On the Equilibrium of the Radioactive Elements in the Hydrosphere. III. On the Ratio of Thorium Emanation to Radium Emanation in the Hydrosphere.

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Introduction.

In the previous paper[1], the occurrence of thorium emanation in the mineral waters of Masutomi was reported by the present authors.

The ratio of thorium emanation to radium emanation in the spring B7 expressed in Mache unit was unexpectedly high and showed the values larger than 10. The authors tried to verify this quite unexpected fact by different methods. The radioactivity of the mineral waters of Masutomi and other famous radioactive springs of Japan was measured and it was found that thorium emanation occurs in many natural waters. The occurrence of thorium emanation was detected and its ratio to radium emanation was determined by the following methods, namely...(1) with the I. M. fontactoscope, (2) with the Lauritsen-type K. Y. fontactoscope, and (3) by the indirect method, in which the radioactivity of the decay products deposited on the negatively charged metals was measured.

I. The Radioactivity Measurements with the I. M. Fontactoscope.

It is not so difficult to obtain the decay curves due to thorium emanation with the I. M. fontactoscope, when the sample water contains considerably high amounts of thorium emanation. However, some technical skill is required, as the half period of thorium emanation is very short, and it disappears if we do not begin the measurements quickly.

The stagnant waters do not contain thorium emanation, and the mineral waters containing large amounts of thorium emanation issue vigorously from the orifice, in most cases directly from the gap in rocks. The samples must be taken very quickly with the injectors of the suitable size, and ejected vigorously in the ionization box of the I. M. fontactoscope. The radioactivity measurement is started as soon as possible. The examples of the decay curves are shown in Fig. 1. a. and Fig. 2. The ratio of the radioactivity of thorium emanation and radium emanation at the time when the sample was taken, is obtained by the extrapolation of the decay curves, as is shown in Fig. 3. Fig. 1. b. shows an example of decay curve of thorium X.

(1) K. Kuroda and Y. Yokoyama, this Bulletin, **21** (1948), 52, 53.

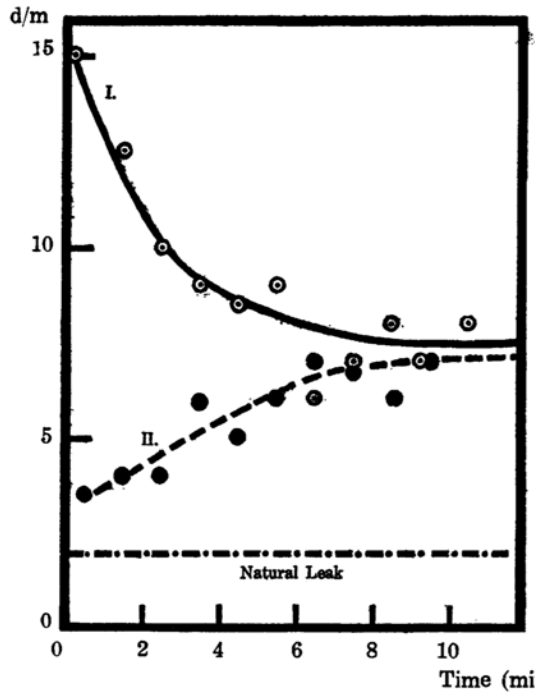

Fig. 1. (a) Decay Curve of Thorium Emanation (I. M. Fontactoscope) O-T-R, Misasa.

Date Aug. 27, 1948, 15h 13′ 30″
Sample; 100 c.c.

I. Radioactivity measurement started as soon as possible.
II. Radioactivity measurement started 5 minutes after the sample was taken with the injector. The decay curve of thorium emanation is not observed.

