Strongly Radioactive Springs Discovered in Masutomi.

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CONTENTS.

1. Preface.

A number of detailed investigations on the radioactive springs have been made by various investigators, but as most of them were made with springs having only low content of radioactive elements, the work was often imperfect with insufficient results for theoretical discussions.

The author discovered recently a number of extremely radioactive springs in Masutomi, their radon contents being one of the highest in the world. The radioactivity of these mineral springs was measured over four hundred times in the course of several months, and among the results obtained were its relations to certain meteorological conditions. Although these strongly radioactive springs newly found are now being studied in detail and it is hoped that interesting informations may be obtained as the result, the author decided to report here the results obtained thus far.

2. The Strongly Radioactive Springs Newly Discovered in Masutomi.

At the beginning of the twentieth century, three springs, namely Kinsento, Ginseito and Ensento, were known in Masutomi as admirable bathing springs.

The radioactivity of these mineral springs was measured for the first time, as far back as 1913, by Mr. Minakawa in the Hygienic Laboratory of Yamanasi Prefecture, who found it to be considerably radioactive. The results obtained by him is shown in Table 1. In 1913, R. Ishizu[(1)] visited there and measured the radon contents, the highest value obtained being 828.34 Mache. He reported the presence of 10 springs in which the radon content ranged between 100 and 500 Mache, that of one spring being higher than 500 Mache, and that of another exceeding 800 Mache. Prof. Kimura and T. Nakai,[(2-4)] who visited this region in 1936, found the radon content of Wadegawara No. 2 spring to be about 1300 Mache.* S. Oana and the

* Dr. Nakai described that the radon content of the sample taken from the surface of the spring was lower than that of the sample taken from the lower parts of the spring. His results are as follows:

Surface .... 1343 Mache Bottom .... 1133 Mache

(1) R. Ishizu, "The Mineral Springs of Japan", (1915).
(2) T. Nakai, *J. Chem. Soc. Japan*, **58**(1937), 638.
(3) T. Nakai, *ibid.*, **59**(1938), 1181.
(4) T. Nakai, this Bulletin, **15**(1940), 363.

author[5-8] measured the radon content of this spring for the first time in July 1939. The radon content of this spring was then 1090 Mache, which, later in October 1940, was found to be 1200 Mache. Since 1941, however, the radon content of this spring declined as follows:

Oct. 1941	615 Mache
May 1942	63 Mache
Dec. 1942	607 Mache (determined by Nakanisi)

The general opinion, therefore, was that the radon content of this most radioactive spring of Masutomi would never again exceed 1000 Mache.

The radon content of a number of mineral springs theretofore unknown was measured on November 2, 1942, with the result that the radon content of two springs, Spring A8 and Spring A9, was higher than 1000 Mache, that of the latter being as much as 1930 Mache.[9] The radon content of this spring was measured a number of times at the end of the same month by T. Nakai, S. Oana, M. Nakanisi, Z. Kikumura and the present author, then with the result of between 1500 and 1600 Mache. Since then the radon content of this spring has been measured daily for about one hundred days—until No. 98 day—by Z. Kikumura and the present author.

Spring A9.

The amount of the flow of this spring is too small to measure with sufficient accuracy. Rough estimates, however, make it less than 100 c.c. per minute. As it rains or snows, the flow increases in amount. When-

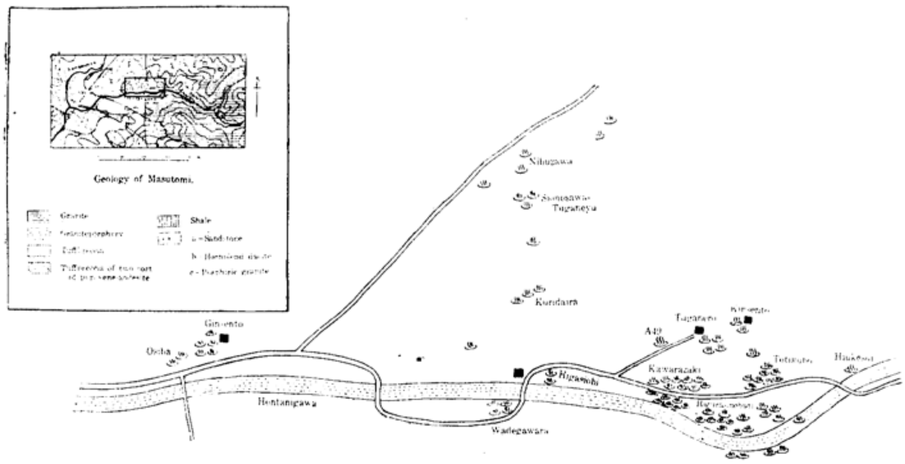

Fig. 1.

(5) S. Oana and K. Kuroda, this Bulletin, **15** (1940), 485.
(6) S. Oana and K. Kuroda, *ibid.*, **17** (1942), 397.
(7) K. Kuroda, *Ber. Japan. Ges. Balneol.*, **1** (1941), 103.
(8) G. H. Schwabe, S. Oana and K. Kuroda, Mitteilungen der Deutschen Gesellschaft für Natur- und Völkerkunde Ostasiens. Band XXXIII. Teil E. (1943).
(9) K. Kuroda and M. Nakanisi, this Bulletin, **17** (1942), 489.

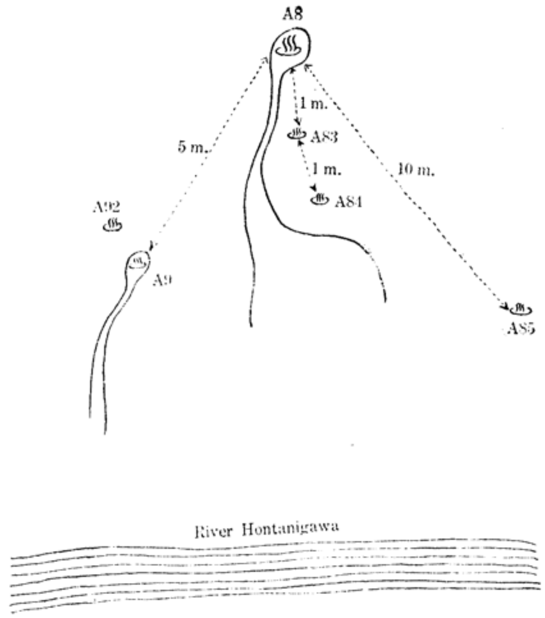

Fig. 2.

ever the amount of the flow of the springs is large, the water from spring A92, another small spring very close to A9, flows into the last named. The radon content of spring A92 being considerably less than that of spring A9, it was supposed that the content of spring A9 decreases considerably whenever the spring A92 flows into the spring A9. Fig. 1 and Fig. 2 show the location of these mineral springs.

Spring A8.

Spring A8 issues at Higurasi-no-huti near spring A9. The amount of flow is not so large. The spring showed the highest radon content (1960 Mache) on No. 85 day.

Table 1.* Radon Content of Mineral Springs of Masutomi, Determined by Mr. Minakawa in the Hygienic Laboratory of Yamanasi Prefecture. (1913)

	Spring	Radon Content (Mache)		Spring	Radon Content (Mache)
(1)	Kuridaira	260	(3)	Nibusawa	154
(2)	Sionosawa	213	(4)	Ginsento	129

* According to Vogt's "Lehrbuch der Bäder- und Klimaheilkunde", the radon content of a spring in Masutomi is 1425 Mache. (See also "The Hot Springs of Japan", (1922) Tokyo.)

Small Springs with Extremely High Radon Content.

After the snow of No. 72 and No. 76 day, the mineral springs in the Masutomi region increased considerably in the quantity of flow, and many small springs issued from places near spring A8 and spring A9. The writer, on measuring the radon contents of these waters, found that they contained very large amounts of radon.

(1) Spring A84. This is a very small spring near spring A8. The amount of flow is less than 1 c.c. per minute. A 1 c.c. sample was drawn by a small injector at 10.40 a.m. on No. 88 day, and carefully diluted with radon-free (sufficiently boiled) water. The radioactivity was measured by an I. M. fontactoscope with the following results.

Sample: 1 c.c. (diluted to 500 c.c.) Time of Shaking: 10.40 a.m.
Natural Leak: 0.9 div./min.

θ (min.)	Velocity of Foil (I_θ)	I_0	θ (min.)	Velocity of Foil (I_θ)	I_0
7	15.2	10.4	17	19.3	11.9
8	16.0	10.5	18	18.5	11.4
9	16.2	10.5	23	19.2	11.6
10	16.8	10.8	24	19.8	11.9
11	15.8	10.1	30	21.6	12.8
12	17.4	11.0			

I_0 (mean value) = 11.29 Radon content = 7130 (Mache)
K = 0.605 Mache radon per div. per min.

The radon content of this water was measured once more at 11.50 a.m., using another fontactoscope (K=0.864), when a value of 4510 Mache was obtained. Why the radon content of this spring declined so rapidly is not clear. The determination was repeated a number of times since, but such high values as 7130 Mache or 4510 Mache were never again obtained. The results of all the determinations are summarized in Table 2.

(2) Spring A9 (when the sample was drawn from the inner and upper parts). Although, when the water sample was drawn in the usual way with a pipette, the radon content of spring A9 is about 1500–2000 Mache, values exceeding 2000 Mache were obtained, when the sample was carefully drawn with an injector from the inner and upper parts of the exit of this spring. On No. 88 day, for example, the radon content of a sample drawn from the inner part of this spring was as high as 6870 Mache.

Sample: 1 c.c. (diluted 500 c.c.) Time of Shaking: (θ_0) 3.29 p.m.
Natural Leak: 0.57 div./min.

θ (min.)	Velocity of Foil (I_θ)	I_0	θ (min.)	Velocity of Foil (I_θ)	I_0
7	11.1	7.45	15	11.9	7.40
8	11.1	7.32	16	12.5	7.76
9	10.8	7.03	22	12.8	7.77
14	13.0	8.12			

I_0 (mean value) = 7.55 Radon Content = 6870 (Mache)
K = 0.864 Mache radon per div./min.

The radon content of this water was measured once more at 4.27 p.m., using another fontactoscope, when a value of 4550 Mache was obtained. The results of all the determinations made with this spring are summarized in Table 2.

(3) Spring A83. This is a very small spring near spring A8. The amount of flow is also less than 1 c.c. per minute. The radon content of this spring, which was 4330 Mache on No. 85 day, gradually diminished.

Table 2. Radon Content of Small Springs.

No.	Name	Date	Radon Content. (Mache)
(1)	A84	10.40 a.m., No. 88 day	7130
(2)	A9	3.29 p.m., No. 88 day	6870
(3)	A9	9.23 a.m., No. 89 day	5200
(4)	A9	4.27 p.m., No. 88 day	4550
(5)	A84	11.50 a.m., No. 88 day	4510
(6)	A83	5.00 p.m., No. 85 day	4330
(7)	A9	2.15 p.m., No. 90 day	3870
(8)	A83	4.40 p.m., No. 85 day	3400
(9)	A83	1.10 p.m., No. 90 day	3400
(10)	A9	3.45 p.m., No. 94 day	3040
(11)	A83	4.10 p.m., No. 93 day	2770
(12)	A9	4.40 p.m., No. 93 day	2500
(13)	A9	4.18 p.m., No. 94 day	2440
(14)	A83	3.30 p.m., No. 93 day	2410
(15)	A85	4.39 p.m., No. 87 day	2400
(16)	A9	10.12 a.m., No. 90 day	2400
(17)	A83	1.43 p.m., No. 90 day	2400
(18)	A9	11.51 a.m., No. 94 day	2360
(19)	A83	9.27 a.m., No. 88 day	2300
(20)	A9	5.30 p.m., No. 89 day	2200
(21)	A9	0.28 p.m., No. 94 day	2190
(22)	A9	0.20 p.m., No. 90 day	2140
(23)	A83	10.10 a.m., No. 94 day	2090
(24)	A92	4.00 p.m., No. 86 day	2000

(4) Spring A85. This is a very small spring issuing from a crevice in granite. The amount of flow is very small. An injector was used in sampling. The radon content of this spring, which was 2400 Mache on No. 87 day, diminished, until after some days it disappeared.

(5) Spring A92. This is a very small spring near spring A9. The amount of the flow of this spring is so small that to measure it is difficult. Rough estimates, however, make it less than 10 c.c. per minute. Following rain or snow, the flow increases in amount.* The radon content of this spring was less than 1000 Mache in 1942 (see page 36), but later it increased considerably. On No. 186 day the radon content of this spring

* In winter, when it is very cold, the amount of the flow of this spring decreases considerably.

registered 3210 Mache.** This spring disappeared, however, on No. 203 day, when the author visited there.

A Strongly Radioactive Spring Newly Discovered (Spring A49).

The writer, on measuring the radon content of a spring that was recently discovered near Hoetl Tuganero, found that it contained a very large amount of radon. The following results were obtained.

Date: No. 155 day.
Temperature: 22.0°C.
pH: 6.2–6.3
Amount of Flow: 0.6 litre per minute.
Natural Leak: 0.3 div. per min.
Sample Taken: 14 c.c. (diluted to 500 c.c.)
Time of Shaking: 12.21 p.m.

θ (Time) (min.)	Velocity of Foil Division	Velocity of Foil Time (seconds)	I_θ	I_0
$\theta_1=6$	10 – 90	29.7	162	111
$\theta_2=6$	25 – 75	18.5	162	111
$\theta_3=7$	20 – 70	17.7	169	113
$\theta_4=8$	20 – 70	17.3	173	115
$\theta_5=9$	20 – 70	17.0	177	115
$\theta_6=9$	20 – 70	16.7	180	117
$\theta_7=10$	20 – 70	16.5	182	117
$\theta_8=11$	20 – 70	16.5	182	116
$\theta_9=12$	20 – 70	16.3	184	116
$\theta_{10}=13$	20 – 70	16.0	188	118

I_0(mean value) = 115
K = 0.517 Mache per div. per min.
Radon Content = 4370 (Mache)

Repeated measurements proved that the radon content of this spring is invariably higher than 3000–4000 Mache (see Table 3 and 4). Values exceeding 5000 Mache were obtained at the time the sample was drawn,

Table 3.

(a) The radon content of spring A49 on No. 156 day. (3.49 p.m.)

N.L.: 2.7 div. per min.
Water Temperature: 21.5°C.
Sample Taken: 2 c.c.

θ (Time) (min.)	Velocity of Foil Division	Velocity of Foil Time (seconds)	I_θ	I_0
$\theta_1=7$	10 – 20	19.2	28.6	19.3
$\theta_2=7$	25 – 35	18.6	29.6	19.8

** Dr. Iwasaki told me recently that he discovered a strongly radioactive spring in Ikeda. The radon content of this spring showed about 3800 Mache only once. This spring is considered to be very similar to spring A92 in Masutomi.

Table 3.—(*Concluded*)

θ (Time) (min.)	Velocity of Foil: Division	Velocity of Foil: Time (seconds)	I_θ	I_0
$\theta_3 = 8$	40 – 50	18.1	30.5	20.2
$\theta_4 = 9$	5 – 15	17.2	32.2	20.3
$\theta_5 = 10$	19 – 29	18.1	30.5	19.6
$\theta_6 = 11$	34 – 54	37.6	29.3	18.7
$\theta_7 = 14$	10 – 60	91.2	30.2	18.9
$\theta_8 = 16$	10 – 60	91.3	30.2	18.7
$\theta_9 = 19$	10 – 60	88.5	31.2	19.1
$\theta_{10} = 21$	10 – 60	87.5	31.6	19.2
$\theta_{11} = 23$	10 – 60	88.5	31.2	18.9

I_0 (mean value) $=19.5$
Q=5200 (Mache)

(b) The radon content of spring A49 on No. 157 day. (3.50 p.m.)

N.L.: 4.0 div. per min.
Sample Taken: 11 c.c.

