Physics. — *"Researches into the Radio-Activity of the Lake of Rockanje"*. By Miss H. J. FOLMER and Dr. A. H. BLAAUW. (Communicated by Prof. HAGA).

(Communicated in the meeting of September 29, 1917).

§ 1. *Introduction.*

In a treatise on the Lake of Rockanje [1] [2] I have communicated that an inquiry into the radio-activity of the samples collected by me was being made by an expert. Now that this inquiry, carried out by Miss H. J. FOLMER in the Physical Laboratory at Groningen, has led to fixed results, we wish to bring them before the public. I shall begin with a short explanation from what considerations this inquiry has been planned and elaborated so amply.

When in July 1915 I began my researches into the flora and history of the origin of the Lake of Rockanje I got acquainted with Dr. BUCHNER's researches, which showed what a surprisingly strong radio-activity the latter had found in the Rockanje mud according to his publication in the Chemical Weekly. [3] Subsequently Dr. B. G. ESCHER communicated to the Geological Mining Society a report [4] made in April **1914**. ESCHER discerned in the Rockanje mud organic and mineral parts and came to the hypothesis that the mineral parts and together with these the radio-active substance, had been blown into it from the dunes. In the said meeting and in the above mentioned treatise I have already pointed out that the large mass of mineral substance is nothing but clay, which has settled on the bottom of salt and brackish water in the form of fine slime and mixed with many organic remains.

If, therefore, this mud were so radio-active there would be a far greater chance that radio-active parts had been carried there as slime by the rivers from the mountains of Central-Europe, than that

[1]) The Lake of Rockanje in the Isle of Voorne, which forms part of the province of "Zuid-Holland". It is situated near Hook of Holland, separated from it by the Isle of Rozenburg.

[2]) A. H. BLAAUW (1917) Treatise. Royal Acad. of Sciences Amst. 2nd Section XIX N⁰. 3.

[3]) E. H. BUCHNER (1913) Chem. Weekly. Part 10 N⁰. 35.

[4]) B. G. ESCHER (1915) Report Geol. Mining Soc. Gen. Geol. Section Part 2.

they should belong to the small quantity of dust from the dunes carried into the lake by the wind. But as it must be very well possible to make out by radio-active measurements of samples from the soil, whether the radio-active substance present in that soil had entered into it from the side of the dunes or rather from the side of the slime-carrying rivers, I thought that it was better experimentally to settle a question which was geologically so important.

For that purpose I have collected a great number of samples as will be mentioned below. After Dr. BÜCHNER informed me, in answer to my letter, that he did not intend to continue these researches himself, I was happy enough to find Miss H. J. FOLMER at Groningen willing to undertake them. Consequently this communication is chiefly the result of researches made by her at Groningen.

First samples 2, 4, 6, 8, 9, 10, 11, 12, 13, 17, 20, 23 were destined for inquiry, taken from the boring made at Rockanje. From these it was to appear in what layer the activity was strongest, after that the geological constitution of those layers and the conditions under which they had been thrown down, had already been discussed before in detail. Samples 20 and 23 are, however, from layers which existed long before the origin of the Lake and of the isle of Voorne [1]). These samples are called: boring 2, 4, etc.

To these others were added:

R. Taken 25 m. to the North East of the Grotto at 40 cm. depth, consisting of brown, entirely organic mud mixed with some grains of quartz of at most 150—300 μ length.

A. Taken from the bottom of the water, on the side of the Noorddijk, behind the Windgat, where formerly the dike often burst, on the North West side [2]).

B. Taken from the bottom of a ditch close to the Molendijk in the Strijpepolder, in order to examine whether the mud at a short distance from the Lake is still strongly active.

C. Sand taken from the dunes nearest the Lake (at ± 800 m. distance) whereabout formerly the Swyn was situated.

D. Light-grey clay, taken at 25 cm. under the meadow of the Drenkeling behind the Vleerdam Dike of the Lake.

M. Slime, freshly settled, from the Meuse at Grave, which Mr. J. DEN DOOP from Grave kindly provided me with.

---

1) Cf. treatise pp. 50—55.

2) Cf. treatise pp. 90—93.