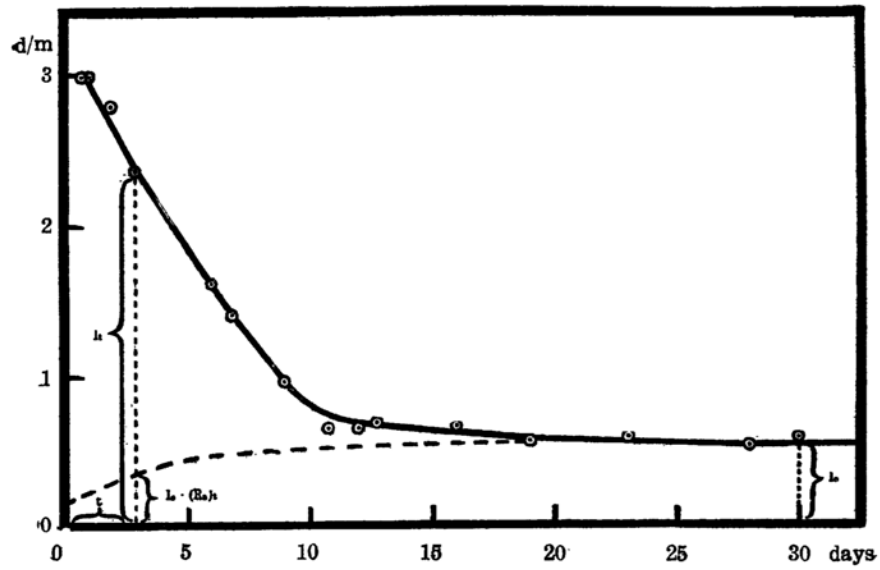

Fig. 1. (b) Determination of the ThX Content and the Ra Content. Masutomi Spring B7.

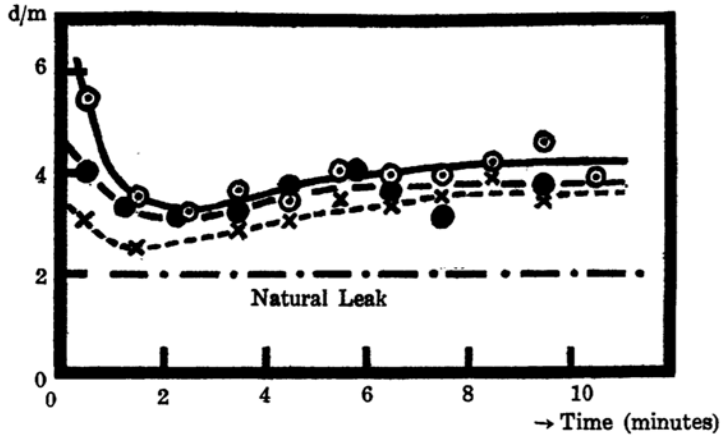

Fig. 2. Decay Curves of Thorium Emanation (I. M. Fontactoscope). O-K-1, Sekigane.

Date: Aug. 28, 1948
Sample: 50~100 c.c.