θ (Time) (min.)	Velocity of Foil: Division	Velocity of Foil: Time (seconds)	I_θ	I_0
$\theta_1 = 6$	10 – 60	24.1	121.9	83.5
$\theta_2 = 6$	10 – 60	23.4	125.8	86.2
$\theta_3 = 7$	10 – 60	22.7	129.8	87.0
$\theta_4 = 8$	10 – 60	22.2	132.7	87.8
$\theta_5 = 9$	10 – 60	22.3	132.2	86.2
$\theta_6 = 10$	10 – 60	22.3	132.2	85.1

I_0 (mean value) $=86.0$
Q=4170 (Mache)

(c) The radon content of spring A49 on No. 158 day. (7.13 a.m.)

N.L.: 0.5 div. per min.
Water Temperature: 22.0°C.
Sample Taken: 11.5 c.c.

θ (Time) (min.)	Velocity of Foil: Division	Velocity of Foil: Time (seconds)	I_θ	I_0
$\theta_1 = 5$	10 – 60	24.3	123.0	84.2
$\theta_2 = 6$	10 – 60	23.6	126.5	86.7
$\theta_3 = 7$	10 – 60	23.5	127.5	85.5
$\theta_4 = 8$	10 – 60	22.5	133.0	88.0
$\theta_5 = 8$	10 – 60	22.4	133.5	88.3
$\theta_6 = 10$	10 – 60	22.3	134.0	86.3
$\theta_7 = 11$	10 – 60	22.1	135.5	86.3

I_0 (mean value) $=86.5$.
Q=4000 (Mache)

Table 4. Radon Content of Dairokuten-no-izumi.

No.	Name	Date	Radon Content (Mache)	Remark (Sample)
390	Exit No. 3	12.21 p.m., No. 155 day	4370	14 c.c.
391	" " 3	2.06 p.m., No. 155 day	3240	10 c.c.
392	" " 2	4.23 p.m., No. 155 day	3460	18 c.c.
393	" " 1	7.36 a.m., No. 156 day	3300	20 c.c.
395	" " 4	10.42 a.m., No. 156 day	3360	10 c.c.
399	" " 3	3.49 p.m., No. 156 day	5200	2 c.c.*
401	" " 3	10.33 a.m., No. 157 day	4950	1 c.c.*
402	" " 4	11.20 a.m., No. 157 day	5200	0.4 c.c.*
404	" " 3	3.50 p.m., No. 157 day	4170	11 c.c.
405	" " 3	7.13 a.m., No. 158 day	4000	11.5 c.c.

* Sample was drawn with an injector.

a small injector used. This spring is composed of at least four smaller ones, the total amount of the flow of these four being about 0.6 litre per minute (see Fig. 3).

According to C. Genser,(10–11) the radon content of the "Hindenburgsquelle" (Oberschlema) is 13500 Mache. This value, however, was obtained only once, and never again since. The radon content of the waters of Oberschlema is usually about 3000 Mache. According to Genser,* the "Hindenburgsquelle" should be called the "Brunnen" with the highest radon content in the world, while the "Wettinquelle" (Brambach) is the "Quellen" (spring) with the highest radon content in the world. The radon content of the "Wettinquelle" is, however, 1800–2400 Mache. Imbó,(12) in 1939, also reported that the radon content of the water of Lacco Ameno was about 5000 Mache. This value was also obtained only once, and it seems that the radon content of the spring is usually considerably lower than 3000 Mache. The spring newly discovered in Masu-

(10) C. Genser, *Z. Deutsch. Geol. Gesellschaft.* **85**(1933), 482.

(11) C. Genser, *Geologische Rundschau,* **23**(1932), 188.

* Dr. Genser measured the radon content of this spring only once. Therefore repeated measurements are necessary. According to him, the total residue of "Hindenburgsquelle" is very low. He concludes that the water of "Hindenburgsquelle" is not deep-water (Tiefeswasser). It is described that the radon source of this spring seems to be uranium mica. In this spring, a number of heavy metals (Co, Ni, Bi, etc.) were detected spectroscopically by Fresenius. A geological map of Oberschlema is shown in his paper and it is similar to that of Masutomi. The radon content of the most important mineral springs of Oberschlema is shown below:

Name	Radon Content (Mache)	Amount of Flow (l./Min.)
(1) Hindenburgsquelle	13500	1
(2) Bismarckquelle	3000	
(3) Bore-hole No. 1	620	12
(4) Bore-hole No. 2	305	33
(5) Radiumgesenk	330	35

(12) G. Imbó, La Ricerca Scientifica ed il progresso tecnico nell' economia nazionale, Anno X(1939), 992.

tomi is therefore probably the most radioactive in the world, or its radioactivity may be said to be at least the second in the world.

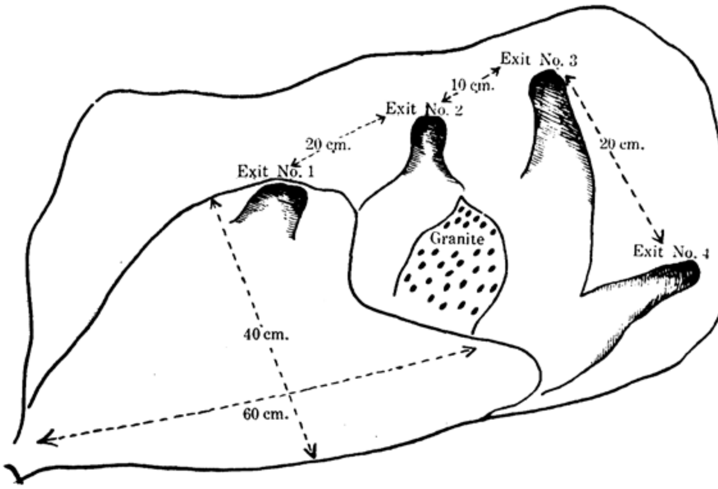

Fig. 3.

Table 5. Radon Content of Dairokuten-no-izumi (A49).

(The sample was taken with a measuring cylinder at the place where the spring water flows away.)

No.	Date	Radon Content (Mache)	
398	2.45 p.m., No. 156 day	2920	Sample : 10 c.c.
403	2.04 p.m., No. 157 day	2800	Sample: 12 c.c.

Table 6. Water Temperature of Dairokuten-no-izumi.

Spring	Date	Water Temperature (°C.)	Spring	Date	Water Temperature (°C.)
No. 1	No. 155 day	22.0	No. 3	No. 155 day	22.0
	No. 156 day	22.0		No. 156 day	22.0
	No. 157 day	23.0		No. 157 day	22.0
				No. 158 day	22.0
No. 2	No. 155 day	21.5	No. 4	No. 155 day	21.0
				No. 156 day	21.0
				No. 157 day	23.0

Radon Content of Spring A49 on No. 183–186 Days.

After rainy days, the radon content of spring A49 increased on No. 183–186 days and showed the following extremely high values.

Table 7.

No.	Date	Radon Content (Mache)	Temp. of Spring (°C.)
(1)	9.24 a.m., No. 183 day	7000	
(2)	10.51 a.m., No. 183 day	9230	
(3)	12.05 p.m., No. 183 day	6500	22.0
(4)	12.55 p.m., No. 183 day	7600	
(5)	1.41 p.m., No. 183 day	6500	
(6)	4.46 p.m., No. 183 day	6000	
(7)	8.14 a.m., No. 184 day	4800	
(8)	10.38 a.m., No. 184 day	5600	
(9)	11.38 a.m., No. 184 day	4700	22.0
(10)	1.58 p.m., No. 184 day	4800	
(11)	3.20 p.m., No 184 day	4700	
(12)	8.58 a.m., No. 185 day	6700	
(13)	11.58 a.m., No. 185 day	7000	
(14)	2.41 p.m., No. 185 day	5500	22.0
(15)	3.57 p.m., No. 185 day	7300	
(16)	4.55 p.m., No. 185 day	6800	
(17)	7.31 a.m., No. 186 day	7920	
(18)	8.07 a.m., No. 186 day	7900	22.0
(19)	9.54 a.m., No. 186 day	6800	

3. Effect of Snow on the Radon Content of Mineral Springs of Masutomi.

In his previous papers,(13-15) the author described the effect of rain on the composition of the hot springs of Yunohanazawa, Hakone.* It is well known that meteorological conditions, as well as volcanic activities and earthquakes, appear to change the chemical composition of hot springs. The variation in the radon content of hot springs, in particular, has been studied by a number of investigators with the object of clarifying the mechanism of hot springs. K. Shiratori,(16) for example, reported in 1927 that the radioactivity of hot springs in the San-in underwent changes since the great earthquake of May 23, 1925. N. P. Pentcheff(17) also reported in 1928 that the great earthquakes of April 14 and 18, 1928, in Bulgaria, changed the radioactivity of the springs. K. Noguchi(18) reported in 1939 that the radon content of the water from the fumaroles of Volcano Asama changed considerably with the eruption of the volcano. As one

(13) K. Kuroda, this Bulletin, **15**(1940), 156.
(14) K. Kuroda, *ibid.*, **15**(1940), 65.
(15) K. Kuroda, *J. Chem. Soc. Japan*, **64**(1943), 153.
* The author studied the seasonal variation of the radium content of "Gongen-no-yu" spring. The seasonal variation of the radon content of this spring was studied by Dr. Noguchi (not yet published).
(16) K. Shiratori, *Science Repts. Tohoku Imp. Univ.*, **16**(1927), 613.
(17) N. P. Pentcheff, *Compt. rend.*, **187**(1928), 243.
(18) K. Noguchi, *J. Chem. Soc. Japan*, **60**(1939), 7.

of the leading investigations on variations in the radon content of mineral springs from meteorological causes, we have P. Ludewig's(19-20) results of continuous measurements of radioactivity of the mineral springs of Brambach. Ludewig found that both the amount of the flow and the radon content of them increase after abundant rain and decrease after dry weather. He reported, further, that the variation in the radon content of "Trinkquelle" in Oberschlema was related to the quantity of the flow of this spring, and the amount of radon in it diminished when the quantity of flow increased. The variation in the radon content of mineral springs and related problems have been discussed by numerous geochemists(21-26) in Japan, the results already obtained being as follows:

(1) The lower the temperature, the higher the radon content of mineral springs.
(2) The greater the amount of flow, the higher the radon content of mineral springs.
(3) The relation between the radon content and the pH of mineral springs is not yet clear.

Seeing that this geochemically important problem has not yet been sufficiently investigated experimentally, notwithstanding that it has been frequently discussed, the author, in this paper, deals with the variation in radon content of one of the most radioactive mineral springs in Japan, which he discovered with M. Nakanisi(9) on November 2, 1942, at Masutomi.

Methods of Experiment.

(1) Determination of Radon. For determining radon, an I. M. fontactoscope was used. Since the radon content of the spring water was too high, radioactivity was measured by diluting 5 c.c. of the spring water to 500 c.c. with distilled radon-free water.
(2) pH. pH was measured colorimetrically, B. T. B. used as indicator.
(3) Cl'. 10 c.c. of the spring water was titrated with a standard solution of silver nitrate, 10% $K_2Cr O_4$ solution used as indicator.
(4) CO_2. 10 c.c. of the spring water was titrated with 0.1 N sodium carbonate solution, phenolphthalein used as indicator.
(5) HCO'_3. 20 c.c. of the spring water was titrated with 0.1 N HCl, methyl orange used as indicator.

(19) P. Ludewig, *Phys. Z.* **22** (1921), 121.
(20) P. Ludewig, *ibid.*, **25** (1924), 280.
(21) I. Iwasaki, *J. Chem. Soc. Japan,* **59** (1938), 1019; **60** (1939), 999; **61** (1940), 367.
(22) S. Matuura, I. Iwasaki and R. Hukusima, *J. Chem. Soc. Japan,* **61** (1940), 225.
(23) I. Iwasaki, I. Ukimoto and K. Hosi, *ibid.*, **63** (1942), 19.
(24) S. Goda, *Shanghai Sizenkagaku Kenkyūsyo Ihō,* **8** (1939), 269.
(25) Imamura, *Science* (Japan), **11** (1941), 372.
(26) K. Noguchi, *Zisin,* **14** (1942), 228.

(6) Amount of Flow. Owing to great difficulties in measuring the amount of the flow of the springs at "Higurasi-no-huti" (A8, A9 etc.), those of other springs, such as A2 and B6, were measured.

Accuracy of the Determinations.

With the I. M. fontactoscope, it is rather difficult to determine the radon content of mineral springs containing so much radon as spring A9 with errors of less than 1%. Provided the sampling, dilution, and the radioactivity measurements were made with utmost care and the experimental conditions were maintained always constant, it is not considered so difficult a matter to determine the 1000–2000 Mache of radon in mineral springs with errors of less than 2–3%. Our measurements were always made near the place where spring A9 issues. In January and February, however, owing to the intense cold in Masutomi even in the day-time, the water sample in the instrument froze while the radioactivity was being measured, so that the writer was compelled to conduct his experiment in a cold room or in a corridor, for the reason that in doing so in a warm room, the radon would escape from the instrument with any expansion of the air. Often the air temperature was below 0°C when the experiment was performed, but in our calculation we had to take 0°C for air temperature, because the partition coefficient of radon for temperatures below 0°C are not given in Dr. Iimori's textbook.

Results.

The results of the experiments are shown in the following Tables.

Table 8 (Omitted).

Table 9.

No. of Day	Temp. (°C.)	pH	Cl′ g./l.)	HCO_3' (g./l.)	CO_2 (g./l.)
(1)	15.0	6.4(6.8)	——	——	——
(2)	14.5	6.3(6.7)	3.822	2.460	0.501
(3)	13.8	6.4(6.9)	3.954	2.470	0.503
(4)	14.2	6.5(6.9)	3.989	2.430	0.438
(5)	13.7	6.5(6.9)	4.007	2.600	0.475
(6)	13.3	6.4(6.9)	3.917	2.519	0.473
(7)	13.0	6.4(7.0)	4.047	2.520	0.467
(8)	13.5	6.4(6.9)	4.114	2.510	0.505
(9)	14.0	6.4(6.9)	3.971	2.518	0.482
(10)	13.5	6.4(6.9)	4.040	2.505	0.475
(11)	15.0	6.4(6.9)	4.044	2.513	0.465
(12)	13.5	6.4(7.0)	4.040	2.526	0.466
(13)	13.0	6.4(6.9)	4.044	2.519	0.470
(14)	12.0	6.4(6.8)	4.042	2.520	0.468
(15)	12.2	6.4(6.8)	4.022	2.521	0.471

Table 9.—(*Continued*)

