Radium Content of Mineral Springs in Japan.

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Contents.

I. Introduction. Since K. Manabe and H. Isitani first undertook, in 1909, to determine the presence of radioactivity in the mineral springs of Japan, a number of reports have been made by R. Ishizu(1), Y. Kinugasa and other investigators, but no determination of the radium content in the mineral springs of Japan has yet been made. To do so is considered important for the following reasons:

First, it will help in solving certain problems in geochemistry. The quantitative analysis of radium in natural waters is indispensable for investigating the distribution of this element in the hydrosphere. Besides, studies of the rare elements, such as that of radium in spring waters, often play an important role in ascertaining their origin and history. It is also very important to know the conditions under which radium migrates when the spring waters act on the rocks through which they pass or when sinter deposits are precipitated from them.

Second, it is helpful in our search for the natural sources of this rare element. It has been reported(2) that the quantity of radium brought up by a group of springs in a single district of North Caucasus, Russia, often amounts to more than one gram per year. Today the supply of radium depends entirely upon radium ores, such as pitchblende, carnotite, etc. Much to our disappointment, these radium ores are very scarce in Japan. If, therefore, a natural water of extraordinarily high radium content is found and a technique for separating the radium from such natural waters without having to use a large quantity of reagents could be developed, it may turn out to a valuable source of radium.

Third, there is the problem of hot-spring therapeutics. Studies in rodioactivity have shown that the activity from mineral spring is one of the most effective of therapeutic agents. It is of course known that some of the physiological reactions of spring waters are believed to be due to the presence of very small quantities of the heavy metals, such as copper, arsenic, etc. According to T. Misawa, the mineral springs of Kinkei, Totigi Prefecture, have proved very efficaceous in anaemia, because they contain a very small quantity of copper in addition to iron. Thus the minor constituents of mineral springs, although their quantities are very small, cannot be ignored. In this respect, the radium of mineral springs may have also some therapeutic effects.

Samples of water for the radium determination were collected from various parts of the country. Those of the waters from the mineral springs of Kasio and Tadati in Nagano Prefecture; Ena, Naegi and Kasagi in Gihu Prefecture; Masutomi in Yamanasi Prefecture; Ikeda in Simane Prefecture; and of those from the hot springs of Misasa, Sekigane, Hamamura and Katimi in Tottori Prefecture; and Sigaku, Koyabara and

(1) R. Ishizu, "The Mineral Springs of Japan," (1915).
(2) I. D. Kurbatov, *J. Phys. Chem.*, **38** (1934), 521.

Yugakai in Simane Prefecture were obtained by the writer. The waters from the mineral springs of Arima in Hyōgo Prefecture, were sampled by Y. Nemoto of the Tokyo Imperial Industrial Research Institute and K. Yamasaki of the Chemical Institute of the Tokyo Imperial University, and that from the hot springs of Tuta in Aomori Prefecture; Matunoyama in Niigata Prefecture; Sionoha and Yosino in Nara Prefecture; and those of Katuura, Yukawa, Yuzaki and Higasi-sirahama in Wakayama Prefecture, were collected by K. Kuroda of the Chemical Institute of the Tokyo Imperial University. The brines from the oil fields in Akita and Niigata Prefectures and that from the natural gas field in Tiba Prefecture were collected by K. Hosino of the Tokyo Imperial Industrial Research Institute. Through the courtesy of the Hot Spring Society of Japan, all the other samples of water were collected by the Hot Spring Owners' Association at each hot spring, for all of which courtesies the writer tenders his warmest thanks.

The samples of water for radium determination were usually drawn direct from the source at each spring. If the orifice of a spring happened to be at the bottom of a bath-tub, samples were drawn after bailing out the water, if that was possible. At the mineral sprirngs of Wadegawara No. 2 in Masutomi, Yamanasi Prefecture, the writer measured the radon content of the water taken from the surface and that direct from the source of the spring. The result in the former case was 1133 Mache's units, whereas that of the latter was 1343 Mache's units. As there may be also some difference in the radium content of water drawn from the source of a spring and that of the surface water, although not so marked as in the case of the radon content, special precautions are necessary in sampling.

