On the Equilibrium of the Radioactive Elements in the Hydrosphere. II.

By Kazuo KURODA and Yuji YOKOYAMA.

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V. Radioactivity of the Rain Water. It was discovered in 1903 by C. T. R. Wilson[1], that the fresh rain water is radioactive, and its half period is about 30 minutes. The radioactivity of the rain water was measured by many investigators since then, and it is believed that the radioactivity is due to the presence of radium B and radium C in the fresh rain water. It is supposed that radium A (positively charged) formed from the radon in the air, is adsorbed by the small particles of rain water (negatively charged). The present authors have measured the radioactivity of the rain water since the summer of 1947, at Masutomi, Yamanashi prefecture, and in Tokyo. Several c. c. of the rain water, taken very quickly, were evaporated, and the radioactivity of the residue was measured with the Lauritsen electroscope. The ionization current diminished slowly at first for 10 to 30 minutes, and after about 40 minutes it diminished quickly almost according to the exponential curves, and the half period of that stage was about 34 minutes. A. Gockel and T. Wulf[2] reported, contrarily, that it diminishes quickly at first and described that the fresh rain water contains much radium C and little radium B at first. The opposite result was obtained, however, by the experiment of the present authors. It is considered that it is improbable that the

(1) C. T. R. Wilson, *Proc. of the Cambridge Philos. Soc.*, **12**, II (1903), 85.
(2) A. Gockel and T. Wulf, *Phys. Z.* **9** (1908), 907.

amount of radium C is larger than that of radium B, if the radioactivity of the rain water is due to the adsorption of radium A, as the half period of radium C is shorter than that of radium B. The results of the measurements are shown in Table 5 a and b.

Table 5*a*. Measurements at Masutomi.

Date	June 15	June 15	Aug 17	Aug. 17	Aug. 17
Time	15.14	16.05	13.30	15.16	16.40
Time needed for sampling	60 min.	30 min.	60 min.	40 min.	20 min.
Sample taken	2.7 c.c	9 c.c	18 c.c.	18 c.c.	20 c.c.
Radium C Content ($\times 10^{-10}$ C/1)	300	340	190	150	180
Remark			thunder rain		

Table 5*b*. Measurements at Tokyo.

Date	Aug. 27	Aug. 27	Aug. 30	Sept. 2	Oct. 6	Nov. 20
Time	14.44	15.02	15.46	14.02	15.30	12.29
Time needed for sampling	15 min.	10 min.	18 min.	11 min.	10 min.	10 min.
Sample taken	24 c.c.	12 c.c.	20 c.c.	10 c.c.	2 c.c.	1 c.c.
Radium C Content ($\times 10^{-10}$C/1)	50	65	250	150	90	80
Remark	thunder rain		thunder rain	thunder rain		

The rain water shows the unexpectedly high content of the radioactive elements, compared with that of the air which is about $20–300 \times 10^{-15}$C/1, and that of ground water, which is usually less than 1×10^{-10}C/1. It is rather difficult to compare the results of the measurements of the radioactivity by many investigators in other countries, as these measurements were carried out very early, and mostly before the confirmation of the units of the concentration of the radioactive elements. A. Goçkel obtained the values of 0.64–7.5 Mache/l ($2—30 \times 10^{-10}$C/1), and they are very low compared with those obtained by the authors in this time. Moreover, the investigators in other countries reported, without exception, that the thunder rain shows the higher radioactivity than the ordinary rain water, but the present authors could not find out such difference. It must be noted that in the old measurements in other countries, considerably long time was needed to collect the rain water, and the decay of radium C in this interval is not taken into consideration. In our experiment, the sampling and the evaporation were carried out as quickly as possible, and the correction for the decay of radium C in this interval was given. The rain water at Masutomi, where many radioactive springs issue, showed the higher content of radium C than that of Tokyo.

VI. Radioactivity of the Fumarole Vapour of the Volcano Hakone. It was found by the present authors that the fumarole-vapour of

the Volcano Iōyama (Sulphur Mountain) at Hakone is also slightly radioactive. This radioactivity is considered to be due to the presence of radium A, its half period being about three minutes. The fumarole vapour was collected with a glass funnel and a long glass tube cooled by snow. The water obtained like this was evaporated up very quickly and the radioactivity of the residue was measured with the Lauritsen electroscope. The sampling must be finished very quickly, as the half period of radium A is very short. Table 6. shows the result of the experiments.

Table 6.

The radioactivity of the fumarole vapour of the Volcano Hakone due to the presence of Radium A.

The amount of water sample: 1 c. c.
The time needed for sampling: 40 seconds.