No. of Day	Temp. (°C.)	pH	Cl′ (g./l.)	HCO_3′ (g./l.)	CO_2 (g./l.)
(16)	11.9	6.4(6.8)	3.711	2.521	0.467
(17)	11.0	6.3(6.8)	4.085	2.519	0.488
(18)	11.4	6.4(6.8)	4.079	2.519	0.488
(19)	13.0	6.4(6.9)	4.078	2.523	0.466
(20)	12.0	6.4(6.9)	4.290	2.519	0.468
(21)	11.6	6.4(6.9)	4.219	2.534	0.472
(22)	11.2	6.4(6.9)	4.078	2.519	0.467
(23)	—	6.4(6.9)	4.090	2.523	0.465
(24)	11.7	6.4(6.9)	4.134	2.521	0.463
(25)	12.0	6.4(6.9)	4.328	2.518	0.459
(26)	13.0	6.4(6.8)	4.645	2.518	0.462
(27)	11.5	6.4(6.8)	4.274	2.514	0.463
(28)	11.0	6.4(6.8)	4.143	2.512	0.467
(29)	10.9	6.4(6.8)	4.140	2.514	0.463
(30)	11.1	6.4(6.8)	4.131	2.518	0.462
(31)	12.0	6.4(6.9)	4.157	2.510	0.467
(32)	12.1	6.4(6.9)	4.136	2.520	0.470
(33)	11.9	6.4(6.9)	4.081	2.516	0.461
(34)	10.9	6.4(7.0)	——	——	——
(35)	9.6	——	4.095	2.520	0.467
(36)	9.0	6.4(6.9)	4.090	2.518	0.467
(37)	10.0	6.4(6.9)	4.104	2.519	0.465
(38)	10.2	6.4(6.9)	4.104	2.518	0.464
(39)	10.0	6.4(6.9)	4.082	2.514	0.467
(40)	9.8	6.4(6.8)	4.082	2.518	0.468
(41)	9.2	6.4(6.8)	4.090	2.515	0.467
(42)	9.0	6.4(6.8)	4.090	2.528	0.468
(43)	8.2	6.4(6.7)	4.086	2.517	0.468
(44)	9.0	6.4(6.8)	4.183	2.518	0.468
(45)	9.1	6.4(6.9)	4.112	2.519	0.467
(46)	8.6	6.4(6.8)	4.115	2.521	0.468
(47)	8.8	6.5(6.9)	4.122	2.521	0.470
(48)	9.2	6.4(6.9)	4.128	2.519	0.467
(49)	10.0	6.4(7.0)	4.131	2.522	0.465
(50)	8.5	——	4.131	2.521	0.473
(51)	8.8	——	4.088	2.519	0.470
(52)	9.2	——	4.085	2.519	0.468
(53)	9.0	——	4.095	2.519	0.468
(54)	8.4	——	4.095	2.520	0.473
(55)	8.4	——	4.081	2.518	0.467
(56)	8.0	6.5(6.9)	4.131	2.521	0.472
(57)	8.2	6.4(6.8)	4.112	2.568	0.473
(58)	8.5	6.4(6.8)	4.196	2.531	0.473
(59)	8.6	6.4(6.9)	4.184	2.546	0.470
(60)	8.0	6.4(6.9)	4.152	2.528	0.466
(61)	9.3	6.4(6.7)	4.237	2.522	0.467

Table 9.—(*Concluded*)

No. of Day	Temp. (°C.)	pH	Cl′ (g./l.)	HCO_3' (g./l.)	CO_2 (g./l.)
(62)	8.9	6.5(6.8)	4.241	2.528	0.472
(63)	8.8	6.4(6.8)	4.219	2.528	0.472
(64)	8.0	6.4(6.8)	4.250	2.542	0.478
(65)	8.3	6.4(6.9)	4.219	2.531	0.471
(66)	7.8	6.4(6.8)	4.184	2.528	0.471
(67)	8.0	6.4(6.8)	4.149	2.528	0.468
(68)	8.6	6.4(6.8)	4.202	2.522	0.468
(69)	8.5	6.4(6.8)	4.219	2.540	——
(70)	—	——	4.198	——	——
(71)	8.0	6.5(6.9)	4.200	2.528	——
(72)	8.2	6.5(6.8)	4.113	2.399	——
(73)	9.0	6.5(7.0)	3.975	2.307	——
(74)	9.0	6.5(6.9)	4.078	2.374	——
(75)	9.0	6.4(6.8)	4.180	2.353	——
(76)	—	6.5(6.9)	(1.638)*	(1.161)*	(0.218)*
(77)	9.5	6.4(6.8)	4.042	2.311	0.482
(78)	9.9	6.4(6.8)	4.134	2.411	0.513
(79)	9.5	6.4(6.9)	4.159	2.440	0.498
(80)	9.2	6.4(6.8)	4.205	2.411	0.503
(81)	9.4	6.4(6.7)	4.184	2.449	0.493
(82)	9.0	6.4(6.8)	4.095	2.443	0.465
(83)	9.0	6.4(6.9)	4.050	2.388	0.537
(84)	9.0	6.4(6.9)	3.976	2.388	0.435
(85)	9.0	6.4(6.9)	3.923	2.452	0.403
(86)	9.0	6.4(6.9)	4.041	2.452	0.447
(87)	10.0	6.5(6.9)	4.014	2.348	0.472
(88)	9.0	6.5(6.9)	3.719	2.300	0.418
(89)	9.0	6.4(6.8)	3.819	2.368	0.435
(90)	9.0	6.4(6.8)	3.787	2.308	0.361
(91)	9.0	6.4(6.8)	3.900	2.328	0.361
(92)	9.5	6.4(6.7)	3.780	2.312	0.403
(93)	9.0	6.4(6.7)	4.093	2.444	0.412
(94)	8.2	6.4(6.7)	4.078	2.368	0.432
(95)	9.0	6.4(6.7)	4.017	2.348	0.386
(96)	9.0	6.4(6.7)	——	——	——
(97)	9.0	6.4(6.7)	——	——	——
(98)	9.0	6.4(6.7)	——	——	——

* The sample was evidently diluted by the thaw water.

Table 10.

No. of Day	Radon Content (Mache)	Radon Content (Curie)	No. of Day	Radon Content (Mache)	Radon Content (Curie)
(1)	1600	5820×10^{-10}	(50)	1240	4520×10^{-10}
(2)	1530	5570 "	(51)	1290	4690 "
(3)	1720	6260 "	(52)	1300	4730 "
(4)	1640	5970 "	(53)	1350	4920 "
(5)	1550	5640 "	(54)	1200	4370 "
(6)	1500	5460 "	(55)	1320	4800 "
(7)	1530	5570 "	(56)	1260	4580 "
(8)	1670	6080 "	(57)	1310	4770 "
(9)	1680	6110 "	(58)	1305	4750 "
(10)	1690	6150 "	(59)	1300	4730 "
(11)	1820	6620 "	(60)	1500	5460 "
(12)	1820	6620 "	(61)	1690	6150 "
(13)	1900	6910 "	(62)	1700	6180 "
(14)	1740	6330 "	(63)	1650	6000 "
(15)	1700	6190 "	(64)	1740	6330 "
(16)	1800	6550 "	(65)	1490	5430 "
(17)	1830	6660 "	(66)	1530	5570 "
(18)	1610	5860 "	(67)	1460	5310 "
(19)	1640	5970 "	(68)	1450	5280 "
(20)	1620	5900 "	(69)	1520	5530 "
(21)	1640	5970 "	(70)	1380	5020 "
(22)	1620	5900 "	(71)	1330	4830 "
(23)	1570	5710 "	(72)	1285	4680 "
(24)	1550	5640 "	(73)	1400	5100 "
(25)	1660	6040 "	(74)	1500	5460 "
(26)	1740	6330 "	(75)	1510	5490 "
(27)	1810	6580 "	(76)	1620	5900 "
(28)	1750	6370 "	(77)	1530	5570 "
(29)	1630	5930 "	(78)	1610	5860 "
(30)	1740	6330 "	(79)	1600	5820 "
(31)	1780	6480 "	(80)	1650	6000 "
(32)	1660	6040 "	(81)	1550	5640 "
(33)	1660	6040 "	(82)	1635	5950 "
(34)	1430	5200 "	(83)	1700	6180 "
(35)	1420	5170 "	(84)	1810	6590 "
(36)	1430	5200 "	(85)	1750	6370 "
(37)	1540	5600 "	(86)	1370	4980 "
(38)	1550	5640 "	(87)	1490	5430 "
(39)	1530	5570 "	(88)	1750	6370 "
(40)	1370	4980 "	(89)	1750	6370 "
(41)	1360	4950 "	(90)	1270	4620 "
(42)	1430	5200 "	(91)	1360	4950 "
(43)	1260	4580 "	(92)	1480	5380 "
(44)	1390	5060 "	(93)	1590	5780 "
(45)	1420	5170 "	(94)	1330	4840 "
(46)	1340	4880 "	(95)	1040	3790 "
(47)	1390	5060 "	(96)	1030	3750 "
(48)	1430	5200 "	(97)	1330	4830 "
(49)	1470	5350 "	(98)	1230	4480 "

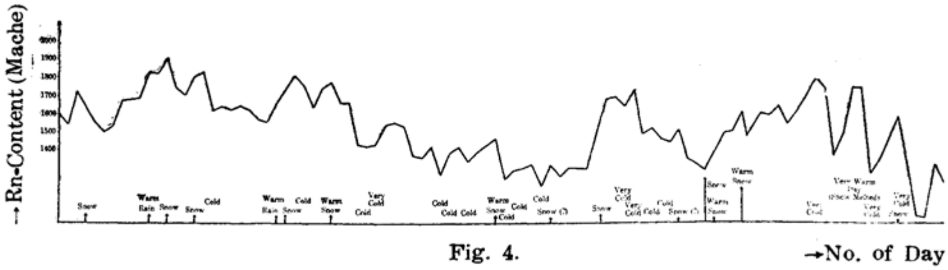

Fig. 4.

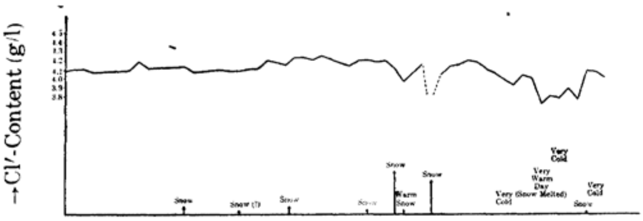

Fig. 5.

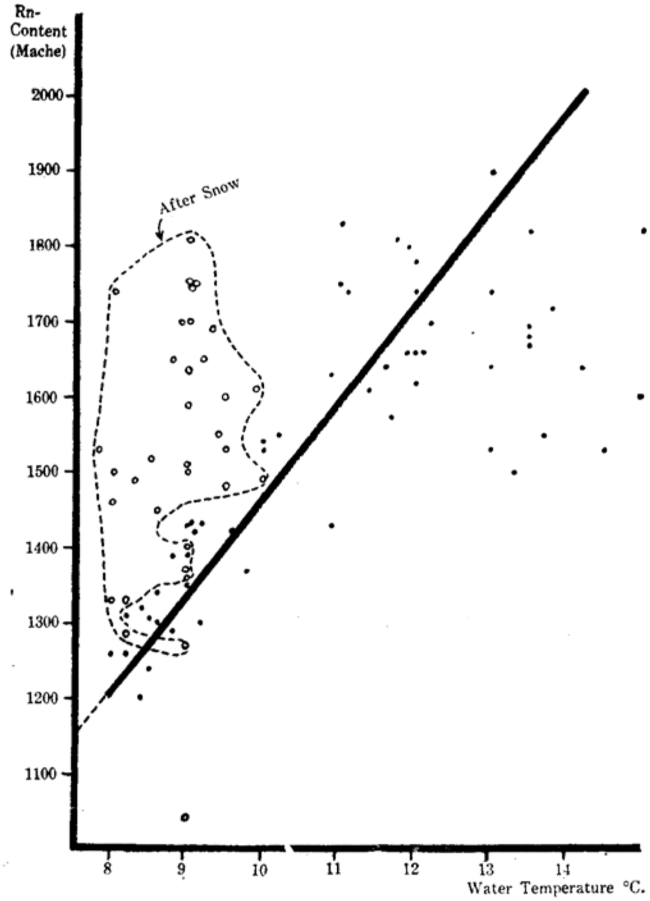

Fig. 6.

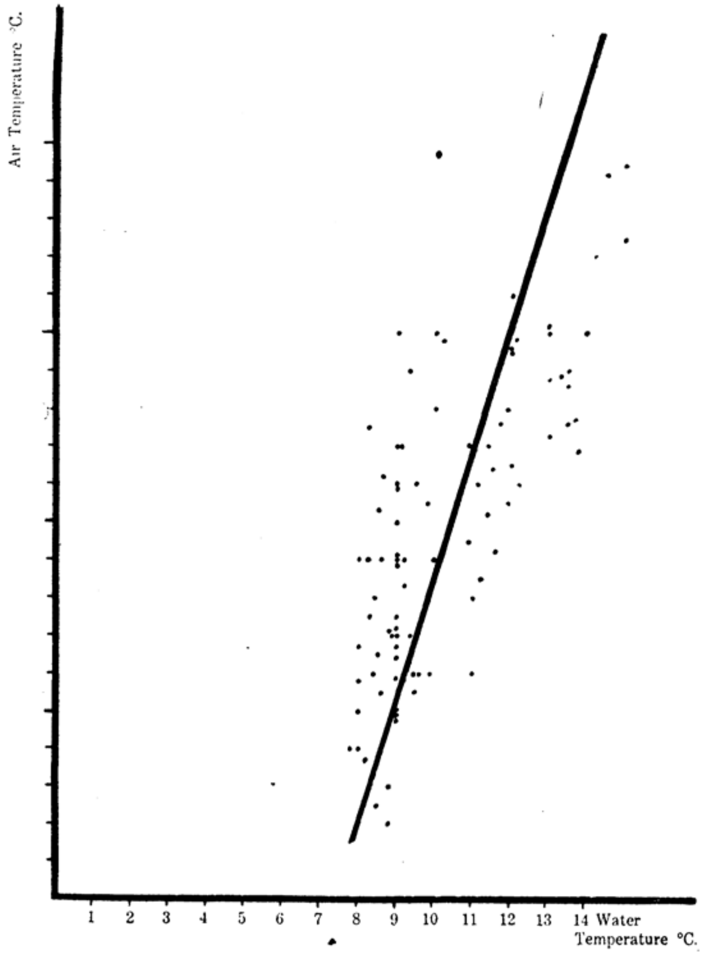

Fig. 7.

Relation between the Temperature and the Radon Content.

From data obtained in the present work it may be concluded that the higher the water temperature the higher the radon content, and the lower the water temperature the lower the radon content.

Fig. 6 shows the relation between the temperature and the radon content of spring A9 during this period. Since, as is shown in Fig. 7, the water temperature of spring A9 evidently depends on the air temperature, it may be expected that the radon content of this spring will increase in summer, and decrease in winter. This expectation, however, has not yet been confirmed experimentally for the reason that this spring having been discovered in November 1942, we are yet without any experimental data covering it for the summer period. The author intends to measure the radon content of this spring as often as possible in the future.

G. Imbó[12] reported in 1939 that the radioactivity of the mineral waters of Lacco Ameno fluctuates with the time as also with the temperature of the water. According to him, rise of temperature is accompanied with increase of radioactivity.

The results of a number of investigators in Japan, however, differ, namely: Lowering of temperature is accompanied with decrease of radioactivity.

K. Noguchi,[18] for example, reported that the radon content of a number of cold springs near Volcano Asama increased when the water temperature declined. He reported further[26] that this rule holds also in the case of the redioactivity of the pond in the Imperial University of Tokyo. According to him, the lower the water temperature the higher the radon content of the pond water. I. Iwasaki[21] found in 1936 that the lower the water temperature the higher was the radon content of the hot springs of Rendaizi, Sizuoka Prefecture. It is notable that the former rule (the higher the water temperature the higher the radon content) holds with springs having high radon content (Lacco Ameno, Masutomi), while the latter rule (the lower the water temperature the higher the radon content) holds in the case of springs with low radon content (the cold springs near Volcano Asama, the pond water of Tokyo Imperial University, the hot springs at Rendaizi, etc.)

Relation between the pH and the Radon Content.

The variation in the pH value of spring A9 was very small during this period and no relation could be established between its pH value and its radon content. Other investigators also failed to find any relation between the variation in pH value and radon content of mineral and hot springs.

Relation between the Chlorine Content and the Radon Content.

The variation in chlorine content of spring A9 is less marked than that of the radon content. The maximum and minimum values of the chlorine content of spring A9 in this period are as follows:

Maximum value	4.645 g./l.	No. 26 day.
Minimum value	3.711 g./l.	No. 16 day.

The variation in chlorine content does not seem to run a regular course. It seems to differ according to circumstances. There are, at least, two modes of variation, namely:

(a) After the snow on No. 60 and No. 61 days, the chlorine content of spring A9 evidently increased, reaching maximum on No. 64 day. The variation in both chlorine and radon content ran quite parallel this time (see Fig. 8).