W. Slime, freshly settled, from the Waal at Nymegen, sent to me by the kind interference of Dr. P. Tesch.

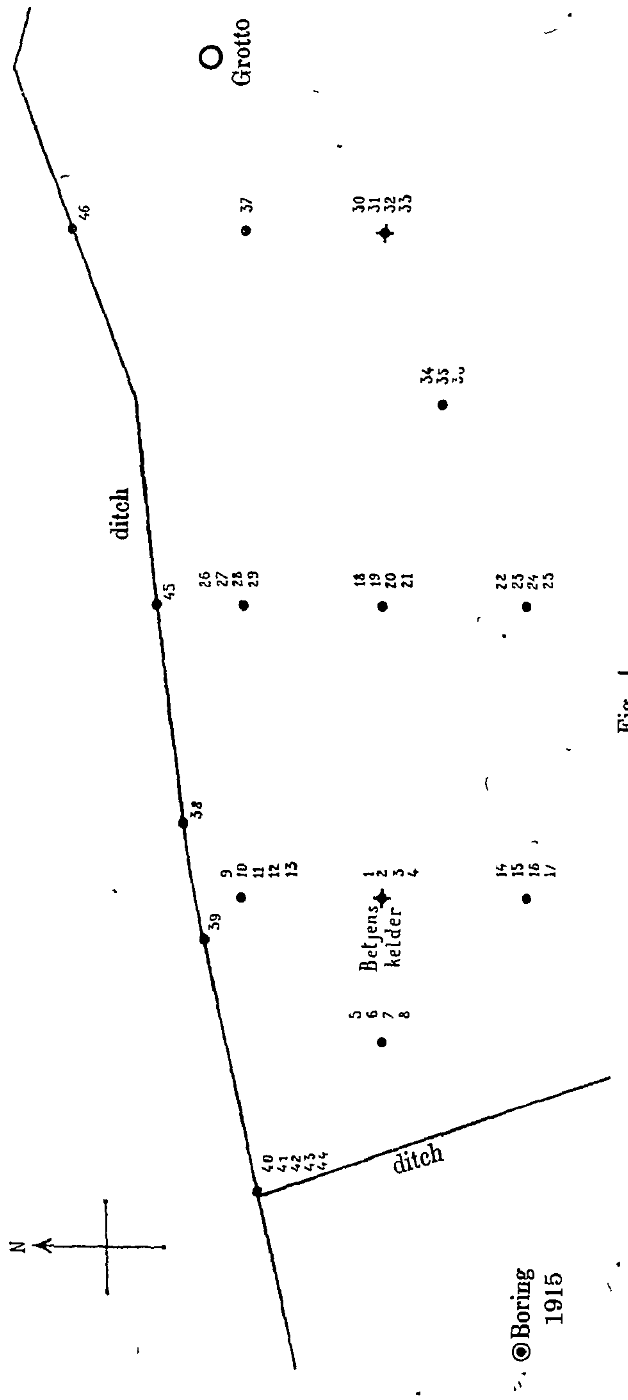


Fig. 1.

Sketch of the shallow borings. Scale $^{1}/_{1340}$.

● = spot of borings; the numbers indicate the samples.

✚ = two spots where, besides, samples I, II, and III had already been taken before

These seven samples enable us to compare:

1 the mud of the Lake with that of the environs.

2 the Rockanje mud with neighbouring dunesands and with the slime from the Meuse, the Waal, consequently from the Rhine and Meuse basins.

For purposes of comparison Miss FOLMER still added mould from the garden round the laboratory at Groningen and Fango of Battaglia and of an unknown find-spot.

After this series had been investigated it appeared from reasons to be mentioned below, that it was indeed necessary to extend the number. First Mr. TROUW at Rockanje kindly sent us three samples taken from the same spots where Mr. BÜCHNER collected the samples in 1913, which he found so extremely radio-active.

These samples are called:

I (about 50 m. to the South-West of the Grotto at a depth of 150 cm.)

II (from the South-East Corner of Betjenskelder 60 cm. deep)

III (ibidem 150 cm. deep).

In conclusion we were obliged to collect yet another series of samples scattered over the whole area and at various depths. To that end I have chosen the spots so as to include entirely the area of the spots from which samples had been taken already, which were said to have shown radio-activity by means of radiograms or emanation (cf. fig. I). They were collected 1 from 10 spots at and about those places where the radio-active samples were collected in 1913 (see above) and besides from 5 spots from the bottom of the ditch that skirts the area of the Lake and Betjenskelder on the North. From these 15 spots 46 samples were taken at different depths varying from 30, 60, 90, 120, 150, and 180 cm. All this is sufficiently indicated in the figure together with the numbers of the samples. We have taken these samples by means of a boring machine 2 m. long, consisting of a tube of galvanized iron. Its stock can be pushed through these layers of clay and peat. To the bottom is attached a box in the shape of a hollow cylinder, on the flattened side of which is a slide, which can be opened by means of a thin rod, running upwards through the iron tube; this slide, therefore, can be opened at the top and fastened by a screw, after the boring machine has been placed at the required depth. With the crank at the top of the tube one turns the whole thing round a couple of times, unscrews the slide-rod, closes and fastens the slide again and draws the whole upwards. In this way we made perfectly sure that we drew up only mud from the depth as wanted. These samples Miss FOLMER put away into small tins shut off by insulating tape, and took them to Groningen for immediate investigation.

Finally a couple of samples the radio-activity of which was said to have manifested itself by means of radiograms, were added to these; and yet: two other samples 1. dregs of sulphur-bacteria 2. very wet mud from the ditch that skirts the lake. All these samples were investigated by Miss FOLMER, who will describe below more fully her mode of working and the results.

§ 2. *Measurements.*

The researches, the desirability of which was demonstrated by Dr. BLAAUW in his Introduction and which in connection with the experience obtained in the course of the experiment were constantly enlarging, can principally be reduced to the two following parts:

I. To this belongs the investigation (cf. Introduction) concerning the 12 samples of boring, the 7 samples for comparison, the samples I, II, and III, the Fango of Battaglia, already long known to be radio-active and finally the sample: mould. All these have been investigated according to the "emanation method", of which further details will follow below.

II. In contradistinction with the preceding the "direct" measuring method defined more closely below, was yet applied to the second series of experiments in which the emanation obtained from the mud was not investigated, but the mud *itself* with regard to radio-activity. This second series of experiments principally extended over the 48 mud samples (cf. Introduction), all of them taken from the neighbourhood of the Betjenskelder or from the Betjenskelder itself.

A closer consideration of researches I and II may set forth the motive for these two series of experiments performed at such various times, as well as the different choice of methods of investigation.

Part I.

Description of the investigation according to the "Emanation method".

Here the first part certainly is the laborious dissolving of the various samples for which I used the method as indicated in outline by various investigators.[1]) 150 grammes were taken from each sample after having been dried by a slight heat, then sifted minutely and shaken forcibly for five minutes; then 5 grammes of this were

[1]) Cf. for this: B. B. BOLTWOOD, Phil. Mag. 1905. J. JOLY, Radioactivity and Geology, 1910. E. H. BUCHNER, Chem. Weekly 1913.

glowed carefully; after having been weighed the remainder was extracted by hydrochloric acid, which was first distilled over chemically pure salt. After this treatment it was filtered, through which a part of the solution to be made, viz.: the acid solution had been obtained. The filter was burnt, the ashes with the dried residue mixed in a platinum crucible with 5 grammes of Na_2Co_3 and 5 grammes of K_2CO_3. [1]) The amalgamation of this mixture took place in a small electric furnace heated for this purpose to about 800 degrees for at least three hours.