For sampling the water, a large bottle of about 2 liters capacity is completely filled with the water, except for the few c.c. of air at the neck to allow for temperature changes. The samples were sent to the Tokyo Imperial Industrial Research Institute, where their radium contents were determined.

II. Methods of Analysis. *The principle of the method.* For determining the radium in mineral waters, the so-called "emanation method" was adopted. The principle of this method is to compute the radium concentration in the sample of water by comparing the radioactivity of the radon that is expelled from the water sample (the radon in it being in equilibrium with the radium) with that produced by the radon from a known amount of radium.

The sample of water, after the sediments in it have been completely dissolved, is collected in a flask, sealed, and left to stand for twenty-eight days and more. When equilibrium is practically established between the radium in the specimen and its radon, all the radon present in it are swept into a previously evacuated ionization chamber by means of the vacuum-boiling method. Its radioactivity is then measured when it becomes maximum, which state is attained three hours after the admittance of radon.

Apparatus. The essential parts of the apparatus are an emanation electroscope and a radon-receiver. A reliable emanation electroscope, made by the Institute of Physical and Chemical Research, Tokyo, was used. The arrangement is shown in Fig. 1.

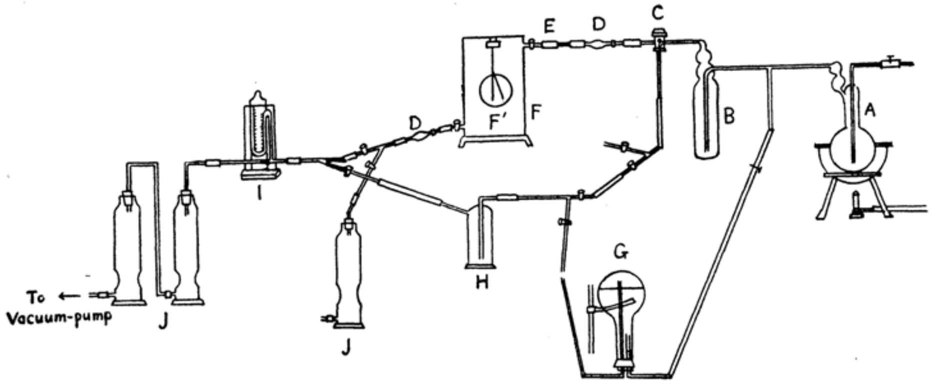


Fig. 1. Arrangement for Emanation Method.

A, which is a preserving flask of about 600 c.c. capacity (curie bottle) containing the solution to be examined, has two side-tubes drawn to a thin end. One of them is connected to the radon-receiver B (see figure), which has a capacity of about 400~500 c.c. and serves as a temporary receiver for the air containing the emanations and the vapour. Thoron disintegrates almost completely during its suspension in B, while radon survives, owing to its longer life, until its activity is measured. On being transferred to the ionization chamber F, the gases, including radon, are passed through a tail stopcock C, a calcium chloride drying tube D, and a capillary tube E. F is a cylindrical brass vessel of about 1500 c.c. capacity, which contains a gold-leaf F′. The reading is done by means of a low power microscope provided with a microscale in its image plane, across which the gold-leaf is observed to move.

G is a flask containing the distilled water that is poured into the radon-receiver B in order to drive the radon in it into the electroscope chamber.

H is a wash bottle, I a manomenter, and J a gas-drying apparatus.

Natural leak. Before each determination, the natural leak of the electroscope, which varies from time to time, must be measured, for which purpose the ionization chamber is evacuated and fresh air introduced through the dryers. As soon as there is atmospheric pressure in the ionization chamber, readings are made and continued at intervals of 15 to 20 minutes for 1 to 3 hours until the leak becomes practically constant.

Since variation in the natural leak interferes with the accuracy of determination, whatever the cause, it is desirable to maintain the laboratory at a state as constant as possible. Should the air that is introduced into the ionization chamber contain even the slightest trace of moisture, the natural leak is liable to be affected to a very marked degree. Special precautions are, therefore, necessary for properly controlling the drying apparatus.