(b) After the snow of No. 72–76 days, the chlorine content generally decreased, whereas the radon content increased.

After the snow of No. 60–61 days, which lay about 5 cm. deep, the radon content of spring A9 increased, reaching maximum (1740 Mache) on No. 64 day. The chlorine content which also increased showed maximum value (4.250 g. per litre) the same day (No. 64 day).

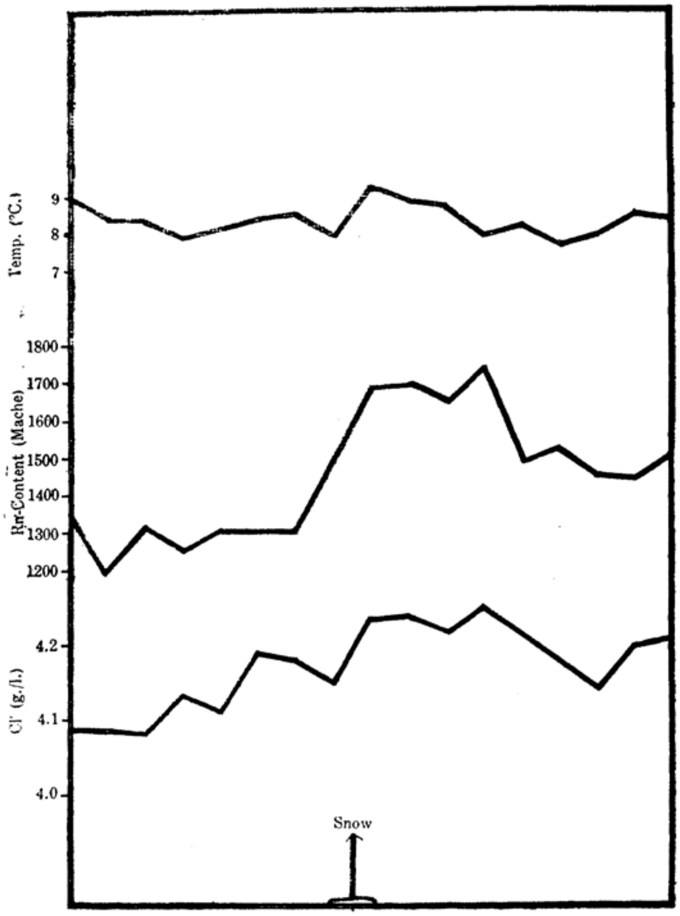

Fig. 8. Effect of Snow of No. 60 and No. 61 days on the Cl' and Rn Content of Spring A 9.

As has already been pointed out by S. Oana and the present author, a relation holds between the chlorine content and the total residue from the mineral springs of Masutomi, that is, the higher the chlorine content the higher the total residue of mineral springs. It may be concluded, therefore, that the total residue of spring A9 increased after snow—a phenomenon quite similar to that which was observed by the author in Yunohanazawa, Hakone Hot Spring. The total residue of the hot springs of Yunohanazawa increased after abundant rain. The water temperature of the hot springs of Yunohanazawa rose also after abundant rain. No such regular variation in the water temperature of spring A9 could be observed, the explanation for which is supposed to be the fact that its water temperature is strongly affected by the air temperature, as mentioned above.

Relation between the Bicarbonate Content and the Radon Content.

The variation in bicarbonate content of spring A9 is not so marked as that of the radon content. The maximum and minimum values of the bicarbonate content of spring A9 in this period are as follows:

Maximum value	2.600 g./l.	No. 5 day.
Minimum value	2.300 g./l.	No. 88 day.

The relation between the bicarbonate and the radon content is not clear.

Relation between the Carbon Dioxide Content and the Radon Content.

The variation in the carbon dioxide content of this spring was not so marked as that of the radon content. The maximum and minimum contents are as follows:

Maximum value	0.537 g./l.	No. 83 day.
Minimum value	0.361 g./l.	No. 90 day and 91 day.

Although, after the snow of No. 60–61 days, the carbon dioxide content of spring A9 fluctuated parallel with that of the radon content, that of the carbon dioxide content of spring A9 after the snow of No. 72–76 days, did not run parallel with that of the radon content. In fact, during the whole period of this investigation, it was very difficult to trace any relations between the carbon dioxide content and the radon content of spring A9.

Relation between Atmospheric Pressure and the Radon Content.

No relation could be established between atmospheric pressure and the radon content of spring A9.

Relation between the Amount of Flow and the Radon Content.

Owing to the great difficulty of accurately measuring the amount of the flow of spring A9, the amounts of the flow of spring B6 (Higasiobi-no-izumi) and spring A2 (Tuganero No. 1) were measured and the results compared.* Generally speaking, it may be concluded that the radon content of spring A9 increases when the amount of flow increases. It was pointed out by P. Ludewig in 1921 and 1924, that the same relation holds in the mineral springs of Brambach and Oberschlema. K. Noguchi also reported in 1939 that this rule holds in a number of cold springs near Volcano Asama. It will be noticed that the opposite relation (the larger the amount of flow the lower the radon content) is yet to be reported.

* The amount of strong radioactive water that flows from the springs in Masutomi is very small. Springs A1, A2, and A3 have probably the highest value in Masutomi, the amount of the flow of spring A1 being 250 litres per minute, according to the report of the Hygienic Laboratory of Tokyo. The amount of the flow of spring B6 was measured three times by Mr. Oana and the present author, preceding this investigation, with the following results.

Date	Amount of Flow (l./min.)
July 8, 1939	1.41
Oct. 18, 1940	1.85
May 20, 1942	2.16

Mr. Oana noticed that the radon content of this spring fluctuates but very slightly, being constant for long intervals of time.

During the intense cold that sometimes follows snow, with the snow on the surface of the ground hardening and remaining so for several days, increase in the amount of flow is not very marked, with the result that the radon content does not increase very markedly.

Effect of Snow on the Radon Content, and the Chemical Composition of Spring A2.

In this section will be found the results of the determinations of radon and the analyses of spring A2 that were made daily during the period from No. 74 day to No. 89 day. The method of analysis and that of determining the radon have been already described. The sample was drawn from the point where the spring water enters the bath room of Hotel Tuganero. For determining the radon, 500 c.c. of the spring water was always used. Both the radon content and the chemical composition of this spring varied greatly, but with some regularity. It will not be difficult to explain the relations between the variations in the radon content and in the chemical composition of the spring water with the aid of the hypothesis proposed in this paper. The experimental data are summarized in the annexed Table.

Table 11.

No. of Day	Temp. (°C.)	Rn Content (Mache)	Cl′ (g./l.)	CO_2 (g./l.)	HCO_3' (g./l.)	Amount of Flow (l./min.)
(74)	30.0	2.64	4.2165	——	2.449	11.50
(75)	—	3.22	4.1520	0.532	2.511	11.40
(76)	29.8	3.50	4.2266	0.538	2.486	12.77
(77)	29.6	2.96	3.9950	0.542	2.532	14.32
(78)	—	3.06	4.2584	0.574	2.494	13.00
(79)	29.4	2.98	4.0953	0.573	2.532	13.75
(80)	30.0	3.70	4.1733	0.613	2.581	10.02
(81)	29.6	3.10	4.1556	0.552	2.615	10.02
(82)	29.2	4.22	4.0847	0.537	2.428	10.65
(83)	29.2	5.18	3.9818	0.588	2.388	10.10
(84)	29.0	3.85	3.9393	0.572	2.360	11.00
(85)	28.8	5.00	3.9038	0.553	2.328	12.52
(86)	—	—	3.9251	0.543	2.367	12.20
(87)	—	4.51	3.9180	0.552	2.348	11.82
(88)	—	—	3.8577	0.588	2.316	10.47
(89)	—	—	3.8612	0.623	2.328	11.49

(1) Variation in the Amount of Flow. The amount of the flow of spring A2 showed its first maximum value on No. 77 day, and the second maximum on No. 85 day. On No. 77 day, the chlorine content showed a sudden decrease. After No. 80 day, the chlorine content gradually declined, notwithstanding the evident increase in the amount of flow. All of these facts point to dilution by thaw water. The chlorine content of the thaw water is regarded, of course, as being very small.

(2) Temperature Variation. The water temperature of spring A2 gradually declined. No reason can be given for the maximum value that was observed on No. 80 day.

(3) Variation in the Chlorine Content. It will be seen that the chlorine content suddenly decreased after the heavy snow of No. 76 day. It decreased gradually after No. 80 day, with the melting of the snow.

(4) Variation in the Carbon Dioxide Content. Although the variation in the carbon dioxide content was very marked, it was rather difficult to ascertain the relation between the quantity of flow and the carbon dioxide content. The first maximum value was observed on No. 80 day, followed by another on No. 83 day.

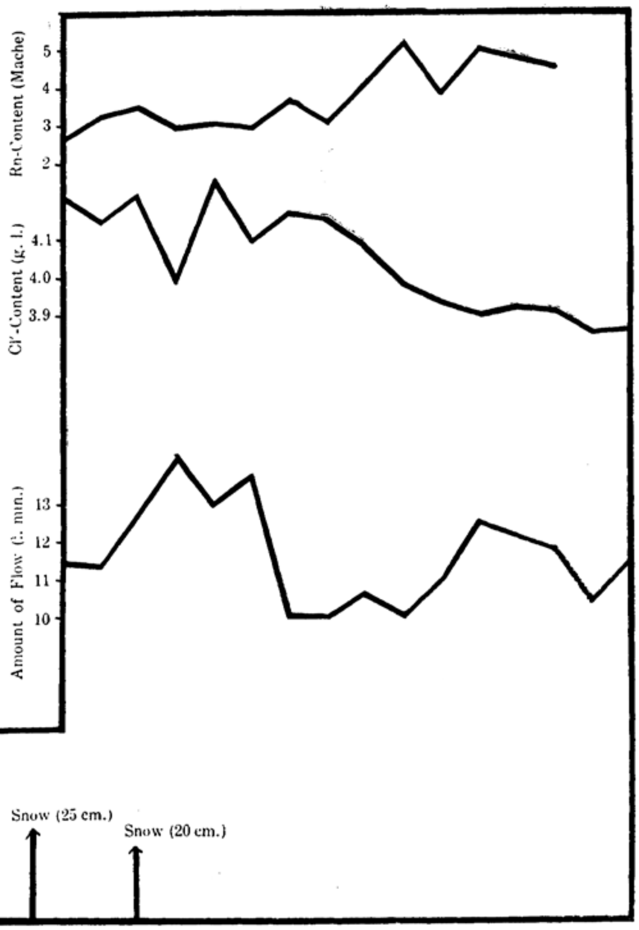

Fig. 9. Effect on Snow on the Radon Content of Spring A 2 (Tuganero-no-yu) (No. 74 Day～No. 89 day.)

(5) Variation in the Bicarbonate Content. The variation in the bicarbonate content was also very marked. A maximum value was observed on No. 81 day, five days after the heavy snow, after which it gradually decreased.

(6) Variation in the Radon Content. Generally speaking, the radon content of spring A2 increased during the period of this investigation. What is notable, however, is that until No. 81 day the radon content and the chlorine content fluctuated in parallel, after which the relation was reversed, (the higher the radon content the lower the chlorine content).

General Considerations.

The variation in the radon content of this spring may be satisfactorily explained by assuming that "the original spring water was diluted by the thaw water, the two having mixed immediately after the snow melted." This thaw water contained very little of either chlorine or radon, because it had not passed through any thick layers of rocks or soils, whose radon it would have dissolved, had it passed through the rocks and soils which were believed to hold large amounts of radon. There is little doubt that during the period between No. 76 day and No. 81 day, spring A2 was diluted by such thaw water. After this period, however, it is believed that the thaw water, which had passed through fairly thick layers of rocks or soils containing the large amounts of radon, or had reached localities where large amounts of radon were accumulated, mixed with the original spring water, with the result that from the following day, the radon content increased and the chlorine content decreased. This conclusion receives support from the fact that the amount of flow increased after No. 81 day, reaching maximum on No. 85 day.

Variation in the Radon Content of Spring A9 during the Period from No. 129 day to No. 151 day.

The radon content of spring A9 was measured daily during the period from No. 129 day to No. 151 day, again. On No. 136 day, an extremely high value (12000) Mache was obtained (Table 12). In view of C. Genser's report in 1933 that the radon content of "Hindenburgsquelle" showed 13500 Mache under the most favourable conditions, the radon content of spring A9 on No. 136 day may also be regarded as an instance showing the large amount of radon that could be concentrated in mineral spring waters under suitable conditions. The I_0 not being so constant, the value of 12000 Mache is not so accurate. The author believes, however, that nothing was wrong with the instrument. Another high value (7400 Mache) was obtained on No. 132 day. It may be noticed that both these high values were obtained on rainy days. On No. 140 day, however, no such high value was obtained, notwithstanding the heavy rain. On No. 132 day and No. 136 day, no marked changes in the Cl′-, CO_2-content and in the pH and water temperature occurred. After the heavy rain on No. 140 day, the chlorine content diminished, then increased daily until it reached maximum value (4.2158 g. per litre) on No. 149 day. During this period the variations in the radon content was almost parallel with that of the chlorine content, as in the case after the snow of No. 60–61 days.

Table 12. Radon Content of Spring A9 on No. 136 Day.

Sample Taken: 5 c.c. Time: 10.00 a.m.

$\theta_1=34$ min.	$I_{01}=43.0$	$\theta_5=62$ min.	$I_{05}=52.5$
$\theta_2=45$ min.	$I_{02}=52.9$	$\theta_6=65$ min.	$I_{06}=60.6$
$\theta_3=50$ min.	$I_{03}=53.3$	$\theta_7=69$ min.	$I_{07}=65.0$
$\theta_4=55$ min.	$I_{04}=51.2$		

I_0 (mean value) $=53.3$ $K=1.11$
$V=5460$ c.c. $Q \doteqdot 12000$ Mache.
$v=500$ c.c.

Table 13.

No. of Day	pH	CO_2 (g./l.)	Cl′ (g./l.)	Radon Content (Mache)	Water Temperature (°C)
(129)	6.3	0.495	4.1981	2500	13.8
(130)	6.4	0.534	4.2123	2820	12.5
(131)	6.3	0.488	4.1626	2600	13.9
(132)	6.4	0.483	4.1445	**7400**	15.0
(133)	6.3	0.477	4.1662	2020	15.6
(134)	6.2~6.3	0.472	4.1520	1760	13.7
(135)	6.4	0.434	4.1485	2900	13.7
(136)	6.3	0.482	4.1623	**12000**	14.5
(137)	6.3	0.463	4.1301	1820	13.0
(138)	6.3	0.478	4.1875	1740	13.2
(139)	6.3	0.465	4.1769	1710	15.0
(140)	6.3	0.467	4.0953	1890	15.5
(141)	6.3	0.459	4.1095	1780	13.7
(142)	6.3	0.489	4.1485	1750	14.0
(143)	6.3	0.479	4.1516	1860	14.0
(144)	6.3	0.479	4.1662	1930	13.8
(145)	6.3	0.467	4.1520	1850	17.2
(146)	6.3	0.484	4.1875	2030	13.8
(147)	6.4	0.520	4.1452	2240	15.3
(148)	6.3	0.436	4.2087	2110	13.6
(149)	6.3	0.405	4.2158	2030	14.6
(150)	6.3	0.413	4.2003	2160	14.6
(151)	6.3	0.415	4.1662	2060	15.0

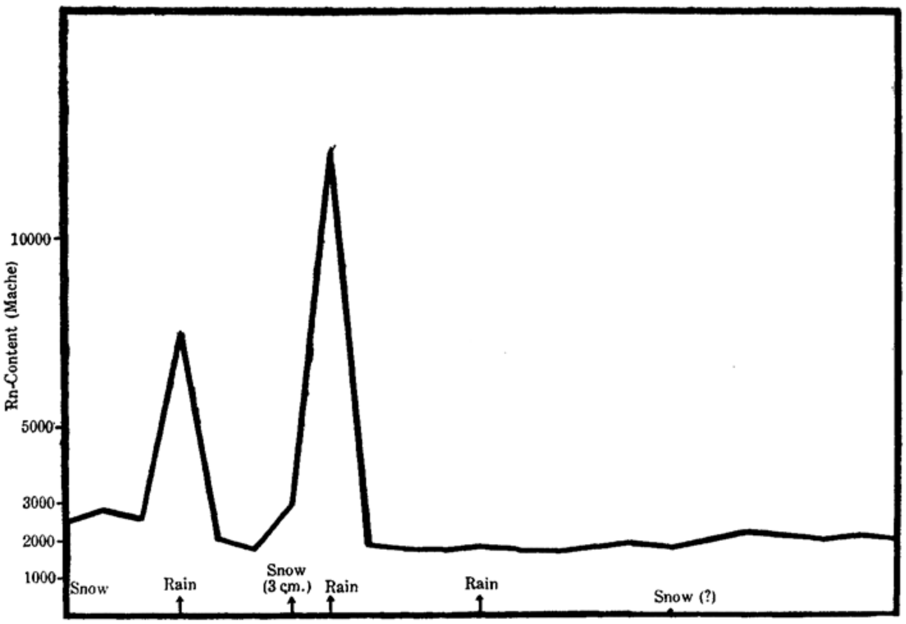

Fig. 10. Variation of the Radon Content of Spring A 9.
(No. 129 Day—No. 151 Day.)