Subsequently this melt of molten mass was extracted by water and filtered in such a way that a perfectly pure alkaline solution was obtained. The remainder was dissolved in strong hydrochloric acid and added to the acid solution obtained first, the alkaline solution being preserved separately. If, however, not everything was dissolved completely, it was filtered once more, the residue amalgamated with only a little carbonate aud treated again as mentioned above; finally these acid and alkaline solutions were added separately to those prepared first. The solutions obtained i. e. of each sample an acid and an alkaline, were shut off air-tight in a one-litre boiling flask of Jenaglass and put away in order to await the emanation equilibrium, which is almost perfect after 50 days. Even after this time the solutions were still found in an entirely clear state and consequently the chance of occlusion for the formed emanation into a gelatinous precipitate, was absent.

As a second part of the investigation now followed the expulsion of the emanation, which was brought about with the assistance of an arrangement with which the various solutions were made to boil and from which in consequence the emanation could develop itself. This together with the escaped gases — the vapour was condensed in a cooler — were caught into a reservoir over a saturated salt solution. From this the emanation was transferred to a jar communicating with a second in order to make the absorption of gas possible, then all the tubes were rinsed again with air and at last the top of the jar shut off. The emanation which was again present here over a saturated salt solution, was then ready to be investigated electrically.

Hereupon followed the third and last phase of the investigation, viz: the measurement of the ionisation current, caused by the emanation described above. For these electric measurements as well as for those according to the "direct method", the electrometer was

[1]) Provided by KAHLBAUM "pro analyse" with "Garantieschein."

used by me the principle of which was published in 1914, [1]) while the description of the apparatus followed in a second article. [2])

The method of measuring I arranged as follows:

Before making the solution boil the electrometer was given a definite not over-sensitive state of charge and that: $a + 12$ Volt, b 0 Volt, c upwards of -4 Volt. The ionisation cylinder was charged to $+80$ Volt. [3]) Having attained this, the natural leak in the measuring cylinder was examined a couple of times. The time was so fixed as to make 10 scale-divisions move under the cross wire until the total of one hundred divisions were passed through; this happened in order to be able to include into the calculation the natural leak, when measuring the samples. After this the ionisation space of the apparatus was exhausted, making use of GAEDE's new single barrel airpump; with this it was possible to exhaust the space of themeasuring cylinder of a volume of 1 litre with the indispensable rubber tubing and manometerspace down to two mm. pressure. The emanation present in the bottle, after having been conducted through drying-tubes with $CaCl_2$, P_2O_5, and a tube with cotton wool, was transferred to the evacuated apparatus. After having awaited the equilibrium of the emanation with the radio-active products, radium *A*. *B*. *C*., so after about 3—4 hours, the measurement proper took place as follows: the time was fixed in which the image of the scale moved under the cross wire, but now, in contradistinction with the determinations just given, over 150 scale-divisions; as this measurement did not last for more than 5 minutes for a sample that is but little active, and also the preceding, viz: that of the natural leak took comparatively little time, the advantage had been obtained by repeating the measurement a couple of times, to be able to include into the average calculation a more exact final value. Herewith the determination proper had come to an end and the radio-activity of the substance could be computed by taking an experiment of the same kind with a normal solution of radium. This I obtained from a quantity of radiumbariumbromide supplied by Messrs. DE HAEN in Hannover, containing 0.0126 m.grammes of radium according to notification.

Here follows, by way of explanation, a calculation of one of the samples examined, viz: N°. I from Betjenskelder.

a. Determination of the natural leak.

[1]) Royal Acad. of Sciences. Amsterdam. Proceedings of June 1914.

[2]) Royal Acad. of Sciences. Amsterdam. Proceedings of September 1917.

[3]) Royal Acad. of Sciences. Amsterdam. Proceedings of June 1914.

Number of seconds wanted for every 10 mm. displacement:

from scale-division 750—700	from scale-division 700—650
41	40
50	42
45	57
48	49
45	52
sum total 3 min. 49 seconds.	sum total 4 minutes.

repeated once more:

750—700	700—650
38	44
40	53
48	43
44	46
41	53
sum total 3 min. 31 seconds.	sum total 3 min. 59 seconds.

b. Determination of the ionisation with the emanation N°. I in the apparatus after equilibrium has set in between this and its radio-active products.

1. time, wanted for the displacement from 750—700: 2 m. 2 sec.
,, ,, ,, ,, ,, ,, 700—650: 2 , 7 ,,
,, ,, ,, ,, ,, ,, 650—600: 2 ,, 25 ,,
sum total 6 m. 34 sec.

2. when this is repeated once more we obtain:
a displacement from 750—700: 2 m. 2 sec.
,, ,, ,, 700—650: 2 ,, 22 ,,
,, ,, ,, 650—600: 2 ,, 54 ,,
sum total 7 m. 34 sec.

The mean of 1 and 2 is . . . . 6 ,, 56 ,,

From *a* and *b* will now follow the correction of the natural leak:

This ionisation together with that of the emanation amounted to 6 min. 56 seconds, measured from scale-division 750—600; it can then be derived from *a*, that the first mentioned ionisation *alone* displaces the scale in 6 min. 56 sec. or 416 seconds from 750—658. Consequently this result has the same effect as if *first* only the natural leak acts and that in 6 min. 56 seconds a displacement from scale-division 750—658 and *then* the emanation *alone* from 658—600.

