SOME PROBLEMS IN RADIOACTIVITY.

By A. S. Eve, D. Sc.

In the month of May, 1908, by the kindness of the Captain and officers of the S. S. "Lake Erie," observations were made, between Montreal and Liverpool, on the radioactive deposit collected on a wire, charged negatively by a Zamboni dry pile, and exposed in the air for a few hours. The wire was insulated by ebonite rods, and suspended from the flag halyards. After exposure it was coiled on a skeleton reel and placed in an electroscope. The latter was clamped to a board, together with the observing microscope. There was no difficulty in obtaining satisfactory readings, in spite of the moderate motion of the ship.

The results obtained in mid-Atlantic did not differ greatly from those obtained in Canada or in England, but it will be remembered that the amount of active deposit determined at any given locality is liable to considerable variations. Such changes are apparently due partly to meteorological conditions, partly to fluctuations in the amount of emanation in the air.

To an arbitrary scale, after deducting the natural leak, the measurements of the active deposit were as follows:

5 May	Montreal	34
6 "	"	26
14 "	Ocean, lat. 50°, long. 45°	21
15 "	" " 52° " 38°	64
16 "	" " 54° " 30°	41
1 July	Hornsea, E. Yorkshire Coast	28
2 "	" " "	80
15 "	" " "	53
20 "	" " "	60
22 "	" " "	48
22 Aug.	Seascale, W. Coast, Cumberland	30
24 "	" " "	270

The large value at Seascale on 24th August was obtained on a vertical wire well exposed to a strong west wind. The small values at Montreal resulted from a horizontal wire on the roof of a house. An uncharged wire at sea gave no effect that could be detected in the electroscope.

It appears then that the active deposit due to radium can be found in mid-ocean to an extent which seems to indicate that the radium emanation, whose period is 3.8 days, can not be entirely

borne by wind from the land, but that emanation must enter the air from the ocean somewhat as from the land.

This is an unexpected result, for Strutt found the average contents of radium per c. c. of rock to be about 3.8×10^{-12} grams, and Joly[1] found the corresponding value of 1.6×10^{-14} grams per c. c. for sea water. It is true that the ratio is about 240 to 1, but on the other hand emanation escapes readily when radium is in solution, especially perhaps from a surface agitated as that of the ocean.

It was my intention to take further observations on the return voyage, and to make long exposures of the wire so as to detect the active deposit of thorium, if present. An accident to the electroscope prevented me from doing this.

It may be advisable to give a few details of the apparatus, all of which, except the microscope and dry pile, could be made quickly by an intelligent plumber at a moderate cost. A rectangular box of zinc, edge 12 cm., had a door behind to admit the reel which stood on 4 legs and was clamped in position by a small wooden wedge. There was a mica window in the door and another in the face opposite. A zinc rod at right angles to the front entered the box and carried the sulphur insulation, vertical rod and aluminum leaf. A wire, acting as charging key, also entered the front of the box through a small ebonite plug. Thus the aluminium leaf and charging key were not disturbed on inserting the reel and the wire around it.

The leaf moved twenty divisions an hour when the wire was free from active deposit. The electroscope and microscope were screwed firmly to a narrow board, and this was held in the hand and lodged against a port hole of the ship and levelled by the horizon. Thus pitching alone affected the position of the leaf, and the angle of pitch of the ship was not large. The reading obtained with a wire, ten minutes after exposure, amounted to as much as a rate of sixty divisions an hour. and the effect disappeared gradually in about two hours. In spite of the disadvantageous conditions on board ship it was, I think, impossible to have been mistaken in my result, that the active desposit of radium may be collected in considerable quantity, relative to that determined on land, even in mid-ocean. It would have been better to have had a stronger battery, for the dry pile gave but 300 volts. The ebonite rods were quite satisfactory for insulation in fair weather.

[1] Pesidential Address to Brit. Assoc., Dublin, 1908.

Other observers have found less active deposit near the sea coast than inland. For example, Elster and Geitel[2] obtained a value 6 for the Island of Juist near Denmark, 18.6 at Wolfenbüttel, 84 at Freiberg and 137 among the Bavarian Alps. Simpson[3] obtained 58 on the coast at Hammerfest, and 126 inland at Karasjok; and at the former station a sea breeze gave much less than a land breeze.

