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

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Introduction. This is the first of a projected series of studies on the equilibrium of the radioactive elements in the hydrosphere. It is well known that many natural waters, particularly hot springs and mineral springs, contain quite an appreciable amounts of radium and radon (radium emanation), and the equilibrium relationships between these elements were discussed by many investigators(1). However, other radioactive elements are very seldom determined in the analyses of natural waters and hence very little is known about their distribution in natural waters. As a preliminary step in the thorough investigation on the occurrence of radioactive elements in the hydrosphere, the methods of determining radon were studied by the present authors and a satisfactory procedure was developed. Instead of I. M. fontactoscope, which is widely used in this country, the present authors also used a Lauritsen-type K. Y. fontactoscope newly devised and constructed in our laboratory. Radon is determined by this new apparatus very quickly and accurately.

.As a first contribution, the occurrence of radioactive elements in the mineral waters, the rain water, and the fumarole vapour of the Volcano

Hakone was studied and the results obtained are here presented. The radioactive elements already determined by the present authors are as follows. (Table 1);

Table 1.

Name	Symbol	Isotope	Half Period	Radiation
Radium	Ra	Ra	1590 yrs.	α
Radon (Radium emanation)	Rn	Rn	3.825 days	α
Radium A	Ra A	Po	3.05 min.	α
Radium B	Ra B	Pb	26.8 ,,	$\beta(\gamma)$
Radium C	Ra C	Bi	19.7 ,,	99.97%β and α
Radium F (Pelonium)	Ra F (Po)	Po	140 days	α
Radiothorium	RdTh	Th	1.90 yrs.	α
Thorium X	ThX	Ra	3.64 days	α
Thoron (Thorium emanation)	Tn	Rn	54.5 sec.	α
Thorium A	ThA	Po	0.14 ,,	α
Thorium B	ThB	Pb	10.6 hrs.	β and γ

These radioactive elements are present in rocks and minerals in the equilibrium amounts to uranium and thorium, excepting some secondary minerals newly formed, and hence it is meaningless to determine their amounts in rocks and minerals separately. On the other hand, these elements are present in the hydrosphere in quite irregular proportions and they are present in the equilibrium proportions only exceptionally. It is considered, therefore, that we can solve some geochemically interesting problems by the study of these "disturbances" of the equilibrium of the radioactive elements in the hydrosphere.

I. Apparatus. For the determination of radon, a Lauritsen-type K. Y. fontactoscope was used. (Fig 1a). This apparatus was planned by the authors. The radon container (Fig. 1b) is made of brass and

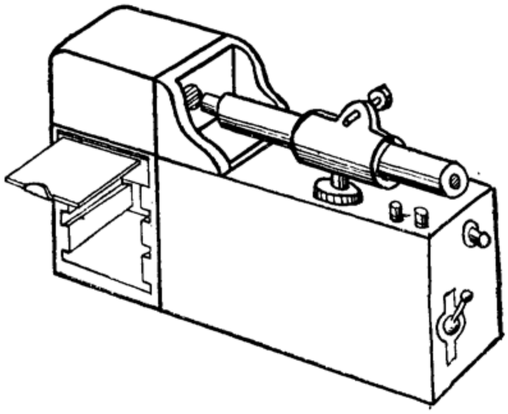

Fig. 1*a*. Lauritsen-type K.Y. Fontactoscope.

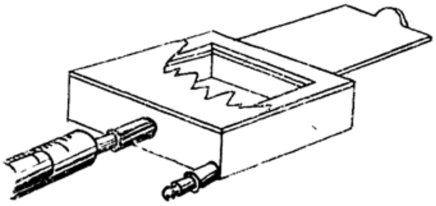

Fig. 1*b*. Radon Container.

its size is 50 mm × 50 mm × 10 mm. The water samples (0.5-several c. c.) are taken by the injectors, shaken in the radon container, and the intensity of the α ray is measured through 7 μ aluminium foils of radon container and of ionization chamber. The accuracy of the measurements was compared with that of I. M. fontactoscope and the satisfactory agreements were obtained. For the determination of other elements, such as radium, thorium, thorium X, polonium, radium A, B, C etc., these elements were co-precipitated with the suitable carriers, and the radioactivity of the powdered samples was measured by the Lauritsen-electroscope under the suitable conditions, as is described in the following section.