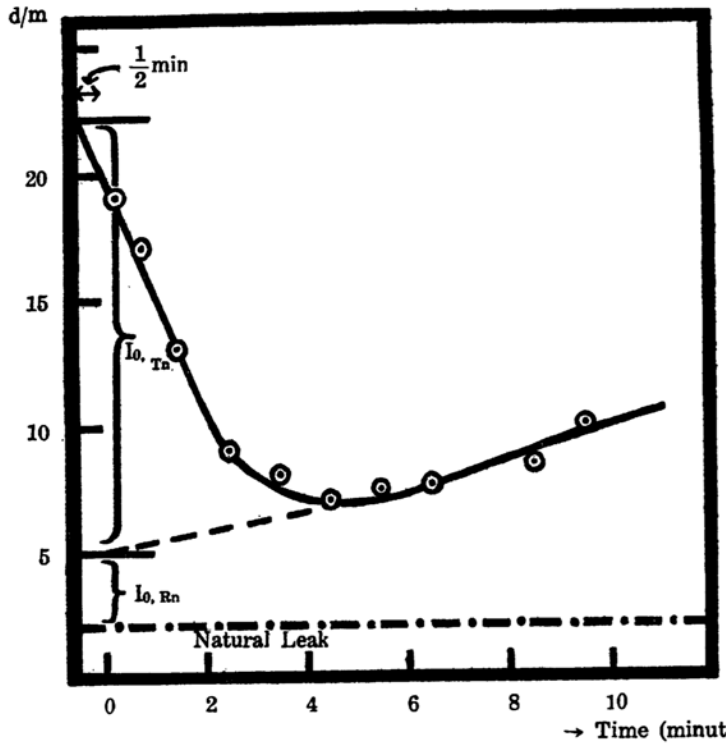

Fig. 3. Determination of the Ratio of Tn/Rn. O-T-R, Misasa. Sample 100 c.c.

II. The Radioactivity Measurements with the Lauritsen-type K. Y.-Fontactoscope.

The decay curves of thorium emanation was also obtained by the radioactivity measurement with the Lauritsen-type K. Y. fontactoscope.

It was rather difficult to determine the ratio of thorium emanation to radium emanation with the K. Y. fontactoscope, compared with the case of the I. M. fontactoscope, as it needed more time to take the sample water into the radon container and to start the radioactivity measurements. However, satisfactory agreements were obtained by these methods. The examples of the decay curves of thorium emanation are shown in Fig. 4.

III. Determination of the Ratio of Thorium Emanation to Radium Emanation by the Indirect Method.

To ascertain the results obtained by the above-mentioned methods, we tried the determination of the ratio of thorium emanation to radium emanation by the indirect method, in which the decay products of thorium emanation and radium emanation are deposited on the negatively charged wires, and the radioactivity is measured with the Lauritsen-electroscope. Fig. 5 shows the apparatus to collect the decay pro-

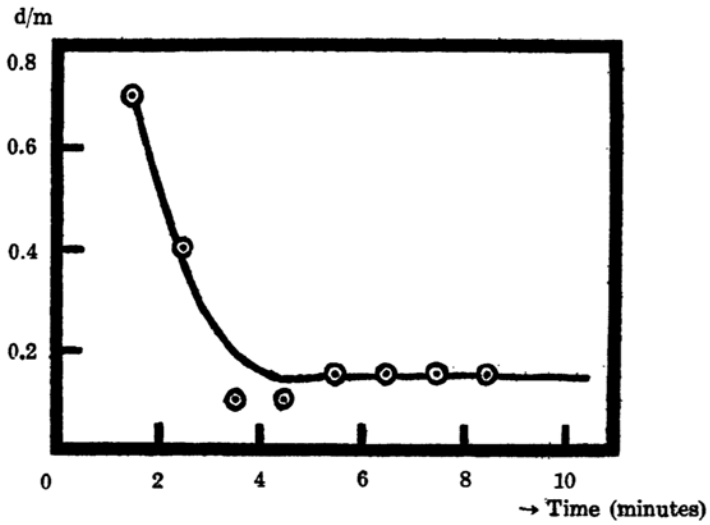

Fig. 4. Decay Curve of Thorium Emanation. (K.Y. Fontactoscope) The Spring B7 in Masutomi.

ducts on the wire. For the high voltage battery, the layer built dry batteries of Toshiba Co. were used. The voltage of single battery was 67.5 volts, and one or two batteries were used in each apparatus. This apparatus was exposed overnight at the places very close to the orifice of the springs, and then the radioactivity was measured with the Lauritsen-electroscope.