Time (Minutes)	Radioactivity (Div./Min.)
1	0.25
2	0.17
3	0.12
4	0.12
5	0.07
6	0.02
7	0.07
8	0.02
10	0.02
13	0.00

The radium A content of the water of the fumarole vapour was calculated from the data of above-mentioned experiment, and the following value was obtained.
The radium A content $= 230 \times 10^{-10}$ Curie/1.

When the longer time was needed for sampling, we could not observe the radioactivity of radium A, and the very weak radioactivity due to radium C was detected. The radium C content was estimated to be as follows.

Table 7.

	The amount of sample	Time needed for sampling	Radioactivity: Time (min.)	Radioactivity: Ionization current (Div./min)
No. 1	10 c c.	2 hours	22	0.02
			42	0.01
No. 2	5.6 c.c.	5 minutes	17	0.06
			27	0.07

The radium C content

No. 1	1.5×10^{-10} Curie/1.
No. 2	9.5×10^{-10} Curie/1.

The radon content of the gas ejected frem the fumaroles was also measured and the results are shown in Table 8.

Table 8. The radon content of the fumarole gas of the Volcano Ioyama, Hakone.

	Radon content (94°C, 660 mm)		Radon content (0°C, 760 mm)	
No. 1	5.5 Mache	20×10^{-10} C/1	8.6 Mache	31×10^{-10} C/1
No. 2	6 7 ,,	24×10^{-10} C/1	10 ,,	37×10^{-10} C/1

The experimental data obtained above are very satisfactorily explained assuming that the radium A is adsorbed completely by the fumarole vapour. If we assume that the radium A, formed from radon, is adsorbed completely by the water particles, and the contacting time of fumarole gas and vapour is t seconds, the following relation will be obtained-:

$$[Ra\ A] = [Rn] \times \lambda_{RaA} \times t \times \frac{1000}{w} \qquad (1)$$

w: The amount of water (g) obtained from 1 litre of the fumarole gas.
$[Ra\ A]$: The concentration of RaA in water. Curie/1.
$[Rn]$: The concentration of Rn in gas. Curie/1.
λ_{RaA}: Disintegration constant of RaA.

If we assume that $[Rn] = 30\times10^{-10}$, $t = 0.5$ seconds, and $w = 0.2$ g., the following value will be obtained for RaA. This is almost equal to the observed velue. (Table 9).

Table 9.

Ra A	
Calculated	Observed
300×10^{-10} Curie/1	230×10^{-10} Curie/1

The 230 $\times10^{-10}$ Curie per litre of radium A will disintegrate and about 10×10^{-10} Curie per litre of radium C will be formed in about thirty minutes. This value is almost equal to the observed value of radium C shown in Table 7.

VII. Geochemistry of Polonium in the Hydrosphere. It was found recently by the present authors that some mineral springs in Masutomi contain considerable amounts of polonium (radium F), and the polonium content of a number of mineral springs was estimated. Polonium was separated from the mineral water as polonium sulphide, a small amount of lead compound being added as the carrier. Radium A, radium B,

thorium B etc. also precipitated, but their radioactivlty disappeared in several days, as their half periods are short, and the radioactivity due to polonium remained. Its half period was found to be about 140 days. The radioactivity was compared with that of polonium samples extracted from known amounts of pitchblende, and the polonium content of the mineral springs was calculated. The polonium content of mineral springs of Masutomi is shown in Table 10.

Table 10.

Name	Polonium content (10^{-10} C/L)
A49	3.9
A9	0.20
B4	0.10
B9	0.05
A1	less than 0.02
A3	,,
A4′	,,
B7	,,

Polonium was detected without exception in the mineral springs containing large amounts of radon. The polonium content of spring A 49 seems to be the highest in the world. It is very interesting that polonium in mineral springs shows some peculiar reactions. It was found, for example, that polonium is not precipitated by ammonia from the mineral water, although the polonium samples, which was obtained by precipitating as sulphide, are precipitated as hydroxide by ammonia or sodium hydroxide solution. These chemical reactions of polonium in mineral springs are now being studied in detail and the results will be reported later.

VIII. Radium A, Radium B, Radium C, and Thorium B Contents of Mineral Spring of Masutomi. As is already described in section II, the radium A,radium B and radium C contents of fresh mineral waters are considerably low, compared with the values expected from their radon contents.

The thorium B content of a number of mineral springs of Masutomi was also estimated recently. The results of these measurements are shown in Table 11.

Table 11.

Name	RaA	RaB	RaC	ThB
Unit		10^{-10} C/l		10^{-12} C/l
Date		June 1947		Feb. 1948
A1				92
A3		6.5	3.5	115
A4′		55	10.5	80
A6		80	10	80
A8	4400	800	100	
A49	4500	140	3	20
B7				280

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(1) The equilibrium relationships of the radioactive elements in the hydrosphere were studied.

(2) The radioactivity of the rain water and the fumarole vapour was measured.

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Chemical Institute, Faculty of Science,
Tokyo University.