II. Equilibrium between Radon and its Decay Products (Radium A, Radium B and Radium C) in the Hydrosphere. The authors have intended the rapid determination of radon in natural waters by the measurement of the intensity of β ray from the decay products of radon, and found that the decay products are contained in most radioactive springs only in very small proportions, when the springs issued; Their amounts increase after the spring water issued, and the equilibrium values are obtained after about three hours. Why the decay products of radon are almost absent in fresh spring waters? The reason for this is considered to be as follows: The mineral water dissolved the radon gas at the "source of radon" near the surface of the earth, and it may be supposed that the decay products of radon were not dissolved in the mineral water at the time. As this "source of radon" is considered to be located at the places not so deep in the earth, it may be supposed that the sufficient time did not elapsed to establish the equilibrium between radon and its decay products. According to this hypothesis, the time (T) elapsed since the radon dissolved in the mineral water at the "source of radon" until the mineral water issued was calculated and the following results were obtained. (Table 2).

Table 2.

Name of Springs	Radon Content (Mache)	T (min.)
A 3	2.7	45
A 4	64	15
A 6	154	10
A 8	1580	10
A49	3700	2

These results are not contradictory to the geological observations, and to the geochemical observations on the variations of the radon content of these mineral springs after the rain or snow, as it is already reported in the previous paper⁽¹⁾.

III. Ratio of Thoron to Radon in Natural Waters. As the half period of thoron is very short, its accurate determination is rather difficult and hence very little is known about the distribution of thoron in natural waters. The present authors have measured the thoron content of a number of radioactive springs, and found that the spring B7, in Masutomi, Yamanashi Prefecture, contains appreciable amounts of thoron, although its radon content is low. (Fig. 2.) It seems that the larger the velocity and the amount of flow of mineral springs, the larger is the ratio of thoron content to radon content.

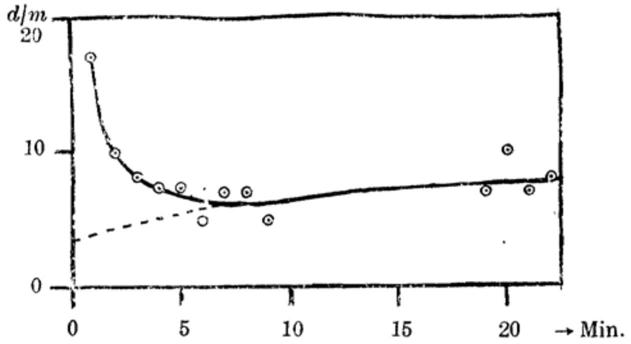

Fig. 2.
Decay Curve of Tn and Rn of the Spring B7-Gas.

A 20–40 c.c. portion of sample water was taken with the injector quickly, and the radioactivity was measured with the I. M. fontactoscope. The thoron content of a number of mineral springs and gas of Masutomi is shown in Table 3.

Table 3.

Name of Springs	Thoron Content (Mache)	Radon Content (Mache)
B7	95 82 85 67 84	6.27
A3	15	2.2
A3′	16	5.9
A3″	10	——
A1	10	——
A49	absent	about 5000
A2	,,	2–4
A6	,,	about 100

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(1) K. Kuroda, this Bulletin, **19** (1944), 33.

Table 3.—(*Continued*).