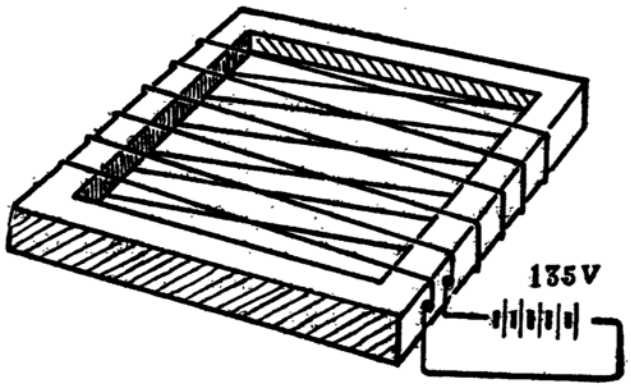

Fig. 5. Apparatus used for collecting the Decay Products of Thorium Emanation and Radium Emanation.

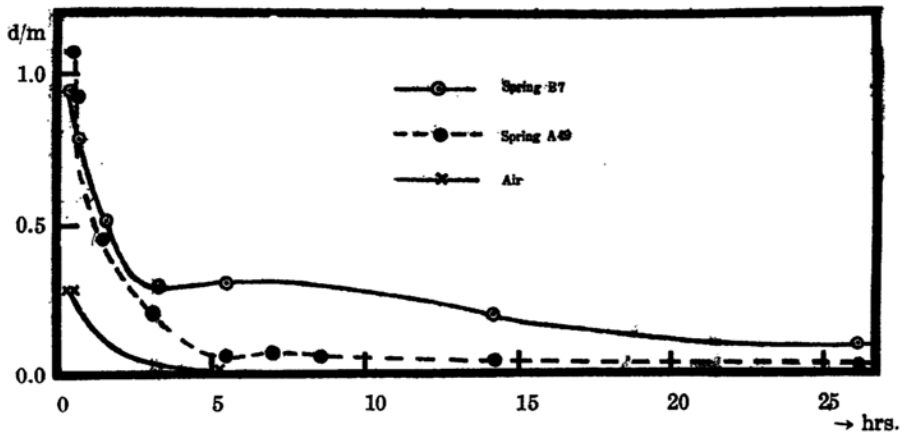

Fig. 6. Determination of the Ratio of Tn/Rn by the Indirect Method.

IV. Results of the Experiments.

The results of the determination of the thorium emanation and radium emanation contents of a number of the hot springs and mineral springs in Japan are shown in Table 1.

Table 1. The Thorium Emanation Content of the Mineral Springs and Hot Springs of Japan.

Masutomi. (Yamanashi Prefecture)

Name	Thorium Emanation Content (Mache)	Radium Emanation Content (Mache)	Apparatus
B7	95	6.27	
A3	15	2.2	
A3′	15	5.9	I.M.
A3″	10	—	
A1	10	—	
A49	absent	about 5000	
Gas from the spring B7	107	19.5	

Ikeda. (Simane Prefecture)			
No. 1	absent		K.Y.
No. 3	330	130	
Misasa. (Tottori Prefecture)			
O-T-R (Ōhasi-Ryokan)	450	80	I.M.
Matubara	8	26	
Sekigane. (Tottori Prefecture)			
O-K-1 (Onseirō-Ryokan)	100	30	I.M.
O-K-2	30	30	
O-U	30	30	
O-N-C	30	30	
Arima. (Hyogo Prefecture)			
Tansan-Onsen (Small spring)	20–40	ca. 5	I.M.
Sakae-Kōsen	10	ca. 5	

Remark: The thorium emanation content was expressed in Mache unit. It is defined as follows: it is called 1 Thoron-Mache when 0.001 e.s.u. of saturation current is obtained by thorium emanation and its equilibrium amount of thorium A in 1 litre of the sample water.

$$1\,\text{Tn Mache} = 1.52\times10^{-10}\ \text{Curie/l.}$$
$$1\,\text{g Th unit} = 1.11\times10^{-7}\ \text{Curie.}$$