4. Earthquakes and the Radon Content of Mineral Springs of Masutomi.

It is said that the spring at Ginsento changed considerably after the great earthquakes of 1923. Notwithstanding that K. Shiratori(16) has reported on the effect of earthquakes on the radon contents of mineral springs, this geochemically interesting phenomenon is not yet satisfactorily studied. It is very difficult to ascertain any marked relation between the variations in the radon content of the mineral springs of Masutomi and the small earthquakes* that occurred in Yamanasi Prefecture, during the period of this investigation. (see Table 14).

Table 14.

No. of Day	Centre of Disturbance	Preliminary Tremors	No. of Day	Centre of Disturbance	Preliminary Tremors
(-10)	Hunazu	3.9 seconds	(49)	Kohu	2.2 seconds
(37)	Hunazu	6.0 seconds	(123)	Kohu	(?)

In the opinion of the author, who intends to study this problem more closely in future, the change in radioactivity of mineral springs is independent of phenomena occurring at depths within the earth, such as earthquakes, but is dependent on causes due to external conditions, mainly meteorological.

There seems to be no relation between the radioactive elements and earthquakes or volcanic activities. The only way in which the radioactivity of mineral springs would change with an earthquake is when the path of the mineral springs changes, with consequent changes in the amount of flow or in the velocity of flow. In such cases, the radon content of mineral springs changes, because the amount of radon accumulating in the mineral water would change with the flow velocity of the spring water.

5. Radon Content of Frozen Mineral Water.

In early January 1943, the author found that the spring water of A9 had frozen several meters distant from the place where it issued. Its thickness was about 10 to 20 c.m., and its area several square meters. This spring water, of which only very little flowed, cooled from its original temperature (16°C) to 0°C when it flowed several meters from where it issued. The radon content of this spring water, the temperature of which was 0°~1°C, was found to be about 250~470 Mache. With the intention of determining the radon content of its frozen mass, it was carefully crushed and washed with distilled water. Several blocks of this frozen mass (20 g.) were added to warm distilled water (480 g.), and after the pieces had melted the water was placed in the I. M. fontactoscope, shaken

* The author expresses his hearty thanks to Mr. S. Homma, of the Central Meteorological Institute, for his courtesy in acquainting the author with details of the intensity and the time of the earthquakes.

vigorously for thirty seconds and its radioactivity measured. The frozen mass of this spring water was found to contain determinable amounts of radon. In order to test whether or not this radioactivity was due to that of the spring water which adhered to the surface of the frozen mass and the frozen mass itself was not radioactive, the author tried the following experiment.

A large block of the frozen spring water was carefully washed first with distilled water and then with hot water until almost the whole of its outer part had melted away, leaving only the center of the mass (25 g.), which was carefully dissolved in warm distilled water and the total volume made up to 500 c.c. The water sample thus prepared was placed in the fontactoscope, and the radioactivity measured, with the following results (see Table 15, No. 3).

Table 15. Radon Content of Frozen Mineral Water.

		Sample Taken	Radon Content (Mache)
(1)	A8 No. 1	10 g.	6.5
(2)	A8 No. 2	20 g.	14.0
(3)	A8 No. 3	25 g.	15.0
(4)	A8 No. 4	20 g.	4.5
(5)	A6 No. 1	30 g.	0.0

The value obtained comes close to that obtained in the former experiment, whence it may be concluded that about 15 Mache of radon is contained in 1 k.g. of the frozen mass of spring A8. The radon contents of samples taken elsewhere are shown in the annexed table.

The radon content of the frozen spring water taken from places more distant from the exit of the spring (Sample No. 4) is lower than that of samples taken from places nearer the exit (Sample No. 2 and No. 3) showing that the frozen masses from more distant places froze earlier than those from nearer places. This may be explained by assuming that radon cannot escape from ice, but that it decays, following the well-known decay equation,

$$Q = Q_0 e^{-\lambda t} \quad \ldots\ldots\ldots\ldots \quad (1)$$

Exit of Spring A8.

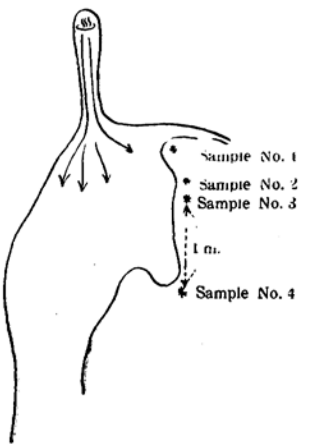

Fig. 11.

Assuming that (1) the radon had not escaped from the spring water when it froze and (2) that the spring water was not diluted by thaw water, the age of the ice may be calculated from the foregoing equation.

Q_0 ...... radon content of spring water cooled to 0°C. (250~470 Mache)

Q ...... radon content of the sample (ice). (15~14 Mache)

From the calculations the conclusion is that the ice was about 15~18 days old. These values are, of course, not so exact, seeing that some of the radon must have escaped when the water froze. The value obtained by the foregoing calculations is therefore believed to show the upper limits of the age of the ice. Therefore it may safely be said that the ice was formed not more than 15~18 days before—a conclusion not contradicted by actual observations.

6. Day-Time-Variations of the Radon Content of Spring A9.

The radon content of the water of spring A9, which was drawn from the inner part of the exit of the spring by means of an injector, was measured a number of times during the day-time. On No. 94 day it was very cold in the morning, but much warmer later in the day. The ground which froze in the morning melted in the afternoon, and the radon content showed the following variations. The content which was very low in the morning (1050 Mache) increased in the day-time, reaching at 3.45 p.m. the maximum of 3040 Mache. In the evening, when the air temperature dropped below 0°C and the ground froze, the radon content diminished again considerably. On No. 95 day, no such great increase in the radon content was observed, it being cloudy and cold the whole day. The ground which froze hard the preceding night did not melt later in the day.

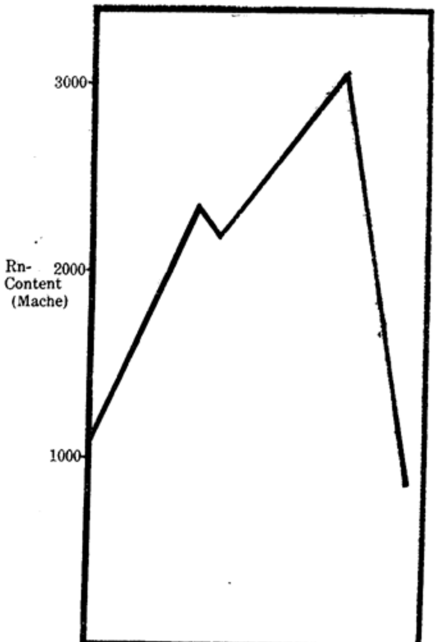

Fig. 12. Day-time variation of the rodon content of the water of spring A 9, which was drawn from the inner part of the exit of the spring by means of an injector. (No. 94 day.)

The experiment which was repeated a number of times led to the following general conclusions:

(1) In winter, the radon content is very low in the morning, increases in the day-time with the thawing of the frozen ground and suddenly diminishes in the evening when the ground freezes again.

(2) On cloudy days in winter, no such marked increases in the radon content is observed.

(3) In spring, when it is warm, the variations during the day-time are not so large.

(4) The radon content of the water of spring A9, which was taken in the usual way, a 5 c.c. pipette used, does not show such marked variations in the day-time.

The constitution of spring A9 is supposed to be like that shown schematically in the following Fig. 13. The spring-water issuing from very small exits in the inner part of the spring contains, according to external conditions, different amounts of radon (large in the day-time when it is warm, and small when it is cold). These marked fluctuations in the radon content of the spring water are probably owing to the fact that the amount of water flow is very small and the velocity of flow very low. The radon content of the water that issues from the main exit of the springs is almost constant, even when the external conditions change considerably. The results of the present experiment show that the radon content of mineral springs is influenced by delicate changes in external conditions, such as air temperature, snow and rain, these changes manifesting themselves at points very close to the exits of the mineral springs.

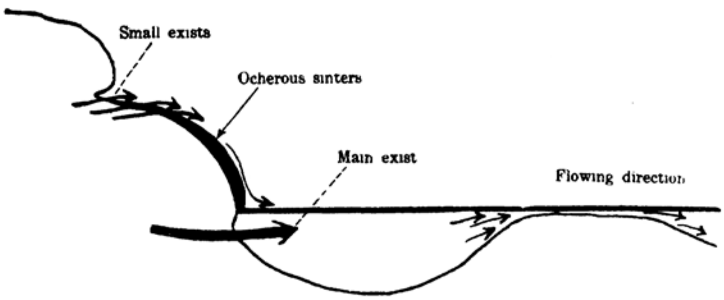

Fig. 13. Constitution of Spring A 9.

Table 16. The radon content of the water of spring A9, which was drawn from the inner part of the exit of the spring by means of an injector. (No. 94 day)

	Sample Taken (c c.)	Radon Content (Mache)
(1) 9.08 a.m.	1	1050
(2) 11.51 a.m.	1	2360
(3) 0.28 p.m.	1	2190
(4) 3.45 p.m.	0.5	3040
(5) 4.18 p.m.	2.5	2440
(6) 4.54 p.m.	4.5	1530
(7) 5 30 p.m.	2.5	860

Table 17. The radon content of the water of spring A9, which was drawn from the inner part of the exit of the spring by means of an injector. (No. 95 day)

	Sample Taken (c.c.)	Radon Content (Mache)
(1) 8.46 a.m.	1	2370
(2) 9.21 a.m.	1	2270
(3) 10.10 a.m.	1	2180
(4) 10.46 a.m.	1	1820
(5) 11.28 a.m.	1.5	1420

Table 17.—(*Continued*)

		Sample Taken (c.c.)	Radon Content (Mache)
(6)	0.05 p.m.	1	1520
(7)	1.07 p.m.	2.5	1620
(8)	1.50 p.m.	1	1830
(9)	2.30 p.m.	1	1570
(10)	3.18 p.m.	1	1250
(11)	4.57 p.m.	1	1820

Table 18. The radon content of the water of spring A9, which was drawn from the inner part of the exit of the spring by means of an injector. (No. 90 day)

		Sample Taken (c.c.)	Radon Content (Mache)
(1)	9.12 a.m.	2	1730
(2)	10.12 a.m.	1	2400
(3)	0.20 p m.	0.5	2140
(4)	2.15 p.m.	0.5	3870
(5)	3.30 p.m.	1	1850

Table 19. The radon content of the water of spring A9, which was drawn from the inner part of the exit of the spring by means of an injector. (No. 97 day)

		Sample Taken (c.c)	Radon Content (Mache)
(1)	10.37 a.m.	0.75	2010
(2)	0.40 p.m.	0.5	2830
(3)	2.10 p.m.	0.6	3290
(4)	3.07 p.m.	0.55	2930
(5)	4.45 p.m.	0.45	2370
(6)	5.50 p.m.	0.55	3780

Table 20. The radon content of the water of spring A9, which was drawn from the inner part of the exit of the spring by means of an injector. (No. 123 day)

		Sample Taken (c.c.)	Radon Content (Mache)
(1)	9.25 a.m.	0.7	2100
(2)	11.13 a.m.	1.0	2220
(3)	12.32 p.m.	1.0	2120
(4)	3.40 p.m.	1.6	1860

Table 21. The radon content of the water of spring A9, which was taken in the usual way, a pipette used. (No. 123 day)

		Sample Taken (c c.)	Water Temp. (°C)	Radon Content (Mache)
(1)	11.48 a.m.	1.0	—	1910
(2)	1.26 p.m.	1.0	14.0	2010
(3)	2.06 p.m.	5.0	14.0	1910
(4)	5.24 p.m.	10.0	14.5	1835

Table 22. The radon content of the water of spring A9, which was drawn from the inner part of the exit of the spring by means of an injeotor. (No. 153 day)

	Sample Taken (c.c.)	Water Temp. (°C)	Radon Content (Mache)
(1) 7.27 a.m.	1	15.3	1830
(2) 8.53 a.m.	1	15.5	2000
(3) 10.22 a.m.	1	16.0	2240
(4) 11.42 a.m.	1	17.0	2060
(5) 0.58 p.m.	1	18.1	2040
(6) 2.22 p.m.	1	18.2	1850
(7) 3.48 p.m.	1	19.0	2140
(8) 5.15 p.m.	1	19.0	1800
(9) 6.37 p.m.	1	19.0	1740

Table 23. The radon content of the water of spring A9, which was taken in the usual way, a 5 c.c. pipette used. (No. 153 day)

	Sample Taken (c.c.)	Water Temp. (°C)	Radon Content (Mache)
(1) 6.27 a.m.	5	15.2	1540
(2) 8.07 a.m.	5	15.3	1390
(3) 9.32 a.m.	5	15.8	1450
(4) 10.57 a.m.	5	16.5	1350
(5) 0.20 p.m.	5	17.1	1270
(6) 1 34 p.m.	5	18.5	1480
(7) 3 05 p.m.	5	18.60	1480
(8) 4.33 p.m.	5	19.0	1350
(9) 5.47 p.m.	5	19.0	1260

7. The Radon Content of Springs in Masutomi, and Its Relation to the Carbon Dioxide Content.

The Radon Content of Spring A2, and Its Relation to the Carbon Dioxide Content.

The radon content of spring A2 was determined at a number of places. The spring water flows into a pipe about three meters long and then into a large bath, forming a fall about one meter high. The places where the water samples were taken are shown in Fig. 14. 500 c.c. of the water was used in determining the radon, with results as shown in Table 24. As will be seen from Figs. 15~17, marked relations hold between radon content and temperature, radon content and pH, and between radon content and carbon dioxide content.

This experiment also showed, as in the case of spring A6, that the radon in mineral springs escapes from the spring water very quickly after it has issued, and that the percentage of the radon that escapes is almost equal to that of the percentage of carbon dioxide that escapes.

Table 24.