Name of Springs	Thoron Content (Mache)	Radon Content (Mache)
B7 Gas	107 96 103 94	19.5
A1 Gas	10	——
A49 ,,	absent	——
B8 ,,	,,	——

IV. Isotopes of Radium in Natural waters. The authors have studied the rapid method of the determination of radium in natural waters, and found that the mineral waters of Masutomi contain also thorium X. Radium and thorium X were coprecipitated together with barium sulphate from 1 litre of the mineral water, and the radioactivity of the powdered precipitate was measured with the Lauritsen electroscope. The radioactivity diminishes slowly for several days, according to the presence of thorium X. Its half period is about three days. The radioactivity due to radium is measured after the radioactivity due to thorium X disappeared. The amount of radium and thorium X is calculated from the curves of the change of radioactivity as is shown in Fig. 3. The thorium X and radium content of mineral springs of Masutomi is shown in Table 4.

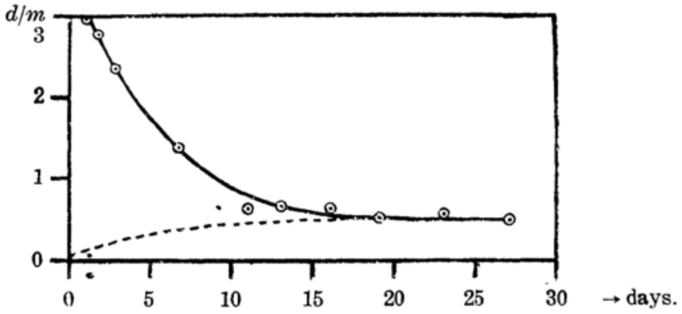

Fig. 3.
Decay Curve of Th X and Ra of the Spring B7.

Table 4.

Name of Springs	Radium Content (10^{-12} g/l)		Thorium X Content (10^{-12} C/l)	
	Aug. 1947	Feb. 1948	Aug. 1947	Feb. 1948
A1	28	23	111	104
A2	37		191	
A3	32	26	149	128
A4′	25	23	74	65
A5	76	52	184	160
A6	23	11	64	29
A7				
A8	14		10	
A9				
A49	8	6	12	2
A50	17		18	
A51	13		68	

Table 4.—(*Continued*)

Name of Springs	Radium Content (10^{-12} g/1)		Thorium X Content (10^{-12} C/1)	
	Aug. 1947	Feb 1948	Aug. 1947	Feb. 1948
B1	9		4	
B2′	less than 0.5		less than 0.5	
B4	3		,,	
B5	19		83	
B6	7		less than 0.5	
B7	31	27	320	340
B8				
B9	9		12	
C1	6		12	
C2	8		3	
C3	less than 0.5		3	
C4	2		4	
Dα	2		less than 0.5	
Dβ	less than 0.5		,,	
Dγ	3		,,	
E1	62		72	

The spring B7 showed the highest thorium X content. It is the highest value ever reported in the world. In the mineral springs of Masutomi, the following tendency was noted, namely: ··· "the higher the water temperature and the larger the amount of flow, the larger is the ratio of thorium X to radium".

This fact is satisfactorily understood, assuming that the source of radium and thorium X is located at a considerable depth in the earth. (As already mentioned, the source of radon is considered to be located quite near the earth's surface). It may be supposed that the considerable amount of thorium X decays when the spring water flows from the source of radium and thorium X to the earth's surface, as the half period of thorium X is short (3.64 days), whereas radium does not decay so much, as its half period is considerably long (1590 years). The determination of actinium X was also tried, and the results will be reported later in detail.

Summary. The equilibrium relationships of the radioactive elements in the hydrosphere was studied. The radium, radon, radium A, radium B, radium C, radium F (polonium), thorium X, thoron, and thorium B content of mineral springs of Masutomi was estimated, and discussed from geochemical point of view. It was found that spring B7 shows the high content of thorium X and thoron. Spring A49, on the other hand, showed the high content of radon and polonium. It was also found that the decay products of radon are contained in fresh mineral springs in

considerably lower amounts compared with those expected from the radon contents.

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