In the previous paper, the authors have reported the high content of thorium X and thorium emanation in spring B7 in Masutomi. From the experiments in Masutomi, it was expected that the radioactive springs containing large amounts of thorium X, also contain large amounts of thorium emanation. The thorium X content of the hot springs of Misasa and the mineral springs of Ikeda is not so high compared with that of the mineral springs of Masutomi. Spring No. 3 in Ikeda, which contains the highest amount of thorium X among several springs in Ikeda, was found to contain thorium emanation, but we could not detect the presence of thorium emanation in other springs of Ikeda, as in most cases it was difficult to take the fresh samples from the orifice. The thorium X content of the hot springs of Misasa is much lower than those of Masutomi and Ikeda, and though eight samples containing considerably large amounts of thorium X were measured, thorium emanation was not detected excepting one sample. This was Matubara-no-yu (yu = spring), in which 2.1×10^{-5} g of thorium was found in 1 litre, by Mr. K. Shimokata. The Ōhasi-Ryokan-Soto-Yu showed the highest content of thorium X among the springs of Misasa, but thorium emanation was not detected. Therefore, we concluded that the hot springs of Misasa do not contain the high amounts of thorium emanation, when we visited there for the first time in May of 1948. However, in August of 1948, the radioactivity was

measured with the special-sized counter, and it was found that some springs in Misasa show unexpectedly high radioactivity, whereas their radon or radium contents are not unusual. For example, a spring in the hotel "Ōhasi", called "Tennen-Gankutu-no-yu-Reisen" (O-T-R), showed unexpectedly strong radioactivity when it was examined with the counter. This spring was carefully examined repeatedly with the I. M. fontactoscope, and it was found that the fresh sample of this spring shows high content of thorium emanation. The thorium emanation content of this spring is the highest ever observed in this country and probably in the world.

The hot springs of Sekigane, and the hot springs of Arima were also studied in the summer of 1948, and it was found that the springs containing not so high amounts of thorium X, sometimes contain the considerable amounts of thorium emanation. The authors consider that most radioactive springs contain thorium emanation, but it was not detected by the previous investigators, as the samples were taken from the stagnant springs and as the radioactivity measurements were started very deliberately. The distribution of thorium emanation accompanied by radium emanation in many mineral waters shows that the source of these elements is not so deep under the ground.

The determination of the ratio of thoron to radon by the indirect method was carried out in the summer of 1948, in Masutomi and in Arima. The experimental results are shown in Table 2.

Table 2. The Determination of the Ratio of Thoron to Radon by the Indirect Method.

	Spring B7		Spring A49		In the Air (At the hotel Tuganero)	
Length of the Wire	2 m.		1 m.		2 m.	
Voltage	135 volts		67.5 volts		135 volts	
Beginning of Exposure	19h 10, July 2		19h 00, July 2		19h 00, July 3	
End of Exposure	7h 04, July 3		7h 06, July 3		9h 16, July 4	
Exposed Time	11 hrs 54 m		12 hrs 06 m		14 hrs 16 m	
t	$I_{obs.}$	$I_{cal.}$	t	$I_{obs.}$	$I_{cal.}$	t	$I_{obs.}$	$I_{cal.}$
m	d/m		m	d/m		m	d/m	
26	0.94	0.90	30	1.08	1.06	22	0.27	0.28
40	0.78	0.82	40	0.92	0.94	27	0.27	0.26
1h 35	0.52	0.59	1h 26	0.46	0.45	5h 17	0.00	0.00
3h 20	0.39	0.43	3h 09	0.21	0.12			
5h 23	0.40	0.38	5h 22	0.06	0.08			
14h 14	0.19	0.21	6h 59	0.08	0.07			
27h 33	0.10	0.09	8h 37	0.07	0.06			
50h 11	0.04	0.02	14h 11	0.04	0.04			
			27h 39	0.01	0.02			
I_0ThB, C	0.49 d/m		0.10 d/m		0.00 d/m			
I_0RaB, C	0.48 ,,		1.18 ,,		0.31 ,,			
I_∞ThB, C	0.99 ,,		0.20 ,,		0.00 ,,			
Thoron/Radon (Curie/Curie)	2.06		0.17		0·00			

In Table 2. I_0 *ThB,C* (= ionization current of thorium B and thorium C at t = 0, when the exposure finished) is calculated from the values of ionization current after RaB and RaC have completely disintegrated. The values of ionization current due to ThB and ThC at each time were calculated and these values were subtracted from the observed values of ionization current (I_{OBS}). The values of ionization current due to Ra B and Ra C are obtained here, and from these values, I_0 *RaB, C* (Ionization current due to RaB and RaC at (t = 0) is obtained.