Again, P. H. Dike[4] found radioactive deposit on a negative wire off the coast of Alaska, and in Cook Strait, New Zealand, but stated that in the open sea no increase in the rate of discharge of the electroscope used for testing the exposed wire could be detected. As this result is not in agreement with mine it is clear that further determinations are needed either on shipboard, or at places on the west coast of continents when the wind is blowing wholly from the sea.

The large value found by me at Seascale on 4th August, with a strong west wind, is noteworthy. This wind had, however, passed over Ireland and the Isle of Man.

Observations were also made by Dike[4] on the conductivity of the atmosphere, using Gerdien's apparatus. Like most other observers he found that generally there appear to be more positive than negative carriers in the air near the Earth's surface. But reverse effects are sometimes found, when negative ions exceed positive. Such cases are reported by Dike in the "doldrums" and by Smirnov[5] during balloon ascents.

The positive and negative ions are so often weighted with dust, vapor or gas molecules that all methods of determining their numbers are uncertain, and it appears to be still an open question whether there are actually more positive than negative ions in the air near the Earth's surface. If there is an excess of positive ions an explanation of the excess appears to be still wanting.*

[2] *Physik. Zs.* Leipzig 5, (11-20.)

[3] *London. Phil. Trans. R. S.* Ser. A., 205, 1905, (61-97.)

[4] *Terr. Mag.*, Washington, D. C., 13, (119.)

[5] *Acad. Sci. St. Petersburg*, Bull. 9, 759, 15 May, 1908.

* Some recent experiments by Mr. F. W. Bates and the writer indicate that: (1) The quantities of positive and negative electricity in the atmosphere do not differ to the extent indicated by Ebert's apparatus. (2) That the apparent excess of positive electricity is either due to diffusion, or perhaps to the fact discovered by Townsend that newly generated positive ions have sometimes lost two electrons. Thus one positive ion may be formed and two negative ions, and the former may therefore be initially more mobile; and in consequence an excess of positive electricity may be indicated in Ebert's or Gerdien's apparatus, if the current is not saturated.

RADIUM EMANATION IN THE ATMOSPHERE.

The first attempt to measure in terms of radium the amount of emanation in the atmosphere was described by the writer in a communication to the *Philosophical Magazine*, July, 1905. Charged wires were placed in a large cylinder out of doors, and also in a larger iron tank in the Engineering Building, McGill University. The active deposit was measured in an electroscope within which the wire was coiled. Another and shorter wire was exposed in a small cylinder containing the emanation from a definite amount of radium bromide taken from a standard solution. The emanation per cubic kilometer of the atmosphere was found to be such as would be in equilibrium with between .14 and .49 gram of radium bromide. Unfortunately, the first standards soon proved inaccurate, because the radium bromide adhered to the glass of the vessel containing the solution. When weaker solutions were made, they therefore proved too weak. This trouble was obviated by adding hydrochloric acid to the standards subsequently prepared. The values given above should perhaps be halved and, if so, they would lie between 41 and 143, 10^{-12} grams of radium per cubic meter. This magnitude agrees fairly with more recent and direct determinations of the radium emanation in the air. The agreement indicates that the emanation and active deposit of radium are in radioactive equilibrium in the atmosphere.

Since that time direct measurements of the radium emanation in the atmosphere have been made by absorbing it in coconut charcoal or by condensing it with liquid air. The determinations thus made are in good agreement, considering the diversity of local conditions and the fluctuations in quantity at a given spot.

The results are as follows:

Satterly[6]	Cambridge	100
Ashman[7]		89
Eve[6]	Montreal	60

The radium equivalents are here stated in multiples of 10^{-12} grams of radium per cubic meter. In other words, there are on the average about 1.4 atoms of radium emanation in a c. cm. of the atmosphere, near the Earth's surface.

[6] *Phil Mag.*, London, Oct. 1908.

[7] *Amer. J. Sci.*, New Haven, Conn., **26**, Aug. 1908.

Ionization due to Radium Emanation.