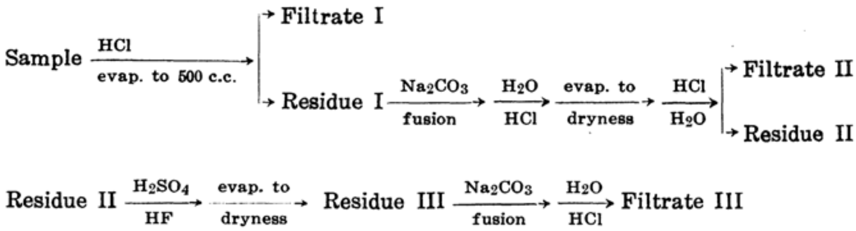
Determination of radium in mineral water. The first method. For determining the radium, about 2 liters of water are used for each sample. After adding a few c.c. of hydrochloric acid, it is evaporated to 500 c.c. on a water bath. Any insoluble residue is collected on the filter paper and the filtrate sealed into the curie bottle, leaving a space of about 100 c.c. for subsequent boiling. The sample sealed, is then allowed to stand for twenty-eight days or more, during which time equilibrium is practically reestablished between the radium in the specimen and its radon.

The solution in the curie bottle is vigorously boiled and the free radon that accumulates in the bottle is swept into a previously evacuated radon receiver and then introduced through the drying tube and the capillary tube into a previously evacuated ionization chamber. When all the air containing radon is expelled from the receiver, air for washing out is introduced through the drying tube into the ionization chamber to sweep away the radon remaining in C, D and E into the ionization chamber until there is an atmospheric pressure in the chamber. The reading is taken when the rate of fall of the leaf becomes maximum, which state is attained three hours after the admittance of radon. The rate of fall of the gold-leaf across the scale is expressed in terms of one scale division per minute. From this must be subtracted the natural leak, determined immediately before the determination, the figure thus obtained corresponding to the radioactivity from the solution in the curie bottle. By comparing the radioactivity produced by the radon with a known amount of radium, the radium content of the solution is readily computed. The standard solution used in the present investigation was prepared in the laboratory of S. Iimori, at the Institute of Physical and Chemical Research, Tokyo.

After completion of the determination, the ionization chamber is exhausted and dried air again introduced. By repeating this process several times, the ionization chamber is completely washed. The next determination is made after the radioactive deposits that have formed in it have completely decayed away.

The insoluble residue on the filter paper mentioned above is converted into solution by the following method. It is ignited and fused with anhydrous sodium carbonate. The fused mass is treated first with distilled water and then with hydrochloric acid. The solution is evaporated to dryness to render the silica insoluble, the salts thus obtained being moistened with concentrated hydrochloric acid and dissolved in water. The insoluble residue, mostly silica, is ignited and treated with a mixture of sulphuric and hydrofluoric acids. If a residue still remains, the operartion just described must be repeated until no residue can be detected. The solution thus obtained is placed into a curie bottle and sealed and its radium content determined by the method already described. The sum of this result and that obtained from Filtrate I is the radium content of the original specimen. The amount of radium absorbed by Residue I is usually negligible.

The maximum observational uncertainty in the radium determination by this method is $0.01\sim0.02\times10^{-12}$g. Ra per liter of water when 2 liters of sample of water are used.



The second method. With some water samples, the following method was adopted. About 2 liters of the water preserved in the bottle is filtered. 500 c.c. of the filtrate is taken and put into a curie bottle and sealed. The total residue, together with that remaining in the bottle, is dissolved in hydrochloric acid. The insoluble residue in hydrochloric acid is dissolved by the method described in the preceding section. The solutions thus obtained are separately put into curie bottles and sealed. The radium contents of the solution in each curie bottle are determined. The sum of these results, each result being calculated to that of 1 liter, is the radium content of the original specimen.

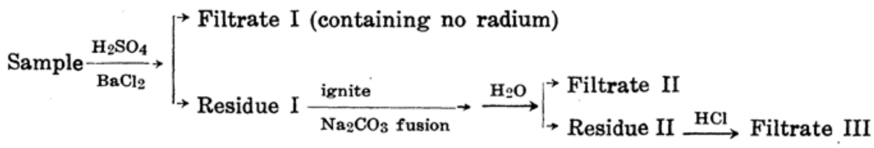
As will be seen from Table 1, the amount of radium adsorbed in the process of precipitation, deposited from the water after sampling, differs greatly with the spring water, in some of which this value is very large, so that they cannot be ignored.

