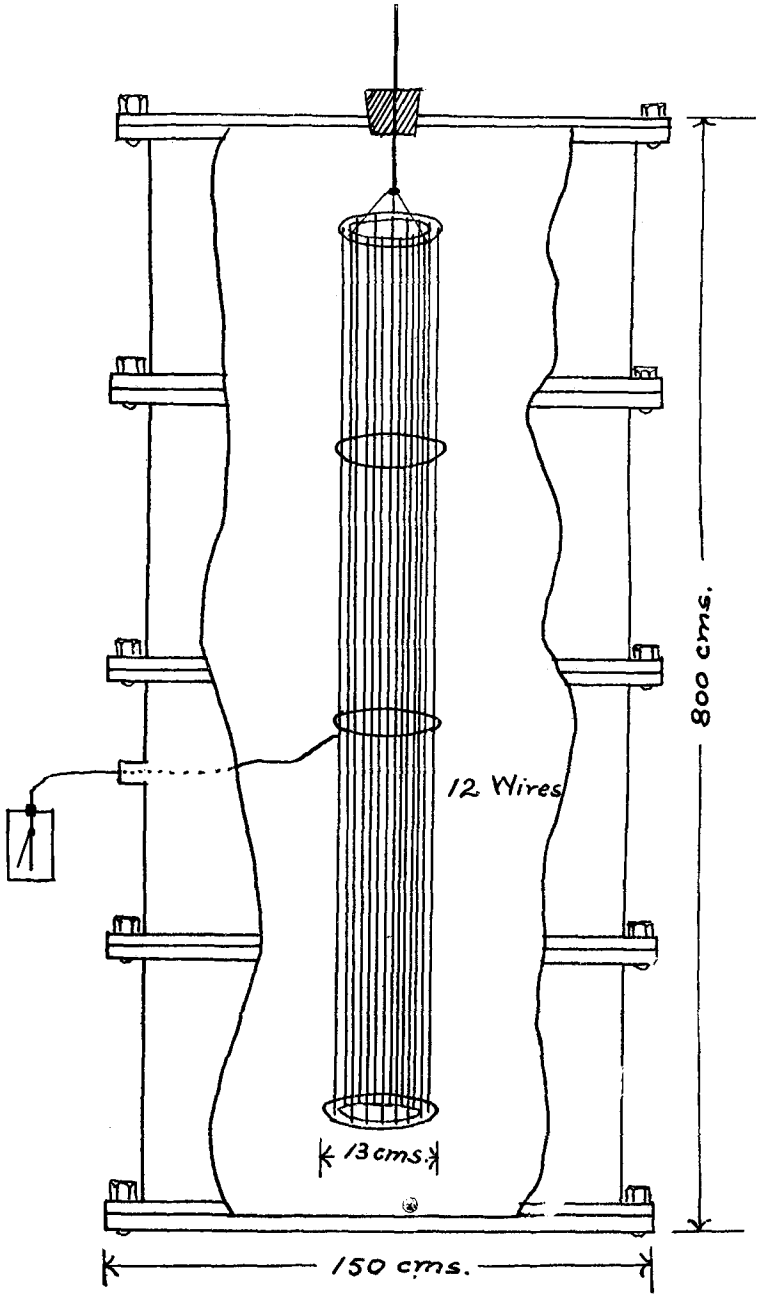
Two wires were then suspended in the air about 90 cms. apart, and the third wire was fixed at a distance of 10 metres from this. It was found that all three wires collected almost the same amount of excited activity, so that the two neighbouring wires were not drawing radioactive matter from overlapping volumes. Similar experiments were made with wires hung in the Engineering Building at various distances from the outside of the tank. The experimental evidence is not easy to summarize, but the general conclusion was obtained that a wire raised to about −10,000 volts collected activity from a cylindrical volume of about 40 to 80 cms. radius. This result is not in agreement with previous suppositions, for some observers have wrongly concluded that the collecting distance is very great.