Sample (See Fig. 13)	Radon Content (Mache)	CO_2 Content (g./l.)	pH	Temperature (°C)
No. 1	3.76	0.686	6.4	29.0
No. 2	3.16	0.518	6.4	29.0
No. 3	2.38	0.296	6.5	28.0
No. 4	1.38	0.234	6.7	27.0

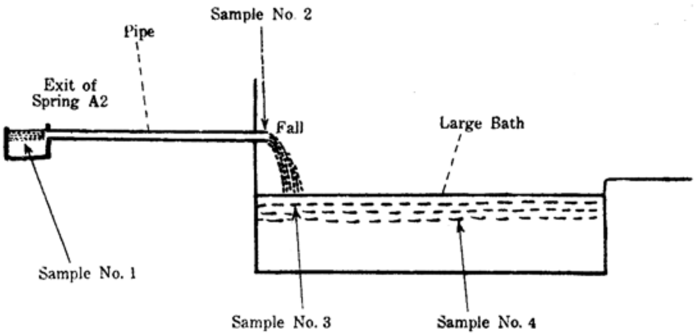

Fig. 14.

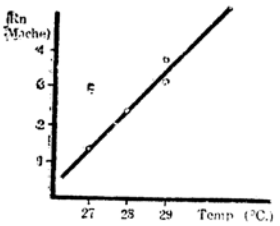

Fig. 15. Rn Content and Temp. of Spring A 2.

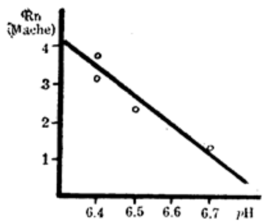

Fig. 16. Rn Content and pH of Spring A 2.

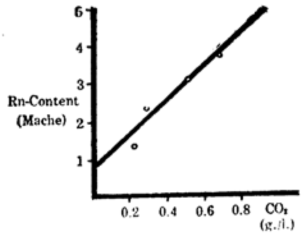

Fig. 17. Rn and CO_2 Content of Spring A 2.

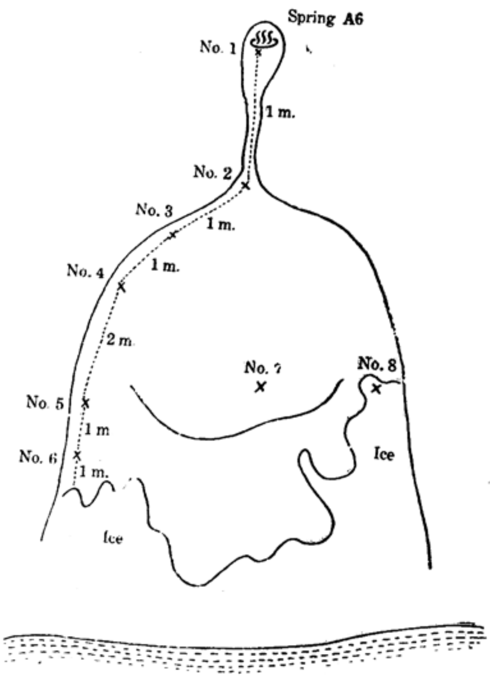

Fig. 18.

Radon Content of Spring A6, and Its Relation to the Carbon Dioxide Content.

The author intended to determine the radon content at various places in the path of the flow of Spring A6. The places where water samples were taken are shown in Fig. 18. The results of the experiment are given in Table 25. As will be seen from Figs. 19~20, marked relations hold between radon content and pH, radon content and carbon dioxide content, and between the radon content and distance from the place where the spring issues. From this experiment, it was found that radon and carbon dioxide diminish in fairly equal proportion.

The considerable change in the value of pH is believed to be owing to the escape of carbon dioxide.

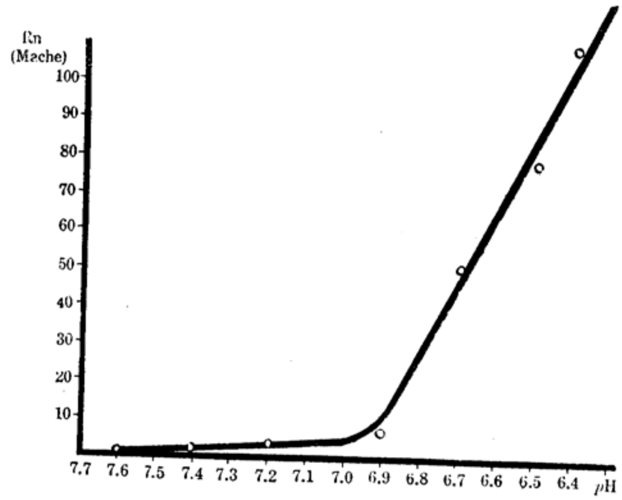

Fig. 19. Rn Content and pH of Spring A 6.

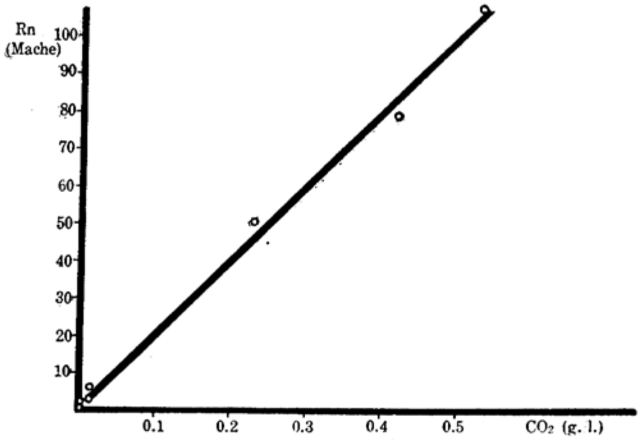

Fig. 20. Rn and CO_2 Content of Spring A 6.

Table 25.

	Radon Content (Mache)	CO_2 Content (g./1.)	pH	Temperature (°C)
No. 1	110	0.538	6.4	25.0
No. 2	79	0.422	6.5	23.8
No. 3	51	0.232	6.7	22.2
No. 4	7	0.013	6.9	21.7
No. 5	2	0.0070	7.4	14.0
No. 6	1	0.0011	7.6	7.0
No. 7	3.6	0.014	7.2	19.0
No. 8 (ice)	0	—	—	0

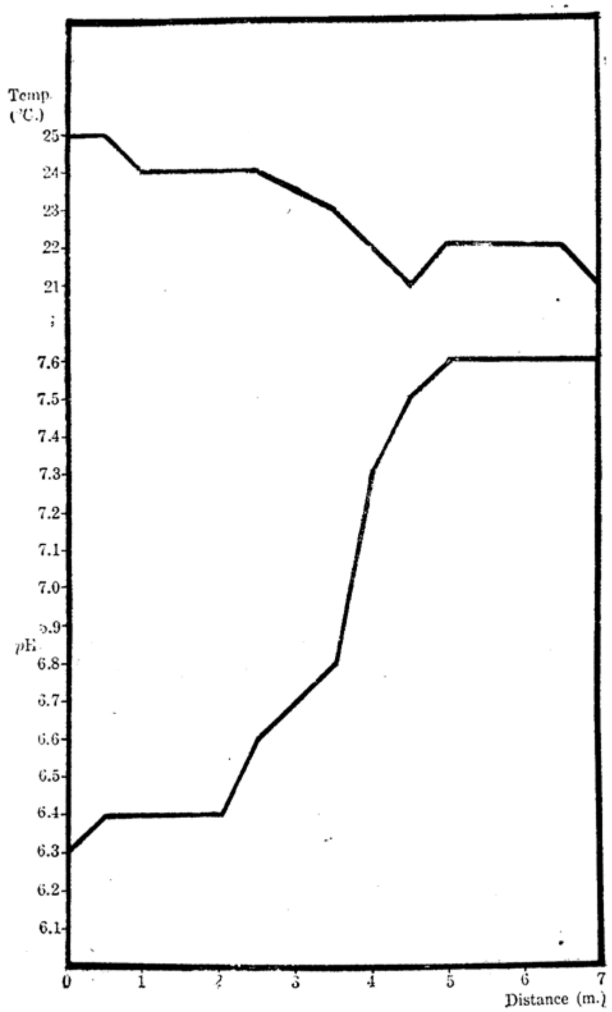

Fig. 21. Temperature and pH of Spring A 6.

Table 26. Spring A7. (March 1943)

Distance from the place where the spring issues (c.m.)		Temperature (°C)	pH	CO_2 Content (g./1.)	Cl′ Content (g./1.)
No. 1	0	24	6.3	0.277	4.085
No. 2	50	24	6.3	0.258	4.120
No. 3	100	23	6.3	0.204	4.127
No. 4	150	23	6.4	0.185	4.124
No. 5	200	23	6.6	0.152	4.109
No. 6	250	23	6.7	0.111	4.124
No. 7	300	23	6.7	0.166	4.145
No. 8	350	23	7.0	0.084	4.127
No. 9	400	22	7.2	0.030	4.063
No. 10	450	22	7.5	0.020	——

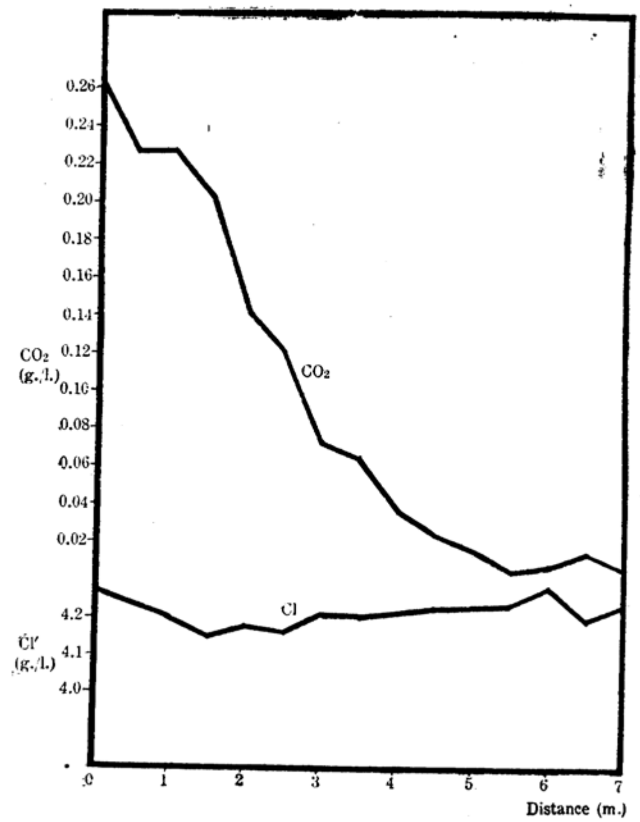

Fig. 22. Cl' and Rn Content of Spring A 6.

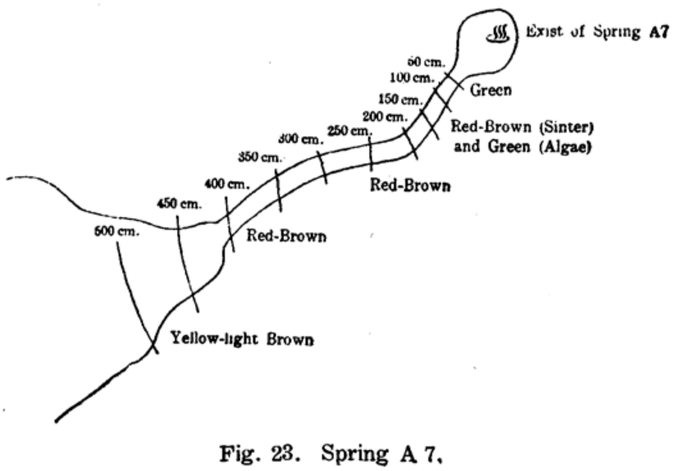

Fig. 23. Spring A 7.

Table 27. pH and CO_2 Content of Spring A6. (March 1943)

Distance from the place where the spring issues (cm)		Temperature (°C)	pH	CO_2 Content (g./l.)	Cl′ Content (g./l.)
No. 1	0	25	6.3	0.265	4.273
No. 2	50	25	6.4	0.228	4.237
No. 3	100	24	6.4	0.228	4.202
No. 4	150	24	6.4	0.204	4.152
No. 5	200	24	6.4	0.142	4.184
No. 6	250	24	6.6	0.1205	4.163
No. 7	300	23.5	6.7	0.0740	4.219
No. 8	350	23	6.8	0.0662	4.205
No. 9	400	22	7.3	0.0389	4.219
No. 10	450	21	7.5	0.0253	4.237
No. 11	500	22	7.6	0.0175	4.237
No. 12	550	22	7.6	0.0058	4.244
No. 13	600	22	7.6	0.0097	4.290
No. 14	650	22	7.6	0.0156	4.202
No. 15	700	21	7.6	0.0078	4.248
No. 17	250 (left)	24	6.6	0.1655	4.202
No. 18	300 (left)	24	6.8	0.0878	4.343
No. 19	350 (left)	23	7.2	0.0741	4.237
No. 20	400 (left)	23	7.5	0.0547	4.280
No. 21	450 (left)	23	7.6	0.0195	4.138
No. 22	500 (left)	23	7.6–7.7	0.0156	4.180
No. 23	550 (left)	22	7.7	0.0097	4.234
No. 24	600 (left)	22	7.7	0.0058	4.265
No. 25	650 (left)	22	7.7	0.0058	4.184
No. 27	700 (left)	21	7.7	0.000	4.230

Table 28. Spring A9.

Distance (c.m.)		Temperature (°C)	pH	CO_2 Content (g./l.)	Cl′ Content (g./l.)
(1)	0	15.0	6.4	0.286	4.148
(2)	50	—	—	0.206	4.163
(3)	100	—	—	0.129	4.163
(4)	150	—	—	0.175	—

Table 29. Spring A9.

Distance (c.m.)		Temperature (°C)	pH
(1)	0	9.0	6.4
(2)	100	7.0	6.4
(3)	200	5.0	6.6
(4)	300	3.5	6.6
(5)	350	3.0	6.7
(6)	400	2.0	6.8

The Results Obtained at Spring B4.

The results obtained at Spring B4 are shown in the following Table.

Table 30.

	Temperature (°C)	Cl′ (g./l.)	CO_2 (g./l.)	pH
No. 1	22	1.670	0.561	6.0
No. 2	21	1.666	0 462	6.1
No. 3	18	1.574	0.475	6.1
No. 4	—	1.605	0.440	6.1
No. 5	19	—	0.411	6.1
No. 6	18	—	0.390	6 1
No. 7	17.5	—	0.318	6.1
No. 8	16.5	—	0.187	6.4
No. 9	14.5	—	0.102	6.7

[Rn/CO_2] Ratio in the Mineral Springs of Masutomi.

It has already been shown that although radon and carbon dioxide diminish in fairly equal proportions from springs A6, A7, A2, etc., the chlorine and bicarbonate contents are almost constant at various places in the path of the flow of these springs. It was also pointed out that after the snow of No. 60–61 days, the radon conent and the CO_2 content of spring A9 fluctuated quite parallel.

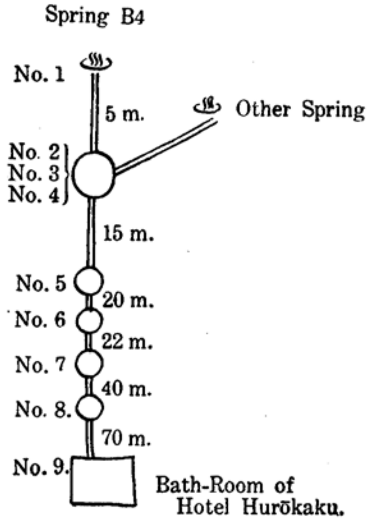

Fig. 24.

It seems very difficult to explain why the Rn/CO_2 ratio of springs at Masutomi, issuing so close to one another, should differ so much. Table 31 shows, for example, the Rn/CO_2 ratio of springs A2, A6, A7, A8 and A9. The following are two probable explanations:

Hypothesis (1). The CO_2 and Rn have the same origin.