For further conclusions we then want to know the ionisation, only caused by the emanation of the normal solution and this also taken from 658—600. The latter, obtained in the same way, indicated a displacement from 750—600 in exactly 23.0 seconds.

In order to derive in how much time this normal solution will ionise from 658—600, the ratio of the average sensibility of charge of the distance 658—600 of the scale to that of 750—600 had to be still defined. For this determination I made use of a special experiment taken very accurately and which was of equal importance for the calculation of all the other samples. This consisted in examining the ionisation of a substance of which the ionisation was not too small, from scale-division 750—600, and that in such a way, that the time was continually checked which was wanted for passing through 10 scale-divisions. The values in seconds amounted from 750 to:

first determination: 21, 23, 24, 20, 22, 31, 24, 20, 24, 29, 32, 27, 31, 33, 34.

c. second determination: 31, 27, 29, 21, 25, 25, 29, 26, 30, 25, 36, 34, 34, 28, 28.

From this follows that the charge wanted to displace the needle from 658—600 amounted to 180/411 of the value, which applies to parts of the scale 750—600. Then the real ionisation time of the normal solution for 658—600 was: $180/411 \times 23$ sec. $= 10.1$ sec. Consequently the sample was $416/10.1 = 41.2$ times weaker than the other, so that every gramme of the sample contained: $\frac{1.25 \times 10^{-10}}{5 \times 41.2} = 0.61 \times 10^{-12}$ grammes of radium. In this way the values for the radio-activity of every sample of part I have been derived [1]).

[1]) The calculation might yet be executed somewhat differently and that with the aid of ionisation values of *only* 100 scale-divisions, e.g of 750—650, and this for a natural leak, emanation as well as for normal solutions, for from the data as given above follows (cf. *b*) that the substance ionises from 750—650 on an average in 4 min. 16 sec. or 256 sec., whereas the natural leak (cf. *a*) ionises over the same space in 7 min. 40 sec. of 460 sec. From which follows that the natural leak together with that of the emanation would ionise in 460 sec. $\frac{460}{256} = 1.8$ time over that part of the scale, (only suppose for the calculation of the sensibility of charge constant from 750—650), so *only* the emanation 0.8 time over that part mentioned in that time. For the final calculation we then derive from *c*, that the normal solution, that ionises in 23.0 sec. from 750—600, displaces the needle from 750—650 in $\frac{253}{411} \times 23$ sec. $= 14.1$ sec., consequently in 460 sec. $\frac{460}{14.1} = 32.6$ times ionises over the division of the scale mentioned. Finally there follows from all this that the activity of the normal solution is $\frac{32.6}{0.8} = 40.8$ times

The results obtained are communicated in the subjoined table.

TABLE 1.

Indication of the samples	Soil	Depth in Metres	Radio-activity expressed in 10^{-12} gr. rad. pr. gr.
boring 2	lightgrey mud with remains of peat	2.70	0.43
" 4	grey mud and dark clay........	4.40	0.54
" 6	grey-black clay................	6.05	0.55
" 8	blue-black clay................	7.40	0.61
" 9	black fat clay..................	8.30	0.78
" 10	black mud......................	9.30	0.66
" 11	bluish-grey clay with grains of sand	9.55	0.62
" 12	blue-black clay................	10.50	(2.16)
" 13	black clay with remains of peat.	12.90	0.64
" 17	sand with little slime..........	± 13.50	0.1
" 20	light-grey clay.............	21.20	0.75
" 23	sand	24.80	0.36
" 23			0.37
A	brown-black peaty mud.........	—	0.39
B	clay..........................	—	0.78
C	sand of the dunes	—	0.23
D	light-grey clay.................	—	0.59
M	slime of the Meuse	—	1.09
W	slime of the Waal	—	1.49
R	brown organic mud............	0.40	0.24
mould	....	—	0.96
No. I	blue-black clay................	1.50	0.61
No. II	brown-grey clay...............	0.60	0.49
No. III	blue-black clay................	1.50	0.57
	Fango of Battaglia.............	—	34.0

stronger than the substance investigated. The first calculation is preferable as in it the ionisations could be examined over a larger number i.e. over 150 scale-divisions. Even then the second calculation could be applied, if also the natural leak had been examined down to 600. This, however, is difficult in connection with the length of time of observation, if one makes several and very accurate observations.

From this table it is clear amongst other things that the investigated samples of the boring, as well as those of the Betjenskelder or its neighbourhood only show a radio-activity in the order of 10^{-12} grammes of radium pro gramme; i. e. no larger quantity than is normally found in most rock [1]). The activity of mould is of the same order, whereas only the sample of comparison: Fango exhibits an effect considerably higher. Notwithstanding this small radio-activity, however, it shows in relation to the depth and the nature of the layers, a certain regular course as may appear more distinctly yet from the geological interpretation of the results by Dr. BLAAUW as mentioned below [2]). The two values for N°. 23 which agree within the limits of errors of observation, relate to the values of activity of two solutions, made and investigated at quite different periods. Having arrived at the end of these experiments we could not but conclude that our results did indeed contrast greatly with the values for samples of mud from the Lake as indicated by Dr. BÜCHNER in 1913; he even found a value of 462×10^{-12} grammes of radium pro gramme in the blue mud from the Betjenskelder.

At the very moment that these researches had proceeded thus far, a circumstance intervened, which especially has been the occasion of our resolving not yet to consider the experiments as being finished; on the contrary: to continue them in a yet more extensive way. From Utrecht Prof. ZWAARDEMAKER sent us two samples of Rockanje mud; from one of these, which Prof. ZWAARDEMAKER had received in 1913 from Mr. TROUW at Rockanje, the radio-activity had been clearly shown in the Utrecht Laboratory, because they had succeeded there in obtaining radiograms through this sample. After all that preceded it was certainly remarkable that, according to the "direct method" of investigation (Cf. Part II) I could determine that this sample certainly possessed radio-activity $1^1/_2$ times that of Fango of Battaglia. In order to find out whether we might have been mistaken with other samples, Dr. BLAAUW investigated this sample with regard

1) In the table the values for activity only apply to those of acid solution; in the alkaline solutions, even in the Fango I could find no activity. An experiment with only chemical substances was made to check this with a solution which contained the quantity of chemical substances only necessary for each experiment; also this had a negative result.