III. *On the Rate of Production of Ions.*

It is important to obtain as many determinations as possible of the value of q, or the rate of production of ions per cubic centimetre of the air. A well-insulated wire was therefore hung down the middle of the large iron tank in the Engineering Building, and the wire was connected to the gold leaf system of the electroscope used throughout the experiments, but from which the zinc cylinder was now removed. On charging the wire to two or three hundred volts, it was found that a saturation current was not obtained. Twelve wires were therefore taken (fig. 3), and fastened to a few zinc disks, 13 cms. in diameter, so that the wires were along the generating lines of a cylinder of that diameter. This system of wires, connected to the electroscope, and charged to 300 volts, was found to give a saturation current.

By this arrangement, the whole tank practically became a very large electroscope, and the discharge of the central wires was due to the radioactive matter present in the air within the tank. The capacity of the system was found by connecting

Fig. 3.



the wires to a condenser of known capacity consisting of two concentric cylinders, and by observing the potential, both before and after the connexion.

The subsequent calculations are similar to those given by Rutherford in 'Radioactivity,' page 72.

Since S, the volume of the tank, equals $18{\cdot}7 \times 10^6$ c.c., and C, the capacity, was 140 E.S. units, the current, i, is equal to CV, when V is the fall of potential per second; q is the number of ions produced per second in a cubic centimetre, and e, the charge on an ion, is taken as $3{\cdot}4 \times 10^{-10}$. But the gold-leaf fell at a rate measured by 2·3 scale-divisions of the microscope in one minute, and a scale-division represented 3·6 volts or $\frac{3{\cdot}6}{300}$ E.S. units.

Hence $$i = 6{\cdot}44 \times 10^{-2},$$

and $$q = \frac{i}{Se}$$
$$= 10{\cdot}1.$$

A large number of similar experiments were made, and the values ranged from 9·1 to 10·2, with a mean value

$$q = 9{\cdot}6 \text{ ions per c.c. per second.}$$

This is the smallest value yet obtained, and I think that the error is not more than five per cent. H. L. Cooke, using a well-cleaned brass vessel, found $q = 10$ in the Physics Building at McGill University about two years ago. Professor Schuster, in the laboratory at Manchester, found $q = 12$. These small values for q are only obtainable when the instruments employed are well-cleaned, and removed from the neighbourhood of radioactive substance.

IV. *An Estimate of the Total Depth in the Earth from which the Radium Emanation passes to the Atmosphere.*

Professor Rutherford has given in 'Radioactivity' an estimate of a higher limit to the amount of radium which can be present in the earth. If this amount were exceeded, the temperature of the earth would have a value in excess of that observed. He calculates that, on an average, not more than $4{\cdot}6 \times 10^{-14}$ grammes of radium can be present per one gramme of the earth's constitutents. But the mass of the earth is $6{\cdot}1 \times 10^{27}$ grms. Hence the total amount of radium in the earth cannot exceed 28×10^{13} kilos, or 28×10^7 British tons.

The consensus of observers seems to show that the emanation

in the air escapes from the earth ; and if we suppose that radium is uniformly distributed to the above amount, we can readily calculate the depth from which the emanation must freely come in order to keep up the supply in the atmosphere.

Let x be the average depth through which that radium is distributed which gives rise to the emanation in the atmosphere. Since 610 tons of radium suffice to produce the amount of emanation over the land of the globe, we have

$$\frac{x \times \pi r^2}{\frac{4}{3}\pi r^3} = \frac{610}{28 \times 10^7},$$

where r is the earth's radius, and since

$$r = 6{\cdot}4 \times 10 \text{ cms.}$$

$$x = 18 \text{ metres.}$$

If we take the lower estimate of 170 tons of radium, the corresponding depth is about 5 metres.

Emanation arising from greater depths than these would probably disintegrate before reaching the surface of the earth.