In this hypothesis, we suppose that the CO_2 and the radon have the same origin. The Rn/CO_2 ratios of all the mineral springs are supposed to be practically the same at depths in the earth. Since the velocity of flow differs with attendant conditions, the time required for the spring waters to reach the surface of the earth from the place where the carbon dioxide and the radon are supplied differs with each spring. Since radon decays, following the equation,

$$Q = Q_0 e^{-\lambda t} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (1)$$

the Rn/CO_2 ratio of the springs, whose velocity of flow is small, is supposed to be also small, while in those springs, whose velocity of flow is large, the Rn/CO_2 ratio is supposed to be also large.

Hypothesis (2). The CO_2 and Rn have different origins.

In this hypothesis, we suppose that the CO_2 and the Rn have different origins. It is supposed that the Rn/CO_2 ratio of the original spring water of Masutomi (at depths within the earth) is not so large. Its ratio is supposed to be almost the same as that of spring A2. The reason why the above-ratio for springs A8 and A9 has an extremely high value is probably that springs A8 and A9 issue through layers of radioactive soils or of uranium minerals formed secondarily from radium or uranium in the spring waters. Springs A8 and A9 take up considerable amounts of radon passing through such layers near the surface of the earth. It is supposed that psring A2, on the other hand, does not pass through such layers as have high contents of radioactive elements. Owing to the insufficiency of one data, it is not easy to say which of the two hypotheses is more likely to be correct. In the author's present opinion, the latter hypothesis is more probable than the former, because springs with apparently large velocity of flow, such as A2 and A3, have low Rn/CO_2 ratios notwithstanding that, according to the former hypothesis, such springs with higher velocity of flow ought to have larger Rn/CO_2 ratios.

Table 31. Rn/CO_2 Ratio of a Number of Springs.

	Rn Content (Mache)	CO_2 Content	$[Rn/CO_2]$
Spring A2	3.76	0.686	5.5
Spring A6	110	0.538	204
Spring A7	60	0.277	216
Spring A8	1300	0.292	4460
Spring A9	1530	0.467	3280

8. Chemical Composition of the Mineral Springs of Masutomi.

The analysis of the cold spring "Nibuzawa" No. 1 by the Tokyo Imperial Hygienic Laboratory, which was performed as early as 1914, gave the result as shown in Table 32. This spring may thus be classified as "Earthy Common Salt Springs". The cold springs "Kamigawara" (also called "Wadegawara") No. 1, "Yunokubogawara-no-yu", "Tugane-yu" No. 1 (A), and "Totikubo" No. 1 having nearly the same composition as that of "Nibuzawa" No. 1, were classified as "Earthy Common Salt Springs".

The cold spring "Kuridaira" No. 1, the composition of which is nearly the same as that of "Nibuzawa" No. 1, differing from the latter only in the smaller proportion of bicarbonates of alkaline-earth metals, was classified as "Common Salt Spring". The analyses of spring A9 and spring A49 are shown in Table 33. The chemical composition of these two springs being almost alike, they may be classified as "Earthy Common Salt Spring". The total analyses of the other springs have not yet been made.

The chlorine contents of A6, A7, A8, A9 and the four springs in Hotel Tuganero are almost alike. The chlorine contents of the springs of "C-Group" (five springs in Ginsento) are slightly higher than those of the other springs. The chlorine content of spring B4 is exceptionally low.

Table 32. Chemical Composition of Spring B1 (Nibuzawa). (Tokyo Imperial Hygienic Laboratory). (1914)

Temperature 23.5°C. Total residue: ca. 7.41. Flow of water: ca. 77 hectolitres in 24 hours.

Cations		Anions.	
K^+	0.13765	Cl^-	3.14674
Na^+	2.43382	SO_4^{--}	0.46988
NH_4^+	0.00106	HCO_3^-	1.47765
Ca^{++}	0.23677		
Mg^{++}	0.02085	HBO_2	0.16910
Fe^{++}	0.00140	H_2SiO_3	0.16792
		CO_2	0.26432

Radioactivity . . . . 394.12 Mache's units

Table 33.

	Spring A 9	Spring A 49
Total Residue (g./l.)	9.14	9.11
Ca^{++}	0.311	0.303
Mg^{++}	0.010	0.007
$(Fe_2O_3+Al_2O_3$	0.090	0.086)
H_2SiO_3	0.144	0.124
SO_4^{--}	0.611	0.598

9. The Constituents in Minute Quantities of the Mineral Springs of Masutomi.

Aside from the radon and the radium contents, the constituents of the mineral waters were studied in detail at Masutomi, a part of which was described in the previous papers. In this section, a general survey of our investigation into the minute constituents of the mineral springs of Masutomi will be made.

(a) *Spectroscopic Analysis.* A number of mineral springs of Masutomi were studied spectroscopically by Prof. Kimura, and the following elements were detected. (See Table 34).

Table 34. An Example of the Results of the Spectroscopic Analysis by Prof. Kimura.

Tuganero No. 1 (Spring A2)

Li Na K Cu Be Mg Ca Sr Ba B Al Si Ge Ti P As V Mn Fe

The presence of beryllium in Tuganero-no-yu (A2 and A3) is very interesting.

(b) *Boron.* An appreciable amount of boron is often found in the mineral springs of Masutomi.[27] The geochemical significance of the high

(27) T. Tagaya, *Bull. Inst. Phys. Chem. Research*, **21** (1942), 165.

content of boron and the strong radioactivity of mineral springs is not yet clear.

(c) *Heavy Metals, Such as Copper, Lead and Zinc.*

In the previous paper, it was reported that the copper and the lead contents of most of the mineral springs in Masutomi seem to be less than about 50γ per litre. Exact experiments recently performed showed that the copper content of the "Kinsento" spring, the "Tuganero" spring, etc. is about 10γ per litre. The copper was determined volumetrically by the dithizone method as described in the previous papers. The lead content of some of the mineral springs in Masutomi, according to the author's recent analysis, is believed to exceed 100γ per litre; these will be described in greater detail later.

The zinc content of the springs of Masutomi is 26~224γ per litre, and *the smaller the pH value the higher the zinc content of these springs.*

(d) *V, Cr, Mo.* Vanadium in the mineral springs of Masutomi was detected by Sandell's colorimetric method. Chromium and molybdenum were, however, not detected by this method.

(e) *Radium.* The radium content of mineral springs in Masutomi was measured for the first time by T. Nakai, in 1936–1937. The author measured the radium content of 38 springs and his determinations are still being continued. The spring E1 showed the highest value (84.36×10^{-12} g. Ra per litre.).

(f) *Uranium.* The presence of uranium was detected by the fluorescence method, its concentration in spring A9 being estimated by M. Nakanisi to be about $10^{-4}\sim10^{-5}$ g. per litre. The results will be reported later in detail.

10. Sinters of Masutomi.

It is mentioned in the "Mineral Springs of Japan" that beautiful sinter-coated leaves occur on the creek-bed into which the water of Kinsento flows. The yellowish-brown deposits are sometimes coated with green algae. Beautiful sinters are formed near the exits of springs in Masutomi, that of spring B1 (Nibusawa), spring A6, and spring A7 being the most beautiful. Old sinter-deposits are found on the roadside near "Osiba-spring", the site being now under cultivation. At "Totikubo" a large old sinter, nearly several meters high, is found, the old exit of the spring on top of it having stopped. The spring issues at present from an exit a slight distance away from this old sintre forming a new beautiful sinter deposit (Table 35). The presence of the old sinters near the present exit shows that the exit of the springs had displaced itself in remote times. The chemical composition, surface form and physical properties (porosity and capillary properties) of sinters are supposed to be greatly influenced by living organisms (algae, bacteria etc.) in the springs. The relations between the properties of springs and the sinter deposits require to be chemically studied in greater detail in order to clarify the geochemical meanings of living things in mineral springs as a "catalyzer" in the formation of sinter deposits.

The chemical composition of the sinter of spring A6 taken on Dec. 17, 1943, is shown in Table 35. The sample was taken at places 1–2 meters distant from the exit of the spring. The usual method of gravimetric analysis was used (CO_2 content obtained by difference). Fig. 25 is a

Table 35. Chemical Composition of the Sinter of Spring A6.

CaO	33.52%	45.70%
MgO	0.07	0.10
Fe_2O_3 / Al_2O_3	9.12	12 68
MnO	tr.	tr.
CO_2	22.97	31.90
SO_4''	0.71	0.96
H_2SiO_3	6.23	8.66
H_2O	27.38	——
	100.00%	100.00%

sketch of the external appearance of the sinter of spring A6. The surface colour of the deposits is yellowish-brown to greenish-brown, and its back reddish-brown. Although in external appearance they seem to be ocherous sinters, chemical analysis showed that their main constituent is calcium carbonate. In the usual hot springs, it is believed that when the bicarbonate ion (HCO_3') breaks up, losing carbon dioxide in the atmosphere, normal calcium carbonate is formed and, being insoluble, is precipitated, the reaction being

$$Ca(HCO_3)_2 = CaCO_3 + H_2O + CO_2 .$$

The author tried to analyze the flowing spring at various points over the surface of the sinter deposits, at spring A6, with the object of ascertaining the exact manner in which the sinters of this spring, the external appearances of which so markedly differ at various points, are formed (See Table 25 and 27).

It is interesting that, notwithstanding that the Cl′- and HCO_3' contents are almost constant everywhere, the CO_2 content rapidly diminishes until at the lowest places (about 7 meters distant from the exit of the spring) the CO_2 content of the over-flowing spring water is almost 0 m.g. per litre. Analyses of the sinters at different places are now in progress, the results of which will be reported in due course. Although it is not yet impossible to discuss the

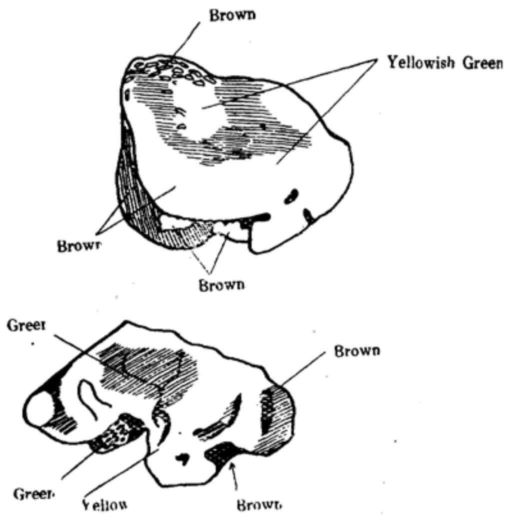

Fig. 25. Sinter-Deposits of Mastomi Spring A6.

"geochemistry of sinter", the results of the present investigation show how important the chemical analysis of the sinters in biological investigations of mineral springs is. Generally speaking, the sinter is supposed to be formed and characterized by the following factors:

(1) Chemical composition and the temperature of the spring water.
(2) Location of the exit of the spring.
(3) Conditions of precipitation.

(a) Oversaturation by cooling.
(b) Oversaturation by evaporation.
(c) Oversaturation by the escape of gas.
(d) Life-phenomena.

The author intends to study the above-mentioned phenomena in greater detail in future in order to be able to understand how the elements, especially the radioactive ones, behave geochemically throughout the following processes:

Lithosphere (Granite)
↓
Hydrosphere (Mineral Springs)
↓
Biosphere (Algae, Bacteria etc.)
↓
Lithosphere (Sinters).

Table 36. Radon Content of Mineral Springs at Totikubo.

	Temperature (°C)	Rn Content (Mache)	Date
Spring No. 1	12	128	No. 96 day
Spring No. 2	12	204	No. 92 day
Spring No. 3	17	950	No. 185 day
Spring No. 4	20	51.1	No. 184 day
Spring No. 5	27.0	15.6	No. 185 day
Spring No. 6	23	4.4	No. 184 day
Spring No. 7	19	11.4	No. 184 day
Spring No. 8	17.4	121	No. 185 day
Spring No. 9	16	9.0	No. 185 day
Spring No. 10	—	7.4	No. 185 day
Spring No. 11	7.0	6.89	No. 92 day

11. General Discussions.

The author recalls here the following two experiments that were made in the laboratory with the object of ascertaining the general laws governing the transition of radon from minerals into liquid media so that with their aid the variations in radioactivity of various springs could be explained.

Spitzuin's Experiment.(28)(29) V. I. Spitzuin placed radioactive mine-

(28) V. I. Spitzuin, *Trav. radium et minerais radioactifs acad. sci. U.R.S.S.*, **2** (1926), 264. *Chem. Abstracts*, **25**, 2914. *Chem. Zentralblatt*, 1927, II, 2442.
(29) V. I. Spitzuin, *ibid.*, **2** (1926), 272.

rals (pitchblend, autunite, samarskite, thorianite, etc.) in flowing water and measured the radioactivity of the water and found that.

(1) *The amount of radon(x) which dissolves in flowing water in unit time is independent of the velocity of the stream.*

He expected from this result that in the case of hot springs, the radon content will diminish with increase in the amount of flow.

(2) *The smaller the size of the particles of the radioactive minerals the larger the amount of radon dissolved(x).*

(3) *Except for thorianite, the higher the water temperature the larger the amount of radon dissolved(x).*

Mache and Markstein's Experiment.[30] In 1935, H. Mache and G. Markstein studied the escape of radon into flowing water from rocks containing radium and experimentally proved that, owing to the slight occlusion property of the rock for various gases, the amount of gas entering the water from unit surface in unit time is independent of the temperature of the water, as also of the amount of radon already present.

They reported that in nature these conditions are only imperfectly approximated.

The Experiments in Masutomi.

The results of the present investigation on the effect of snow and rain on the radon content and on the chemical composition of radioactive springs may be summarized as follows:

(1) In spring A9, the radon content fluctuates more than that of the chlorine, bicarbonate, and carbon dioxide contents.

(2) During the dry season, the lower the temperature the lower the radon content of spring A9.

(3) After snow or rain, the radon content increase considerably.

(4) On very cold days, the radon content suddenly decreases. With these facts, together with the results of other investigators as basis, the mechanism of radioactive springs will now be briefly discussed.

The mechanism of the formation of radioactive springs in Masutomi can be easily understood by assuming that these springs pass through a layer of radioactive minerals (or soils rich in radioactive elements, such as radium and uranium), as in the case of Spitzuin's experiment. Fig. 26 gives a good idea of radioactive springs as they are supposed to be. Spring A has a low radon content (spring A2 in Masutomi, having 3~4 Mache of radon) while spring B has an extremely high radon content (spring A9 in Masutomi having 1500~2000 Mache of radon). Spring B passes through a layer of radioactive elements (not yet clear whether they are radioactive minerals or not). The bulk of the radon in spring B comes from this source of radon, that coming from the original springs being very small in amount, almost the same as that in spring A which does not pass through any source of radon, as will be seen from Fig. 26.

The author's result obtained in Masutomi is *the higher the temperature the higher the radon content of mineral spring,* a result in agreement with Spitzuin's experiment. The author's second conclusion, namely *the higher the amount of flow the higher the radon content,* however, does not

(30) H. Mache and G. Markstein, *Sitzber. Akad. Wiss. Wien, Math.-Naturw. Klasse.* Abt. II a, **144**(1935), 489.

agree with the results obtained by Spitzuin. This contradiction may be explained by assuming that in nature there are numerous sources of radon and paths of spring water. The author's idea will be seen from Fig. 27, in which R_1, R_2, R_3, R_4, R_2', R_3', R_4' are the sources of radon in paths P_1, P_2, P_3, P_4, P_2', P_3', P_4'. In the dry season, when the amount of flow from the springs is very low, the spring water is believed to flow only through

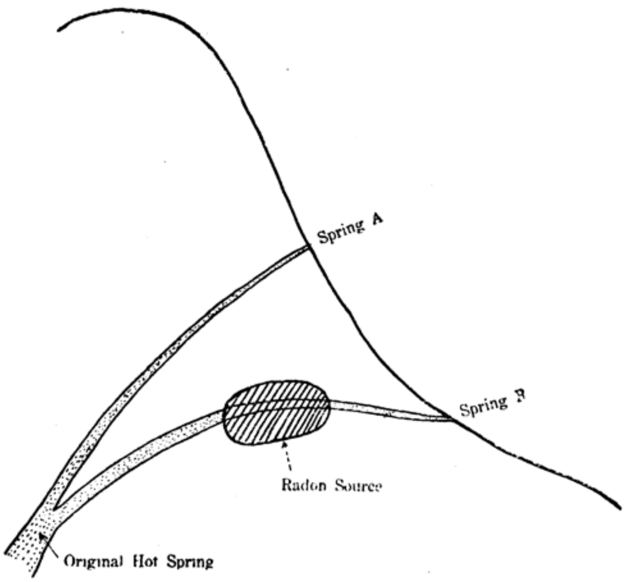

Fig. 26.

path P_1, the other small paths being dry during this period. Since after rain or snow, however, the amount of flow increases, and the spring water passes through all paths, the spring water takes up large amounts of radon that have accumulated in paths P_3, P_3, P_4, P_2', P_3', P_4' during the dry days, whence it follows that the radon content of radioactive springs increases with an increase in the amount of flow following rain or snow.