The view advocated in 1905 by the writer was that the ionization of the atmosphere in large vessels could be mainly attributed to the active matter in the contained air, provided the penetrating radiation was excluded. The number of ions thus produced by the active matter in the air was estimated at 9 per c. c. per second—an overestimate, as subsequent work proved. In the *Philosophical Magazine* for October, 1908, Satterly gives a revised estimate of 2.5 ions per c. c. per second. But if we take Rutherford's more recent value for the α particles projected from a gram of radium per second, namely, 3.4×10^{10}, and the average number of ions produced by an α particle from *Ra. Em.*, *A*, and *C* as 130,000, then the number of ions produced per cubic meter per second is

$$100 \times 10^{-12} \times 3 \times 3.4 \times 10^{10} \times 1.3 \times 10^{5}$$

or about 1.3 ions per c. c. per second.

The effect due to the β and γ rays is probably but a moderate percentage of this value, inasmuch as the α ray's energy is much greater than the combined β and γ rays' energy.

Since the active deposit of thorium on charged wires exposed for two or three days gives radioactive effects not infrequently from 40 to 70 per cent initially of those due to radium active deposit, there is an uncertain addition to be made to the above. But, all told, the productions of ions in the atmosphere due to all the radioactive matter therein does not seem to exceed 3 ions per c. c. per second.

Ionization Theory.

In the atmosphere if n_1 is the number of positive, and n_2 the number of negative ions present at a given instant per c. c., then in a steady state the number forming per second and the number combining per second, in a cubic centimeter, may be denoted numerically by $\frac{dn}{dt}$, or q, and this varies as $n_1 n_2$. Or if the numbers n_1, n_2 are nearly equal, $\frac{dn}{dt} = \alpha n^2$, where α is the constant of recombination.

The value of α has been determined by Langevin, McClung and Townsend, and the mean value of their determinations for air is $3330e$, where e is the ionic charge. Taking the old value of e, 3.4×10^{-10}, we have $\alpha = 1.5 \times 10^{-6}$. Hence the following values should correspond:

q	n
1.5	1000
6.0	2000
13.5	3000

In measuring n the same value of e may be taken. Using an Ebert apparatus the value of n fluctuates considerably from time to time, but it is usually from 1000 to 2000. Langevin found that the large loaded ions escape detection by Ebert's apparatus, so that we may perhaps take n as large as 2500, and in that case $q = 9.4$, or approximately 10.

It has been shown that the radioactive matter in the atmosphere may account for a value of $q = 3$. Whence come the remaining 7 ions produced per c. c. per second? The answer may be given that such ionization is caused by the penetrating or γ rays proceeding from the radium and thorium products known to be present in the ground. This penetrating radiation was first foretold by Rutherford, and then detected and measured by Cooke[8], who showed that when an electroscope was screened with large masses of lead the current or natural leak of the electroscope decreased 30 per cent. The addition of more lead did not cause further decrease in the natural leak. Lead itself usually contains active matter and the screens may have contributed a little to the leak, though obviously they screened far more than they caused.

The penetrating rays on striking or passing through a metal plate give rise to secondary rays, and the ionization due to such rays may be as large as that due to the penetrating rays themselves. An example of this may be given:

If 14 mg. of pure radium bromide be placed three meters from electroscopes made of different metals, then the value of q within the vessels are as follows:[9]

Substance	Thickness	Atomic Weight	q
Lead	1.6	207	550
Lead	2.7	207	482
Zinc	0.57	65	351
Copper	0.57	63.5	345
Iron	0.60	56	320
Aluminium	0.40	27	297

The values of q follow the order of atomic weight as we might expect from McClelland's work. In the same way the penetrating radiation from the radium C and thorium C in the Earth will produce more ionization in a metal vessel of high than of low atomic weight, and more in a close vessel with moderately thin walls than in the open air.

[8] *Physic. Rev.* Lancaster, Pa. Mar. 1903 **16** (183.)
[9] *Phil. Mag.*, Sept. 1906.

If Q grams of radium produce N ions per c. c. per second at a distance r from the radium, then $N = K \frac{Q}{r^2} e^{-\lambda r}$ where K is a constant and λ is the co-efficient of absorption of the γ rays by air.