IV. *On the Cause of the Natural Ionization observed in the Atmosphere.*

The question arises whether the natural ionization of the air at the surface of the earth is due to the radioactive matter contained in it, and whether this cause will wholly account for the effect, or whether there are other causes at work, known or unknown in character.

Take, for example, the discharge of the cylinder of wires suspended in the large iron tank in the Engineering Building. This wire lost its charge at the same rate whether it received initially a positive or negative charge. The saturation current from the wire to the sides of the vessel was $6{\cdot}44 \times 10^{-2}$ E.S. units, and the rate of production of ions was given by $q = 9{\cdot}6$ per c.c. in a second. Is such ionization of the atmosphere to any extent an inherent property, or would air entirely free from radiating matter, or its influence, cease to produce ions, and would it become a non-conductor? The only ionizing agents under such conditions are (1) radiation due to radioactive matter contained in the air, (2) radiations due to active matter on the surface, or in the material of the sides of the

vessel, (3) penetrating radiation through the sides of the vessel, due to radioactive matter in the surrounding bodies.

In the present experiments, the sides of the tank consisted of iron one inch thick, and were sufficient to cut off all but γ rays coming from without. The deposit on the sides of the tank could not have been appreciably radioactive, or the low values of q (9·6) would not have been obtained. For in no apparatus, however carefully cleaned, has a lower value of the volume ionization been noted. The chief factor, probably the only factor, in producing the discharge of the wire is the emanation in the air, and the successive products of rapid decay. And of this there is undoubted evidence, inasmuch as the excited activity was actually collected on a negatively-charged wire in the tank.

The experiment shows that the radioactive matter is in the tank, and the only question is whether it is sufficient to account for the effect obtained.

In order to test this point, Professor Rutherford proposed to me a new method of directly determining this important point. I venture to describe it at some length, because it is desirable that similar experiments under various conditions should be carried out in other places. Such a radioactive survey of the atmosphere would be of interest at the present time.

If the radioactive matter in the air is the sole cause of the ionization observed, there should be a direct and proportional relation between the excited activity and the ionization ; for both these effects must originate from the same cause, namely, the emanation present at any given moment in the mass of air under observation.

The method employed was as follows :—The natural ionization of the air in the tank was observed, and the active deposit in the tank was then collected ; the first with a cylinder of wire connected with the electroscope (as in fig. 3), the second on a wire suspended in the tank, and charged to −10,000 volts. The wire after removal was wound on the reel, and measured by the same electroscope, as in figure 2.

Thus, if C_1 be the capacity of the cylinder of wires in the large tank, V_1 be the potential fall per minute in the electroscope whose gold-leaf system is connected with the central wires, then C_1V_1 is a measure of the natural ionization.

Again, if C_2 be the capacity of the electroscope, fitted with the upper cylinder and reel as in figure 2, and if V_2 be the fall of potential per minute due to excited activity on a negatively-charged wire, as observed ten minutes from the

cessation of the charge, then C_2V_2 is also a measure of the emanation present in the tank. Therefore, whatever may be the amount of emanation, whether naturally present or artificially introduced, we should have, under all circumstances, a constant value for the following percentage, namely,

$$\frac{C_2V_2}{C_1V_1} \times 100 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

The constancy of the quantity must, of course, depend upon the current being fully saturated, and on the active deposit being completely collected in every case.

If a smaller vessel be now filled with the emanation of radium, so as to give a discharge large compared with that due to the natural ionization, the same value for (1) should be found for the air mixed with emanation in this small tank, as for the natural air in the large iron tank.

The advantages of this method are due to the fact that the measurements are all taken with the same electroscope, and may be expressed in terms of scale-divisions of the same instrument, which need not even be disturbed from its position.

Now, in the actual experiment with the iron tank,

$$C_1 = 140, \qquad C_2 = 3{\cdot}5 \text{ E.S. units.}$$

$$V_1 = 2{\cdot}17, \qquad V_2 = 3{\cdot}6 \text{ scale-divisions per minute.}$$

$$\therefore \quad \frac{C_2V_2}{C_1V_1} \times 100 = 4{\cdot}1.$$

This result is the mean of many experiments extended over some months.