Table 1.

Spring	Location	Prefecture	Radium content $g./l. \times 10^{12}$	
			in water	in sediment
Monkawa-Onsen	Monkawa	Kanagawa	0.41	0.00
Yuzawa No. 1	Masutomi	Yamanasi	71.74	0.07
Toyotomi-Onsen	Toyotomi	Hokkaidō	9.62	0.48
Totikubo No. 1	Masutomi	Yamanasi	16.60	16.02
Kaimonzi-Kōen-Onsen	Beppu	Ōita	0.13	0.12
Tuganerō No. 1	Masutomi	Yamanasi	14.57	49.96
Makiyama-Kōsen	Makiyama	Mie	0.17	1.63
Matudaira-no-yu	Masutomi	Yamanasi	0.00	1.52

The third method. To the water sample a few c.c. of sulphuric acid are added, followed by a solution of barium chloride (about 0.5 g.). Radium is precipitated from the water as radium sulphate, along with barium sulphate. The quantity of radium contained in the water of mineral springs is so small that the solubility product is not attained despite the high sulphate ion concentration, but by adding barium chloride, radium is precipitated along with barium sulphate in the form of mixed crystals.

The precipitated sulphate, after it is filtered off, is ignited and fused with anhydrous sodium carbonate. The fused mass is leached with distilled water in order to dissolve the sodium sulphate. The insoluble residues of barium carbonate and radium carbonate are dissolved in hydrochloric acid. The two solutions thus obtained, the alkaline solution of sodium sulphate (Filtrate II) and the acidic solution of barium and radium chlorides (Filtrate III) are separately put into curie bottles and sealed and their radium contents determined.



As will be seen from Table 2, the results obtained by this method agree fairly well with those obtained by the first method. Since this method, however, is more troublesome than the other two, in the present experiments only the first and second methods were used, almost all the samples being determined by the first method.

Table 2. Comparison of the two methods of determination.

Method	Sample	Radium content g./l. $\times 10^{12}$
First	Yakendo-Kōsen, Arima,	61.57
Third	,,	61.50
First	Hon-Onsen, Arima	64.97
Third	,,	64.88
First	Netu-no-yu, Matunoyama	6.43
Third	,,	6.44

Determination of radium in sediments from mineral springs. About 10 g. of the sample is dissolved in hydrochloric acid. The insoluble residue is ignited and then fused with anhydrous sodium carbonate. The fused mass is then dissolved by the method described in a previous section. The solutions thus obtained are separately placed in curie bottles and sealed and their radium contents determined.

III. Results of Determinations. *Results of Determinations.* 477 water samples drawn from various mineral springs in Japan and 15 brine samples from the oil fields in Akita and Niigata Prefectures and from the natural gas field in Tiba Prefecture, were determined for radium content, with results shown in Table 3. The orifice temperatures of these spring waters as measured at time of sampling are also given for reference in the table.

Geographical distribution of radium content. The geographical distribution of the radium content of Japanese springs is shown on the accompanying map, Fig. 2, the radium content of each grorup of springs