The total number of ions produced by one gram of radium would, therefore, be:

$$\int_0^\infty K \frac{4 \pi r^2}{r^2} e^{-\lambda r} dr = \frac{4 \pi K}{\lambda}$$

I have calculated the value of K from the experiments referred to above and deduced that the total number of ions which would be produced by the γ rays from a gram of radium is about 9×10^{14}. The α rays from the radium emanation, A and C, in equilibrium with one gram of radium, would give about 1.3×10^{16} ions. Thus by this estimate the γ rays cause 7 per cent of the ionization as compared with the α rays. But near the Earth's surface we have to halve the γ ray effect, but not the α ray effect. Hence, the γ ray ionization due to the active matter in the *air* will be 3.5 per cent of the α ray ionization. This is the order of magnitude which might be expected from the heat experiments made by Rutherford and Barnes, and from the general theories of Rutherford which prove that the energy of the α particle exceeds considerably that of the β and γ rays combined.

If, however, we take the most penetrating rays, for which Wigger[10] found λ/density = .02, then λ = .000026, not .000044 as from McClelland's[11] work. With this smaller value of λ, the γ rays will give, near the Earth's surface, not 3.5 per cent, but 6 per cent compared with the α ray ionization, the ionization in each case being due to the radioactive matter in the air. This appears to me an over estimate, because all the γ rays are not of this penetrating type. Actual determinations of the co-efficient of absorption of the γ rays by air are certainly needed.

Experiments on the Penetrating Radiation.

But we are not compelled to consider this matter merely from a theoretical basis. Cooke endeavored to determine the direction from which the penetrating radiation comes by placing screens in various positions round the electroscope, and it appeared that the

[10] *Jahrb. der Radioaktivität*, **2**, 1906, (391).
[11] *Phil. Mag.*, London, July, 1904.

effect came equally from all directions. It is possible, however, that a screen above would give rise to secondary radiations from the lower surface. C. T. R. Wilson found the natural leak of an electroscope in a tunnel the same as in open air, but again the effect would be dependent on the radioactive matter present in the surrounding brick and rock. Geitel observed less penetrating radiation in a rock salt mine than at the Earth's surface, so that relatively the rock salt screened more than its contents radiated.

But the most important of recent observations are those made at the University of Toronto by C. S. Wright, described by McLennan in a letter to *Nature*, 30th April, 1908.

The values of q, the rate of production of ions per c. c. found over land, and over the ice and waters of Lake Ontario, were:

Electroscopes made of	q over land	q over lake
Pb	15.3	8.6
Al	13.4	6.0
Zn	12.5	6.35

On a spit of sand jutting into the lake q had an intermediate value 9. Since the waters of Lake Ontario may be described as practically free from radioactive matter they afford an ideal screen. The conclusion drawn is that the bulk of the penetrating radiation comes from the land, not from the air. The natural leak in the electroscopes is doubtless due in part to radiation from impurities in the metals comprising the electroscopes. It seems from these experiments that we shall not be far from the truth if we take $q=7$, due to the penetrating radiation from radioactive matter in the ground. This value will naturally vary somewhat according to locality.*

Add to the above the value of q due to the α, β and γ rays from radioactive matter in the atmosphere and we obtain $q=10$, whence $n=2500$.

If we include large and small ions in the atmosphere this value of n appears consistent with observations.

RATIO OF PENETRATING RADIATION VALUES FOR LAND, SEA AND AIR.

Assuming that the co-efficients of absorption of the more penetrating rays are proportional to the densities of the absorbing matter (Wigger) we are able to make good estimates of the relative mag-

*In the *Phil. Mag.*, London, Feb. 1909, received whilst correcting proofs, Mr. C. S. Wright, Toronto University, gives a much lower estimate for the ionization due to penetrating rays from the soil.

nitudes of the effects due to the penetrating radiation from the active matter in earth, sea and air.

Let Q be the amount of radium C present per c. c. of the atmosphere, and let Q be expressed in terms of the amount of radium with which the radium C is in equilibrium. For an electroscope placed at the ground level we have for the number of ions produced per c. c. per second:

$$N = \int_0^\infty K \frac{2\pi r^2 \, dr}{r^2} \cdot Q \cdot e^{-\lambda r} ,$$

$$= \frac{2\pi K Q}{\lambda} .$$

If accented letters denote corresponding quantities for the Earth, and a double accent for sea water, then

$$N' = \frac{2\pi K Q'}{\lambda'} , \quad N'' = \frac{2\pi K Q''}{\lambda''} ,$$

the same electroscope being used throughout.