2) It is of importance to remark here, that very little information has been given up to now on the relation of the quantity of radium to the depth under the surface of the earth. However, experiments showed no relation. Cf A. F. EVE and D. MC. INTOSCH, Trans. Roy. Soc. Canada 1910; E. H. BÜCHNER, Jahrbuch Rad. u. El. 10, 1913; H. E. WATSON and G. PAL, Phil. Mag. 28, 1914.

to Diatoms after which he could ascertain that it was certainly mud from Rockanje and that from a layer agreeing with samples 4 to 6 from the deep boring. Thus the Fango of Battaglia was investigated and it appeared that this is also recognisable by a definite kind of Diatoms. The Diatoms as "characteristic fossils" could again be of use here. When, therefore, the strong radio-activity of the mentioned sample had also appeared in my case, we passed on to a second enlarged investigation with the purpose of obtaining yet closer indications about the question that concerns us here. This research contains the second series of experiments arranged higher up under Part II (Cf. also Introduction).

Part II.

Description of the method of inquiry, viz.: the "direct method". This is principally to the effect that a part of the dried mud is weighed and placed into the ionisation-space; the radio-active substances, so not *only* the emanations present in it, cause ionisation of the air, which is again measured. This method, introduced already by ELSTER and GEITEL for definitions of the radio-activity of rock and soils and at present still used for these, among others by Prof. GOCKEL in Freiburg, has, however, been disputed on various grounds[1]) and it unquestionably has considerable drawbacks as a measuring method; among others this, that when applied as above mentioned it is not possible to obtain an absolute definition or mutual comparison of the quantity of radium, nay a mutual comparison of the activity as a whole cannot even strictly be carried out. For the ionisation may be the consequence of radium and its radio-active products, but at the same time of thorium and other active substances, the rays of which ionise in a very unequal degree. Then, the absorption of the rays is also disturbing, because it gives differences, for the various substances as well as for the divers radiations. Overagainst this we may, however, mention as a very important advantage of the "direct method", that we can get on with it so much quicker; no protracted chemical operations are required as in the case of the emanation method; with the electric measurement we need not await the formation of the equilibrium of the substance employed with the radio-active products to be formed by it. Thus the measurement will only take Prof. GOCKEL an hour, without mentioning the preceding, necessary determination of the natural ionisation. With the use of a sensitive electrometer, however, this advantage is shown more favourably yet. Thus it was possible for me to examine 40 samples

[1]) Cf. E. H. BUCHNER. Jahrbuch Rad. u. El. 1913.

quite accurately in 5 hours, among which occur several measurements which were repeated sometimes for the sake of a greater certainty [1]). Nor was it necessary for me to glow the substance before measuring its ionisation. Prof. GOCKEL, namely, experienced the disadvantage that during the hour of the measurement emanation escaped, the radio-active products of which settled on the sides of the vessel, thus causing the ionisation to grow stronger during the measurement; as several circumstances, a. o. the nature of the substance, are of great influence on this so-called "emanating", GOCKEL tries to avoid this difficulty by glowing the substance beforehand, or: to deprive it of all emanation present. This is an uncertain procedure, which BÜCHNER has already pointed out (cf. note p. 725) and I think it is also preferable not to shut out its activity, if the substance should contain some emanation. This is possible if the measurement is but short, as in that time the quantity of escaped emanation need not be taken into account. In my opinion both methods have great value and the importance of the "direct method" should not be underrated; first because of the advantages mentioned in order to determine the order of radio-activity, as in the case of the samples of Part II, by a speedy inquiry, but secondly to penetrate deeper into the radio-active phenomena of the substances itself. It will never be possible to say beforehand what radio-active products, perhaps new ones, one may happen to meet, or whether, as Prof. GOCKEL also remarks, all the products are indeed present, which arise from a series of active substances the emanation of which is found according to the emanation method. With the help of absorption experiments and especially in very accurate measurements it seems to me that especially in the future this method may become of very great value.

In order to complete the above brief description of the method, I may add the following: first of all the placing of the samples into the ionisation space was done in a particular way. Prof. GOCKEL describes the difficulty occurring in his case, viz: that in the space of experimentation a fresh supply of air will always penetrate, which in itself will modify the ionisation. In order to avoid this disturbance the electrometer employed by me was provided with a special

---

1) It is important to remark here that a sensitive measuring method, "direct" as well as indirect, possesses advantages over an insensitive one; where up to now, many investigators use the latter, this will explain, though only partly, the remarkable fact that as regards the measuring results of various observers one meets with so much contradiction, where determinations of the radio-activity of similar material are concerned.

closure.[1]) When in one of the basins of diameter $11^1/_2$ cm, 50 grammes of the dried substance were strewn equably, it was possible in a very simple way to exchange the basins without allowing the air to enter the ionisation space. As to the electric measurement, for this the ionisation time was only determined for 50 scale divisions over the same part of the scale. This was subsequently done with all the samples, the results of which are given in the table below; only the ionisation time is mentioned in it as, because of the above-mentioned reasons, a proper calculation of activity will not be possible. Yet the various ionisation times will approximately denote the different values of activity. For a closer meaning of the numbers cf. Fig. 1.