Now, the amount of thorium C formed from thorium emanation in 12 hours is 49.6 % of the equilibrium value, the values of ionization current due to thorium B, C when the activation reached the saturation value (I_∞*ThB, C*) are then calculated. The ratio of Tn/Rn is obtained from the ratio of I_∞*ThB, C*/I_0 *RaB, C*. $I_{cal.}$ is the calculated value from the values of I_0 *RaB, C* and I_0*ThB, C*. The decay curves and theoretical values showed satisfactory agreements. However, the ratio of Tn/Rn in the atmosphere above the orifice of the springs is obtained by this method, and this value will be different from the ratio of Tn/Rn in the mineral springs. The thorium emanation and radium emanation content of the spring B7 expressed in Curie unit is as follows.

$$\text{Tn} = 144 \times 10^{-10} \text{ Curie/l.}$$

$$\text{Rn} = 20 \times 10^{-10} \text{ Curie/l.}$$

Therefore, the ratio of Tn/Rn in the spring B7 is about 7 expressed in Curie unit. The ratio in the atmosphere on the orifice of the spring B7 is considerably lower than this value, as is shown in Table 2. The reason for this is probably because the half period of thorium emanation is very short and it disintegrates as soon as it issued from the orifice.

The thorium emanation content of the spring A49 was not determined by the direct methods. However it is estimated from the result of indirect determination to be as follows, if we assume that the conditions are same in both spirngs (B7 and A49).

Rn content of the spring A49 = about 17000×10^{-10} Curie/l.

Tn/Rn in the atmosphere near A49 = 0.17

As the ratio [(Tn/Rn) in the air]/[(Tn/Rn) in the water] was 2/7 in the case of the spring B7, the ratio Tn/Rn in the spring water of A49 will be about 0.6.

Therefore, the Tn content of the spring A49

= about 10000×10^{-10} Curie/l.

= about 6600 Tn-Mache.

(Remark ; 1 thoron-Mache = 1.52×10^{-10} Curie/l.)

The determination of the ratio of Tn/Rn was also tried at Arima. In the atmosphere near the orifice of Tansan-Onsen, the decay products o-

thorium emanation were detected, but their radioactivity was very weak, and the radioactivity due to the decay products of radium emanation was not detected.

Table 3. The Determination of the Ratio of Thoron to Radon by the Indirect Method. "Tansan-Onsen" at Arima, Hyōgo Prefecture.

Exposure		19h 40, Sept. 2, ...7h 40, Sept. 3			
Voltage		−135 V			
T (m)	40	1h 42 m	2h 46 m	5h 16 m	10h 0 m
I (d/m)	0.06	0.07	0.07	0.07	0.045

From this, the ratio of Tn/Rn in the atmosphere near the orifice of the "Tansan-Onsen" is considered to be about 10 or larger. Therefore, the ratio of Tn/Rn in the atmosphere at Tansan-Onsen is considerably larger than that of the spring water.

This result is quite different from the result obtained in Masutomi, and further investigation is necessary.

Summary.

The ratio of thorium emanation to radium emanation in natural waters was measured. Following three methods were used: (1) with the I. M. fontactoscope, (2) with the Lauritsen-type K. Y. fontactoscope, and (3) the indirect method, in which the radioactivity of the decay products of thorium emanation and radium emanation is measured with the Lauritsen-electroscope.

It was found that thorium emanation is considerably widely distributed in the mineral springs and hot springs of Japan.

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