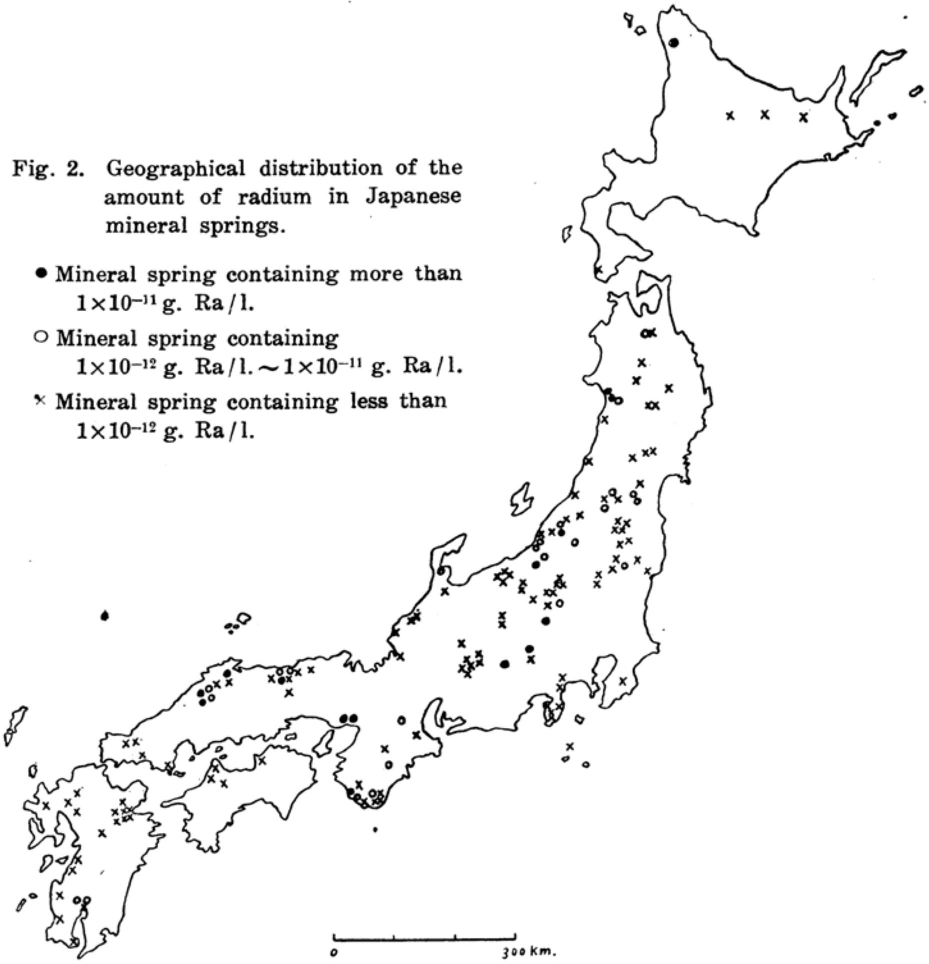


Fig. 2. Geographical distribution of the amount of radium in Japanese mineral springs.

● Mineral spring containing more than 1×10^{-11} g. Ra/l.

○ Mineral spring containing 1×10^{-12} g. Ra/l. $\sim 1\times10^{-11}$ g. Ra/l.

× Mineral spring containing less than 1×10^{-12} g. Ra/l.

being represented by their highest values, from which it will be seen that the mineral springs of high radium concentration are usually localized —an interesting fact.

Comparison of the radium content of mineral waters in Japan with those in other countries. The radium contents of mineral waters in Japan will be compared with those in other countries.

The largest amount of radium in the water of mineral wells and springs previously known was discovered by B. A. Nikitin[(3)] in the salt water from Grosny petroleum district, North Caucasus, Russia. It amounts to 1.83×10^{-8} g. of radium per liter of water. In the Far East, S. Gōda recently found the extraordinary high concentration of radium in salt water from Tzeliutsing, Szechwan, China. It amounts to 7.26×10^{-10} g. of radium per liter of water. In Table 4 are given for comparison the highest concentrations of radium in the world's mineral springs, and in Table 5 for those in Japan.

(3) W. Chlopin and B. Nikitin, *Compt. rend. acad. Sci.*, U.R.S.S., A **1930**, 393.

Table 3. Radium content of mineral waters in Japan.