In order to be able to form a somewhat more accurate notion of the order of radio-activity with which such ionisations agree, I have, while supposing among others that the absorptions of the various samples are the same and, besides, that they only contain radium, traced the calculation of activity for sample I (cf. table 2) namely by comparing the ionisation time of I with that of the Fango of Battaglia. It should be taken into consideration here, that the natural leak will also give its share to attain these effects, so that in reality the ratio of the activity of the Rockanje mud to that of the Fango is still less than one might think one could derive directly from these figures. Thus I found N°. 1 to contain 1.1×10^{-12} grammes of radium pro gramme; i.e. 1.8 times a larger value for this sample than follows for this sample from the emanation method (cf. Table I). That these values do not agree, might partly be caused by the fact that the sample in question contains at the same time thorium. Many minerals are even exempt from radium, whereas they contain a large amount of thorium.

So also this second comprehensive series of experiments had not disclosed an appreciable radio-activity for any of the samples. This notwithstanding, we did not yet consider our experiments as having come to an end and that in connection with the assertion uttered on various sides that radiograms had been obtained by means of Rockanje mud; this had been the matter with the already mentioned sample 1913 and with two samples forwarded by Dr. REYS from the Hague. These samples were investigated, both electrically and photographically; it was already mentioned that according to the first

[1]) Cf. also for the drawing: These Proceedings, p. 684. The closure principally consists in this, that instead of the circular bottom plate a ring is fastened to the ionisation cylinder carrying two ways over which 2 metal basins can slide, which alternately form the bottom of the ionisation space.

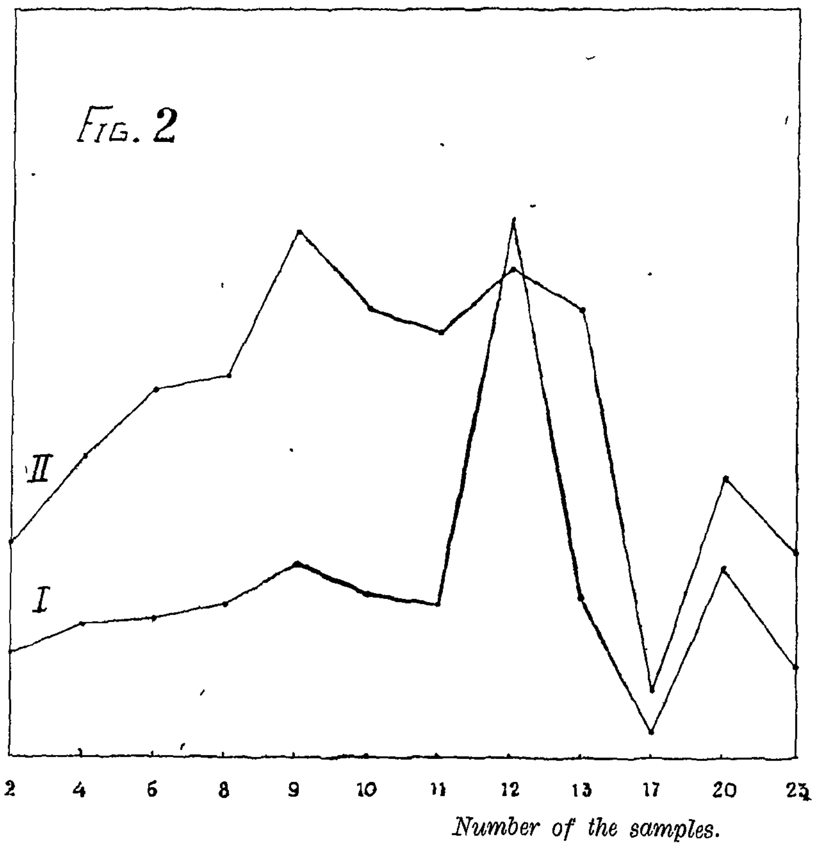
TABLE 2.

Number	Depth in cm.	Ionisation time in sec.	Number	Depth in cm.	Ionisation time in sec.
1	90	178	24	150	93
2	120	116	25	180	66
3	150	88	26	90	152
4	180	125	27	120	104
5	90	150	28	150	115
6	120	150	29	180	117
7	150	78	30	60	161
8	180	100	31	90	121
9	60	150	32	120	90
10	90	88	33	150	87
11	120	90	34	90	95
12	150	100	35	120	91
13	180	100	36	150	95
14	90	163	37	60	173
15	120	122	38	60	182
16	150	99	39	60	143
17	180	123	40	60	125
18	90	159	41	90	82
19	120	133	42	120	78
20	150	77	43	150	74
21	180	96	44	180	127
22	90	105	45	30	209
23	120	107	46	30	187
Sample I		130	Sample II		136
Sample III		120	Mould		80
Fango of Battaglia		18			

method sample 1913 showed a stronger radio-activity than the Fango. The two other samples, however, could again be ranged under the order of 10^{-12} grammes of radium pro gramme.

But also photographically these and other samples were investigated with regard to their action. Prof. H. HAGA was willing to undertake the radiogram experiments at our request, of which the results were as follows: The experiments were made in the usual way in which the photographic plate in a black envelope is exposed to the action of the substance to be investigated; in order to be able to state an eventual action some parts of the plate were protected by placing some small metal discs on the envelope, or metal pieces with figures cut into them. In using "Schleussner Röntgen-plates" only a very small action was obtained after a nineteen days' exposure from Fango. Dried mud of Rockanje N°. 12 and mould would have no action at all. In using "extra speedy Wellington" plates a very strong effect was obtained after a 30 hours' exposure with Uranium-pitchblend, a very weak one with a strip of Uranium glass and no effect at all with a quantity of Rockanje mud 1913. No more was any action obtained after 44 hours by strewing the black envelope with the whole quantity of the last mentioned samples or after a week's exposure with the above-mentioned samples from Dr. REYS. The same negative result was arrived at in experiments in which for a week Schleussner-Röntgen-plates" *without the wrapper* had been placed over a layer of Fango or mud at ± 1 mm. distance or had also been strewn with these substances.