$$\text{Hence,} \quad N : N' : N'' :: \frac{Q}{\lambda} : \frac{Q'}{\lambda'} : \frac{Q''}{\lambda''}$$

$$:: \frac{Q}{\rho} : \frac{Q'}{\rho'} : \frac{Q''}{\rho''}$$

where ρ, ρ', ρ'' are the densities of soil, air and sea water, namely, 2.7, .0013, 1.1, 1.0 and Q, Q', Q'' are 3.8×10^{-12}, 1.0×10^{-16}, 1.6×10^{-14}, (Strutt; Eve, Satterly, Ashman; Joly.)

$$\text{Hence,} \quad N : N' : N'' :: 14{,}000 : 770 : 157 ,$$

$$\text{or} \quad :: 89 : 4.9 : 1 .$$

Thus the γ radiation from radium C in the Earth will produce about 18 times as many ions as that from the radium C in the air. Let us take $q=7$ for the penetrating rays from the Earth, then $q=0.4$ for the penetrating rays from the air. This agrees very well with our previous deductions.

But now we have to consider an important and serious difficulty. The penetrating rays from radium C in sea water will give only one-fifth of the ionization of the radium C in the atmosphere, or $q=0.4$ for the air; and $q=.07$ over the ocean. And here we are assuming that the radium emanation over the ocean is as abundant as over the land, whereas it is most probably less abundant. By addition we have $q=7.4$ for land and air, and $q=0.47$ (or less) from sea and

air. If we add $q=3$ for α rays and suppose this the same over land and sea (thus favoring the latter) we get as final values $q=10.4$ and $q=3.47$ for the same electroscope over land and sea respectively. The value for the latter has been purposely overestimated. But $q=\alpha n^2$ so that

$$n^2 : n'^2 :: 10.4 : 3.47 \quad ,$$
$$\therefore \quad n : n' :: 1.76 : 1$$

It is clear then that the number of ions present at any instant over the land should be nearly twice as great as over the ocean. But many experiments have been made with the apparatus of Ebert and of Gerdien measuring the ionization and conductivity of the air over both land and sea. For example, by Boltzmann, Eve and Dike; and they certainly do not find any such disparity. Thus we arrive at a fundamental difficulty:

Whilst the radioactive matter known to be present in the earth and air is sufficient to account for the observed ionization in the atmosphere near the Earth's surface, the amount present in sea and air appears to be inadequate to account for the ionization of the air over the surface of the ocean.

It is apparently useless to look to the formation of spray, or to the Sun's light, to account for the ionization over the ocean. The ionization is large in calm weather; and Simpson's work during the long Arctic night indicates that sunlight has little influence.

It is premature to suggest any explanation of this difficulty, which must be attacked by further experimental evidence. Until this is done the radioactive theory of the ionization of the atmosphere is not on a satisfactory basis, although it appears to have too firm a footing to allow of doubt as to its general correctness.

It is fair to state that the difficulty would be lessened if W. W. Strong were correct in supposing that the penetrating radiation came equally from the air and the land. This view would entail the belief that the experimental results obtained over Lake Ontario were incorrect, and it would involve the assumption that the γ rays penetrate the air much more readily than the density law warrants us to expect.*

It is not probable that thorium is present in sea water in excess of the amount present in rocks, or sufficient to explain the above difficulty. Nor can we invoke penetrating radiation of the γ type coming to our planet from an external source, for such rays would be absorbed by the atmosphere before nearing the Earth's surface.

*GÖCKEL and WULF have recently published an account of experiments which support the view that the penetrating radiation comes mainly from the earth. The ionization in an electroscope was little affected by elevation, but was nearly doubled when placed inside the Simplon Tunnel.

Thorium Products in the Atmosphere.