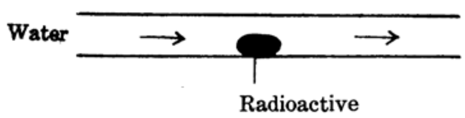

Fig. 27. Spitzuin's Experiment.

In Spitzuin's experiment, the source of radon was radioactive minerals, such as autunite, pitchblend, etc., whereas in Masutomi we do not know whether or not such radioactive minerals exist near the surface of the earth. In the following section, the author attempts to ascertain by calculation whether or not the amount of radium brought from depths in the earth by mineral springs, such as spring A2, A3 (with high radium content), is sufficient to supply radon to the strongly radioactive springs from which only very small quantity of water flows.

One Probable Hypothesis on the Source of Radon.—("Sinter--Theory").

Since the half-value period of radon is 3.825 days, it is believed that the source of the radon in mineral springs is not deep in the earth. It is generally accepted that the radon content of mineral springs is characterized by the last of the rocks through which the mineral water passed. Radioactive springs are usually found in regions of granite. The high radon content of spring A9 is, however, very difficult to explain merely by the fact that this spring issues from a granite region. As will be seen

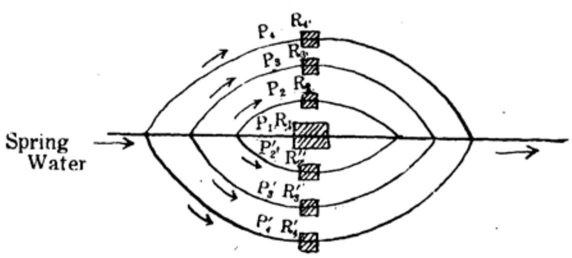

Fig. 28.

from Table 37 by Nakai,[31] the radium content of the rocks in Masutomi does not much exceed those of rocks from otherplaces in Japan. Since no radioactive minerals have yet been discovered in Masutomi-region, it is difficult to conclude that radioactive minerals near the ground surface of Masutomi are the source of radon in mineral springs.

Table 37. Radium Content of the Rocks in the Masutomi Region. (Determined by T. Nakai.)

Rocks	Location	Radium Content g./g. $\times 10^{12}$
Granite	Yuzawa	0.76
Metamorphic rock (from sandstone)	Wadegawara	0.71
Sand stone	Siokawa	0.56
Shale	Siokawa	0.76

Table 38. Radium Content of Sinter Deposits from Mineral Springs of Masutomi.

(Determined by T. Nakai.)

Spring	Radium Content of Deposit. g./g. $\times 10^{12}$
Tuganero-simo-no-yu	175
Higasiobi-no-izumi	19.2
Siokawara	18.4
Ginsento-huru-yu	9.6
Kuridaira No. 3	8.8

According to T. Nakai, ocherous deposits of mineral springs (usually called "Radium-fossil") contain appreciable amounts of radium, as is shown in Table 37. It was our opinion that these ocherous deposits, as

(31) T. Nakai, not yet published.

well as the soils containing it, and the weathering products of granites, are the source of radon in mineral springs, as will be clear from what follows:—

Three springs (A2, A6, and A9) will now be compared, and the relations between the radon content and other factors explained in detail. Spring A9 issues from a spot very close to the River Hontanigawa, spring A2 in the grounds of Hotel Tuganero, and spring A6 from a place slightly higher in level than that whence spring A9 issues. Except for their radon contents, the chemical composition of these three springs is very similar. The large differences in the radon contents of these three mineral springs may be explained by assuming that the ocherous sinters of mineral springs, accumulated since very remote times, cover the surface of the granite whence the mineral springs issue. Since these soils are usually porous,

Table 39. Radon Content of Mineral Springs at Kawarazaki.

(a) Springs A49. 3000–9000 Mache. (See page 39.)

(b) Kawarazaki-kami-no-yu Springs.

	Temperature (°C)	Rn Content (Mache)	Date
Spring No. 1	21.2	290	No. 91 day
Spring No. 2	16.0	400	" "
Spring No. 3	19.0	260	" "
Spring No. 4	21.0	290	" "
Spring No. 5	15.0	485	" "
Spring No. 6	18.5	480	" "
*Spring No. 7	22	275	" "
Spring No. 8	13	650	" "
Spring No. 9	15	905	" "
Spring No. 10	17	465	" "
Spring No. 11	—	17.7	Nov. 1942

* Amount of Flow 1.83 l./min. (No. 185 day).

(c) Kawarazaki-simo-no-yu Springs.

	Temperature (°C)	Rn Content (Mache)	Date
*Spring No. 1	8.8	19.9	No. 92 day
Spring No. 2	8.8	42.7	" "
**Spring No. 3	5	29.4	" "
Spring No. 4	5	43.7	" "
Spring No. 5	5	106	" "
***Spring No. 6	6	235	" "

* Amount of Flow 100–200 c.c./min.

** Amount of Flow 150 c.c./min.

*** Issues from sinter.

holding considerable amounts of radium, they are supposed to be very good accumulators and reservoirs of radon. The high radon content of spring A9 is probably due to the fact that this spring passes through a very extensive area of such sinter deposits, and the low radon content of spring A2 is probably owing to the fact that it issues almost directly from the granite.

Fig. 29 shows that the mineral springs of Group A along the line AB contain the more radon the lower the level whence they issue.

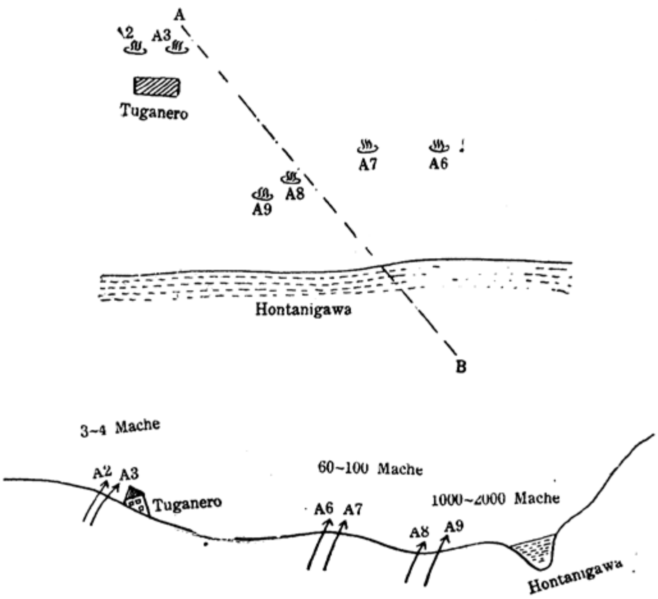

Fig. 29.

Fig. 30 shows that mineral springs of Kawarazaki along the line AB contain the more radon the higher the level whence they issue (Table 39).

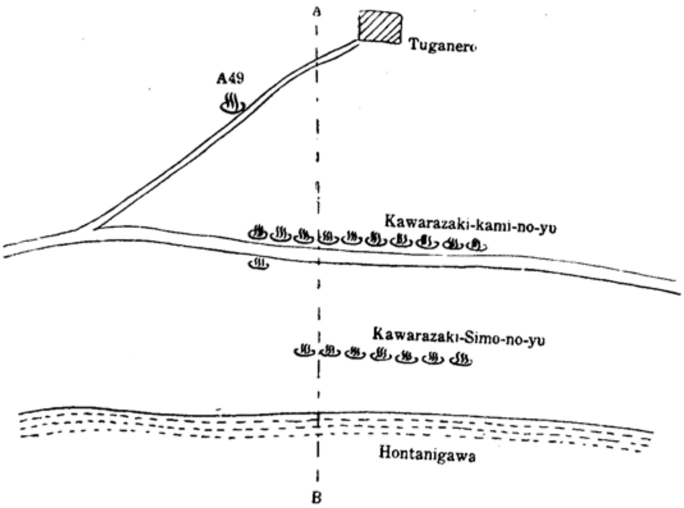

Fig. 30 A.

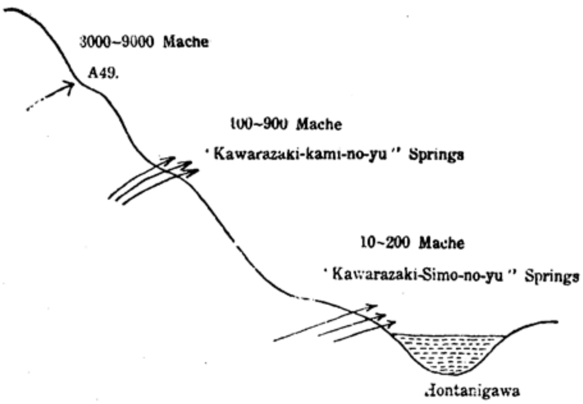

Fig. 30 B.

A Trial of Mathematical Consideration.

The following calculations show that the very large amount of radon in spring A9 (and A8) could be supplied from the radium that had been brought to the surface of the earth by springs A2, 3, etc.

The amount of radium brought to the surface of the earth by the mineral springs in time dt may be given by the equation,

$$dN_1 = K_1 dt \qquad (1)$$

where K_1 is the amount of radium brought to the surface of the earth in unit time.

The radium decays, following the well-known equation,

$$dN_2 = -\lambda N dt \qquad (2)$$

where λ=decay constant of radium

N=the amount of radium, brought to the surface of the earth by the mineral springs and accumulated in this region.

The amount of radium that is taken away in time dt from this region, for example by the rivers, is given by the equation,

$$dN_3 = -K_3 dt \qquad (3)$$

where K_3 is the amount of radium that is removed in unit time by the river.

The increase of radium in this region is, therefore, given by the equation,

$$dN = dN_1 + dN_2 + dN_3$$
$$= K_1 dt - \lambda N dt - K_3 dt \qquad (4)$$

Solving this differential equation, assuming that when t=0, N=0, we get the general equation,

$$N = \frac{K_1 - K_3}{\lambda}(1 - e^{-\lambda t}) \qquad (5)$$

Assuming that $K_1 \gg K_3$

$$N = \frac{K_1}{\lambda}(1-e^{-\lambda t}) \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (6)$$

when $t \to \infty$,

$$N \to \frac{K_1}{\lambda} \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (7)$$

then the amount of radium brought to the surface of the earth, by spring A2 for example, is estimated to be about 5×10^{-12} g. Ra per second. Therefore if this value is given to K_1, and as

$$\lambda = 1.39 \times 10^{-10} \text{sec.}^{-1} \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (8)$$

we get

$$N = 0.28 \text{ g.} \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (8)$$

(8) shows that if spring A2 had been issuing constantly during a very long period of time, and that the radium thus brought to the surface of the earth had not been removed from this region, there ought to be accumulated in the region near spring A2 about 0.28 g. of radium.

The amount of radon formed from 0.28 g. of radium in one year can then be calculated by the equation

$$Q = Q_0 e^{-\lambda t} \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (9)$$

where $\lambda = 1.39 \times 10^{-10}$ sec.$^{-1}$

$$t = 3.16 \times 10 \text{ sec.} \quad (= 1 \text{ year})$$

$$(Q_0 - Q)_{1\,\text{year}} = 0.0026 \text{ g. Rn}$$

As 1 Curie = 6.5×10^{-6} g.

$$\frac{0.0026}{6.5 \times 10^{-6}} = 400 \text{ Curie/year} \quad \ldots\ldots\ldots\ldots \quad (10)$$

The foregoing calculation shows that about 400 Curie of radon is formed every year. If the total amount of flow is assumed to be 1 litre per minute and their average radon content 5000×10^{-10} Curie per litre, then the radon discharge of springs A8 and A9 is calculated to be about 0.26 Curie every year.

Therefore only 0.26 Curie out of 400 Curie of the radon is consumed annually in spring A8 and A9. This is only 0.065% of 400 Curie.* The remaining 99.935% is not consumed in springs A8 and A9, but either escapes into the air or decays while it remains in the rocks or soils.

Summary.

(1) A number of strongly radioactive springs were discovered in Masutomi, Yamanasi Prefecture.

* If it is assumed that this amount of radon (400 Curie) were consumed without loss every year to supply the radon to springs A8 and A9, a radioactive spring having a radon content of 100/0.065 times higher than that of springs A8 and A9, namely, $5000 \times 10^{-10} \times 100/0.065$ = about 7700000×10^{-10} Curie will be obtained.

(2) The radon content of a number of very small springs near spring A8 and A9 at Higurasi-no-huti was measured. On No. 88 day, spring A84 showed a content of 7130 Mache. On the same day the radon content of spring A9 (when the water sample was drawn with an injector from the inner part of the exit of the spring) registered 6870 Mache.

(3) A strongly radioactive spring was discovered in Masutomi on No. 155 day, the radon content of which (spring A49, Dairokuten-no-izumi) is supposed to be always 3000 to 4000 Mache, although values higher than 5000 Mache were obtained at the time the sample was drawn, a small injector used.

(4) The radon, chlorine, carbon dioxide contents, the amount of the flow and the water temperature, of spring A2 were measured daily during the period between No. 74 day and No. 89 day, and the effect of snow on the radon content, together with the chemical composition of the mineral springs was studied. The variation in the radon content of this spring is explained by assuming that the original mineral spring was diluted by thaw water.

(5) The radon content of spring A9 was measured daily during the period between No. 1 day and No. 98 day.

(6) The radon content of spring A9 fluctuated between 1030 and 1900 Mache during this period.

(7) The radon content increases after snow or rain.

(8) The higher the water temperature the higher the radon content.

(9) The variation in the chlorine, bicarbonate, and the carbon dioxide contents were also measured daily during this period.

(10) On No. 136 day, the radon content of spring A9 registered 12000 Mache.

(11) In winter, on very cold and clear days after snow, the radon content is very low in the morning, increases in the day-time with the thawing of the frozen ground, and suddenly diminishes in the evening when the ground freezes again.

(12) The radon content of frozen mineral water was measured.

(13) [Rn/CO_2] ratio in the mineral springs of Masutomi was discussed.

(14) It was very difficult to ascertain any marked relation between the variations in the radon content of the mineral springs of Masutomi and the small earthquakes that occurred in Yamanasi Prefecture, during the period of this investigatoin.

(15) The total analyses of spring A9 and spring A49 were intended.

(16) A general survey of our investigation into the minute constituents of the mineral springs of Masutomi was made.

(17) The chemical composition of the sinter of spring A6 is described.

12. Conclusion.

The author, after repeatedly measuring the radioactivity of one of the most radioactive mineral springs in the world, attempted in this paper to clarify the mechanism of formation of strongly radioactive springs, and obtained a number of geochemically interesting results, thus enabling a theoretical discussion on this problem.

To some readers the discussions proposed in this paper may appear rather speculative and very qualitative, but it is hoped that they may contain some germs of what might in the future be developed into a more perfect general theory of strongly radioactive springs. The author hopes to resume the discussion when more sufficient data have accumulated.

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