These results agree entirely with the photographic action on radio-active substances as described in the literature on the subject. In such photographic experiments one trial experiment should always be taken with a plate from the same pack without any substance strewn upon it, and one should always be careful with the so-called black paper, some kinds of which transmit a sufficient quantity of daylight to obtain a misleading effect. As the photographic experiments had again given a negative result, there only remained to us to set forth more clearly one side of the question. It has namely been thought that there might exist a relation between organisms and accumulation of radio-activity, when BÜCHNER's definitions of the radio-active intensity of the Rockanje mud were known. As, according to this investigator, radium probably occurs in mud as $RaSO_4$ and so many sulphur-bacteria live there, whose sulphur-reserve is oxydated by respiration, so that sulphates again are dissolved into the water, it seemed desirable to us to examine bacteria-dregs and watery mud from the ditch as to their activity For this purpose Dr. BLAAUW once more took two samples and sent them to me, mixed with much water. The research tookplace both according to the "direct" as well as to the emanation method. As regards the latter, no emanation equilibrium was awaited, but only the quantity of emanation present extracted by boiling. If Fango was treated thus, very strong ionisations could again be observed. I shall not mention all the values obtained according to the various methods;



let it suffice to mention here, that also in this case the figures pointed to a radio-activity not exceeding that of 10^{-12} grammes of radium pro gramme.

Finally I will mention one very important experiment taken with the various samples of boring (cf. Part I). From the "conclusions" which Dr. BLAAUW has derived from my data one may notice how he could entirely account for the course of the radio-activity in the series of the borings. I thought it desirable also to apply the "direct method" to these samples of boring to wind up my researches. The result will be found represented graphically in curve II of Fig. 2, whereas curve I fixes the values mentioned in table I according to the emanation method. The conformity in the course will strike one: with the exception in the case of N°. 12 there occur in both curves corresponding fallings and risings. As not only the quantity of radium influences the values of the "direct method" (cf page 725) it is certainly remarkable to have to gather from these curves that either these substances contain only the active radium or that also the other active substances in it must show the same course as that element.

Besides, this conformity in the results of the two methods may be looked upon as a valuable check of the one upon the other.

But to what cause do the different values for sample 12 point? One could notice already that this sample is much more active than any of the others in series I, but again looking up the notes taken when I investigated these various samples electrically, I found that it was just for this sample that a very strange course was mentioned of its behaviour with regard to the radio-activity during the day of its measurement. Though, however, N°. 12 only contains 2.16×10^{-12} gr. of radium pro gramme yet the above mentioned fact considered by itself will be the motive that this sample will be subject to further investigation.

If then, having come to the end of all the mentioned experiments, we recapitulate results, we will see that neither the electric research, according to various methods, nor the photographic research have disclosed any appreciable values for the radio-activity of the Rockanje mud. The only exception to the large quantity of samples is sample 1913, but, as it did not come to us straight from Rockanje, but was kept in the Utrecht-laboratory for 4 years, we need not take it into serious account.

Our final conclusions from these researches can only be that in our opinion the Lake of Rockanje possesses no radio-activity of any importance.

§ 3. *Conclusions.*

After the detailed researches described above by Miss FOLMER there is no occasion left to me to seek geological explanation for an especially strong, local radio-activity, for not one of the samples shows an action which would be ± 100 times stronger than those of igneous rocks (Cf. BÜCHNER 1913, ESCHER 1915) and all the samples have an activity as has generally been found in soils of the kind.

But though the occasion for further inquiry into an hypothesis for the explanation of a strong local activity may be disregarded by me, I yet want to point out that it is now that this question becomes generally of importance: where do the radio-active parts come from which are found in the alluvium (or in a more general sense in the ground)? In this I will not venture too far on a territory with which others are so much more familiar and better entrusted and I will only point out some conclusions to be drawn from the values found by Miss FOLMER. This is all the more important, because we are concerned here for the first time with a great number of determinations of radio-activity of a geologically amply described country.

As has been fully described in the treatise (pp 29—31) we are concerned here with clay, organic material, and sand. In the microscopic investigation into the organisms of the samples I have always mentioned the greater or smaller presence of mineral dust, organic remains and grains of sand, together with their size. Of course, it may not be lost sight of that mineral dust need not exclusively be slime from the rivers, but that also part of it may come from the dunes. And the more we meet with grains of quartz in the layer, the greater the chance will be that also part of the finer mineral dust originates in the dunes or the bottom of the sea. Reversely, very small grains of quartz are thrown down in the slime, for in the freshly settled slime of the Meuse and the Waal they are to be found in a small quantity; I shall leave undecided whether they have been blown into it, carried along or rooted up. Yet it is very well possible to conclude from all the figures Miss FOLMER has given, to which of the three mentioned elements: clay, organic remains, or sea-and dunesand the radio-active parts belong.

The quantity of radio-active substance varies in the 24 samples: boring 2, 4 etc. A. B. C. D. M. W. R. I. II. III in 14 of the 24 cases between rather narrow limits: 0.49 and 0.78×10^{-12} gr. of radium pro gramme dried substance.

The following samples possess a lower number:

Boring 2 with 0.43×10^{-12} grammes. Besides fine mineral dust and a few grains of sand of 200—300 μ length at most, especially *much organic* material.

Boring 17 with only 0.1×10^{-12} grammes. Consists of blue-grey *sand* (sand with little slime).

Boring 23 with 0.36 and 0.37×10^{-12} grammes of *sand;* the grains of sand in this sample are at most 450 μ long. Mixed with very fine mineral dust.