The emanation from radium bromide was next blown into a small tank of 80,000 c.c. capacity, and a similar series of experiments were made. C_1 was now equal to 21, and C_2 to 3·5, and the following values of these quantities were obtained for various strengths of the emanation:—

V_2.	V_1.	$\frac{C_2V_2}{C_1V_1} \times 100$.
1·44	4·4	5·5
0·93	2·6	6·0
1·85	6·3	4·9
1·32	5·8	4·4
	Mean.....	5·2

Thus we obtain 4·1 from the natural air in the iron tank, and 5·2 from the air mixed with the radium emanation in the smaller vessel. These figures are of the same order, and the difference between them is probably caused by the difficulty of drawing all the excited activity to a wire charged even to −10,000 volts from a tank whose sides measure 150 cms. These results show that the ionization in the large tank was very largely due to the presence of radium emanation, and cannot be ascribed to radiations from the walls of that vessel. But the results obtained were not altered after air had been freely driven through the tank, and we thus come to the conclusion that the rate of production of ions in the air of the tank was probably the same as from the air outside the building; and if such is the case, we have seen that the ionization can mainly be attributed to the presence of radioactive matter in the air itself. Since the activity is due to the emanation which decays to half value in four days, its amount cannot have sensibly decayed in passing from the outside air to the tank.

The results strongly indicate that the radioactive matter in the atmosphere near the earth will fully account for the rate of production of ions observed in it.

V. *Summary.*

The general conclusions derived from these experiments are as follows:—

(1) An estimate of the amount of radium required to maintain a steady supply of the emanation in one cubic kilometre of air near the earth's surface, lies between ·14 and ·49 gramme.

(2) This amount of emanation and its successive products cause a production of ions at the rate of about 9·6 per cubic centimetre per second.

(3) The radium emanation in the air is probably sufficient to account wholly for the natural ionization observed in large closed vessels consisting of non-radioactive materials, and for the rate of production of the ions in the atmosphere near the earth.

(4) The collecting distance of a wire charged to −10,000 volts is about 40 to 80 cms. The active matter derived from the carriers is not drawn in appreciable quantities from still air at a greater distance.

(5) Assuming that the radium in the earth is equally distributed in amounts sufficient to maintain the temperature gradients actually observed in the earth, then the radium emanation in the air is derived from an average depth estimated to be between 5 and 17 metres.

All these estimates and results are based on the assumption that the conditions at Montreal are normal, and represent the average. It must be remembered that the country was covered with snow from one to three feet deep, and that the temperature was sometimes as low as 10 degrees, or 15 degrees below zero Fahrenheit. But after the snow had melted, and the frost had left the ground, the active deposit on a wire in the zinc cylinder on the College Campus was almost equal in value to that obtained in the depth of winter. Allan also found that the summer and winter values at Montreal showed but slight variation.

It is most desirable that experiments should be repeated elsewhere similar to those described in this paper, particularly that suggested by Professor Rutherford, and described in Section IV. If any investigators have at their disposal a very large tank which can be rendered absolutely air-tight, it would be interesting to observe whether the natural leak of a well-insulated wire suspended in it and connected to an electroscope would fall in a few days at the same rate as the decay of radium emanation. Fresh air could then be introduced from without, and the experiment repeated. Such an investigation would determine absolutely whether the natural ionization is entirely due to the emanation present in the air. There might be a small residual ionization due to the sides of the vessel. A large steam-boiler might serve as a tank. Observations at sea are also needed, both on the rate of production of ions, and on the excited activity which can be collected.

In conclusion, I am indebted and grateful to Professor Rutherford, both for suggesting the experiment, and for his advice, encouragement, and resourcefulness when difficulties occurred.

McGill University, Montreal.
3rd April, 1905.