If a negatively charged wire is exposed for a few hours in the air it is easy to prove the presence of the active deposit of radium (Elster and Geitel). If the wire is exposed for about three days it is found that the active deposit of thorium is also present, (Bumstead and Wheeler). Moreover, initially the activity of the active deposit of thorium may amount to 40 or even 75 per cent of that due to the active deposit of radium. This result has been confirmed wherever experiments have been made to test the result. It is astonishing that the active deposit of thorium is present to so large an extent relatively to the active deposit of radium. For the period of radium emanation is more than one hundred times that of thorium emanation, and it is only whilst in the gaseous state that the respective products can pass from the earth to the air. Dadourian has made a calculation that the ratio of the number of atoms of radium emanation to the number of atoms of thorium emanation in the air is about 30,000 to 1. But this method of stating the result, though correct, is misleading, because the rates of decay of the two emanations are so unequal. If we reduce the ratio to that of the equivalents of uranium and thorium the ratio becomes 1300 to 1, and if the chance of the escape from the soil of an atom of radium emanation is one hundred times that of an atom of thorium emanation, then the amount of uranium in the Earth near the surface must be about 13 times the amount of thorium. In fact, had we a method for detecting thorium, such as we have for radium, there seems little doubt that we should find as much thorium as uranium distributed through the surface rocks of the Earth.* Now Pegram and Webb[12] have measured the heating effects of thorium in equilibrium with its subsequent products and find 2.1×10^{-5} gram calories per hour per gram, and they point out that the evolution of heat from radium bromide is 3.2×10^{6} times as great as that from thorium. But a gram of radium is in equilibrium with 2.64×10^{6} grams of uranium. Hence the heating effects of the thorium and of the uranium-radium families are not far from equal.

As a first approximation we may conclude (1) that thorium and uranium are present in the surface rocks and soils in nearly equal amounts, (2) that the heating effects due to them and their subseqent products (including the radium family with uranium) are also of the same order. Indeed Blanc, from his observations near Rome, finds that thorium is the more important as a heat producer in the Earth's outer crust. But Strutt found the radium in the surface rocks more than sufficient to account for the existing temperature gradients, and the presence of the thorium intensifies the difficulty.

* W. Wilson, working at Manchester University, has obtained figures with which mine are in general agreement (*Phil. Mag.*, Feb., 1909). Dr. McIntosh and the writer found at Montreal the ratio of the active deposits, thorium to radium, to be about the same as that found at Rome or Manchester.

[12] *Le Radium*, p. 271. Sept., 1908.

It is true, as H. A. Wilson[13] has pointed out, that the presence of the radioactive matter, in the observed quantities, throughout the whole Earth would produce a most gradual temperature rise, but the trouble is to account for the fact that the temperature conditions have arrived at their actual state with the thorium and radium present. Strutt has suggested that the character of the constitution of the Earth changes a few miles down, and his suggestion has been supported by Milne; this theory holds the field perhaps in default of a better.

If the Earth had consisted of a solid sphere of radium 70,000 years ago, then owing to the disintegration of the radium atoms the amount remaining would be the small quantity such as Strutt has detected in the average rock to-day. But radium is a subsequent product of uranium, so that, continuing the argument, if the Earth had consisted of a solid sphere of uranium 94 thousand million years ago it would account for the radium present to-day. The suppositions are not suggested as probable or possible, and the latter period is large, perhaps 500 times geological time. We are driven to the conclusion either that uranium must have formed from some heavier element by its disintegration—a hypothesis we may dismiss—or it must have been integrated from elements of less atomic weight. This reversal of radioactive disintegration probably occurs under conditions difficult to reproduce in a laboratory, and the process may be abrupt or deliberate. There can be little doubt that such a process has existed or does exist, but all efforts by heat or pressure or electrical methods to check or modify the disintegration process have hitherto failed, so that at present the building up of uranium from the lighter elements is profoundly obscure. The future may unfold possibilities which will cause a complete change of position in the present attitude towards the heating effects of radioactive matter in the Earth.

CONCLUSIONS.

Some of the problems awaiting solution are:

(1) *Is the number of positive ions in the atmosphere near the Earth's surface really greater than the number of negative ions?*
(2) *If so, how is the excess caused?*
(3) *What are the relative quantities over land and ocean of the products of radium and thorium?*
(4) *While the radioactive theory of the ionization of the atmosphere over land is on the whole satisfactory, the radioactive matter in the air and ocean appears to be too small to account for the ionization and conductivity of the air over the ocean.*
(5) *How has been obtained or how maintained the supply of uranium, from which the radium existing in the Earth has been derived?*

McGill University, Montreal,
December 31, 1908.

[13] *Nature*, London 77 (365.) Feb. 20 1908.