A with 0.39×10^{-12} grammes. According to notes in 1915: excessively rich in Diatoms; *a great many organic remains*, little mineral dust".

C with only 0.23×10^{-12} grammes. According to notes in 1915: "very regular *sand* without any organic mixtures.

R with only 0.24×10^{-12} grammes. This sample is the same as that described in the treatise p. 17 under No. 2: "brown mud" where no admixture with the blue-grey mud had as yet taken place; it consists *chiefly of remains from plants and animals* with a few grains of quartz of at most 150—300 μ length.

A higher number was found for the following samples:

Boring 12 with as many as 2.17×10^{-12} grammes. This is the highest amount that has been found. "Blue-black" clay; "consists of *very fine mineral dust,* mixed with very *few* grains of sand, at most 80—120 μ long; *few organic remains.* (Treatise p. 38). Neither is there any sample in this series in which the very fine mineral dust is mixed with so little sand and organic material. It was also pointed out that there was a resemblance between the Diatoms to those from the slime of the Meuse near den Briel. (Treatise p. 51). *M* with 1.09×10^{-12} grammes. *W* with 1.49×10^{-12} grammes.

I think that from this will appear that the quantity of radio-active substance is smallest in samples of soil with much sand or with much organic substance and that this quantity will be the higher as the settled slime of the river lacks these elements. This is clearest in the case of boring 12 and the only samples where the quantity also exceeds 1.00×10^{-12} grammes, are these very slimes from Meuse and Waal. If we have to choose between an origin from the dunes or slime of the river then I think I can safely conclude *that the radio-active substance in the alluvium of Rockanje comes especially from the side of the rivers and in a far less degree from the dunes.*

Apparently there is also radio-active substance in the sand of the dunes (cf. sample C), though in a far less degree. Whether this is the same substance as in the stronger active clay will perhaps be settled later on by others. Moreover it remains possible that also that slight quantity in the dunes yet exists of minerals originally carried along, together with slime of the river and which have settled on the bottom of the sea on the shore. I must still add that, of course independent of the quantity of sand and organic remains, the slime settlements may possess different radio-active intensity, even if they come from the same river. The motion of the water, either by the current or the whirl of an eddy, may cause by fractionated settlement on various spots a varying quantity of active minerals, altering with the strength of that motion.

We will now consider the numbers of the samples of boring (2, 4, 6, 8, 9, 10, 11, 12, 13) a little more closely. They belong to that part where in a deep basin hollowed in the sand, slime of the river was thrown down in salt and later on in brackish water, while in later times the slime of the uppermost layers was mixed with ever increasing organic remains. We now see that the radio-activity increases chiefly from the top to the bottom, i.e. with the increase of slime of the river and with the decrease of organic material; boring 2 with 0.43, boring 4 with 0,54 boring 6 with 0.55, boring 8 with 0.61, boring 9 with 0.78, boring 12 with 2.16×10^{-12}! But why that slight fall in borings 10 and 11? It is exactly in these two samples that there is a larger quantity of sand. Sample 9 still consists of very black, fat clay with much fine mineral dust, the *few grains of sand* being 100—150 μ long at most. Sample 10: watery mud of very fine mineral dust (slime), mixed with *more grains of sand*, somewhat coarser, at most 350 μ long, Sample 11: Bluish dark-grey, because the slime is mixed with a few *more grains of sand*, very much particularly fine mineral dust, grains of sand at most 300 μ long; small remains of *peat* and wood. (Treatise p. 38). While in going from 9—12 one might expect a further rise of radio-activity, this rise is temporarily suppressed, because layers 10 and 11 again contain more and larger grains of sand, especially 11 a few more remains of peat. Only when this has entirely passed in 12, there appears a rather high number. But in sample 13 the activity falls again to the value of 10 and 11 and concerning this was noted down at the time: fat, black clay, many *remains of peat*, very much fine mineral dust mixed with somewhat *larger grains of sand* at most 200—400 μ long (Treatise p. 39). Then again a fall, if mixed with more organic substance and larger grains of sand. So in its details it also tallies entirely with the conclusion drawn higher up, but at the same time it appears from this of how much geological value accurate sensitive determinations of radio-activity can be.

Finally we may yet draw the attention to the numbers occurring in table 2 as collected by Miss FOLMER. There the radio-activity is the stronger as the given number of seconds is smaller. We refer to the table and to Fig. 1. Where 4 or 5 samples were taken from one spot, one higher than the other, it is again striking that the smallest action is always found in the highest layers that were bored; that first the activity increases in the downward direction, is strongest at 150 cm. depth generally and often decreases again at 180 cm. The same may apply here as to the great boring, that the upper layers contain a larger quantity of organic material. The layer of clay goes down to the depth of 13 m. on the spot of boring, on the spots of these shallow gaugings to 5 m. at most, generally far less deep, so that for the rest one can hardly compare the same depths. Moreover, it will be a good thing to pay attention to numbers 38, 45, and 46. These are the most weakly active, the only ones where the value rose beyond 3 minutes and it is exactly these samples that consist principally of sand. Besides these 3 samples there are only two left (37 and 39) which also chiefly contain sand, but these too belong to the least active ones. In 37, 38, 39, 45, and 46 the layer of sand was already present at 60, 60, 60, 30, and 30 cm. below the level of the sea.

So here it is once more corroborated, that radio-active action grows less according as the sample of the soil contains more sand or organic substance. The curves in Fig. 2, when compared mutually or with the above-mentioned particularities of the soils may demonstrate clearly, that two very different methods have led to this same conclusion. That sample 12 shows a surprising difference in the two methods is very striking indeed. May be that it points out that the very high number of sample 12 in this series is yet of particular importance.