1. Radium content of the mineral springs of Japan.

No.	Spring		Section	Village	County	Prefecture	Radium content g./l.×10^12	Temp. of spring °C	Date of sampling
1	Katura-no-yu	Sōunkyō	Sōunkyō	Kamikawa	Kamikawa	Hokkaidō	0.12		Aug. 1937
2	Reikō-no-yu (Onna-yu)	,,	,,	,,	,,	,,	0.08		,,
3	,, (Otoko-yu) A	,,	,,	,,	,,	,,	0.08		,,
4	,, (,,) B	,,	,,	,,	,,	,,	0.10		,,
5	Hana-no-yu	,,	,,	,,	,,	,,	0.12		,,
6	Kawayu-Onsen	Kawayu	Kawayu	Tesikaga	Kawakami	,,	0.00		
7	Ponyu-Onsen	Ponyu	Ponyu	Aiuti	Tokoro	,,	0.00		Aug. 1937
8	Toyotomi-Onsen	Toyotomi	Kami-sarobetu, Saru	Horonobe	Tesio	,,	10.10		,,
9	Kozima-Kōsen No. 1	Kozima	Kozima	Nebuta	Matumae	,,	0.00		July 1937
10	,, No. 2	,,	,,	,,	,,	,,	0.00		,,
11	Netu-no-yu	Sukayu	Sukayu-sawa, Arakawa	Arakawa	Higasi-tugaru	Aomori	0.32		Aug. 1937
12	Hie-no-yu	,,	,,	,,	,,	,,	0.15		,,
13	Sika-no-yu	,,	,,	,,	,,	,,	0.17		,,
14	Sibu-rokubu-no-yu	,,	,,	,,	,,	,,	0.00		,,
15	Sin-yu	Sinyu	Arakawa	,,	,,	,,	0.05		,,
16	Tetu-Kōsen No. 1	,,	,,	,,	,,	,,	1.16		,,
17	,, No. 2	,,	,,	,,	,,	,,	0.63		,,
18	Megusuri-no-yu	,,	,,	,,	,,	,,	0.07		,,
19	Tuta-Onsen	Tuta		Towada	Kamikita	,,	0.24	46.0	Aug. 1939
20	Nanasigure-Kōsen	Nanasigure	Nanasigure	Terada	Iwate	Iwate	0.60		
21	Hanamaki-Onsen	Hanamaki	Itini-tiwari	Yumoto	Hienuki	,,	0.00	72.0	Aug. 1937
22	Usi-no-yu (Ō-yu)	Yumoto	Yumoto	Yuta	Waga	,,	0.08		
23	Taki-no-yu	,,	,,	,,	,,	,,	0.05		

Table 3.—(*Continued*)

1. Radium content of the mineral springs of Japan.—(*Continued*)

No.	Spring		Section	Village	County	Prefecture	Radium content g./l. $\times 10^{12}$	Temp. of spring °C	Date of sampling
24	Ōkama-no-yu	Tamagawa	Sibukuro-zawa, Tamagawa	Tazawa	Senhoku	Akita	0.60		Nov. 1937
25	Yuze-Onsen	Yuze	Yubata	Miyakawa	Katuno	,,	0.00		
26	Onogawa-Onsen	Onogawa	Onogawa	Misawa	Minami-okitama	Yamagata	8.37	76.0	Aug. 1939
27	Kyokuato-gensen	Akayu	Akayu	Akayu Town	Higasi-okitama	,,	0.09	62.0	,,
28	Matusima-kwan-no-yu	,,	,,	,,	,,	,,	0.09	54.0	,,
29	Turuzune-Onsen	Kaminoyama		Kaminoyama Town	Minami-murayama	,,	1.07		July 1939
30	Sin-moto-yu	,,		,,	,,	,,	0.27		,,
31	Tukioka-hotel No. 1	,,		,,	,,	,,	1.58		,,
32	,, No. 2	,,		,,	,,	,,	1.07		,,
33	Kawara-no-yu	Takayu	Takayu	Hotta	,,	,,	0.23	42.0	Aug. 1939
34	Gensen	Akakura	Akakura, Tomizawa	Higasi-Oguni	Mogami	,,	0.12	65.0	Aug. 1937
35	Gensen No. 1	Sin-akakura	,,	,,	,,	,,	0.10	65.0	,,
36	,, No. 2	,,	,,	,,	,,	,,	0.15	67.0	,,
37	,, No. 3	,,	,,	,,	,,	,,	0.17	67.0	,,
38	Kyōdō-yokuzyō-no-yu	Atumi	Atumi	Atumi	Nisi-tagawa	,,	0.58		Jan. 1938
39	Kawanaka-no-yu	,,	,,	,,	,,	,,	0.87		,,
40	Kamasaki-no-yu	Kamasaki	Kamasaki, Kuramoto	Hukuoka	Katta	Miyagi	1.00		Aug. 1937
41	Mogamiya-no-yu	,,	,,	,,	,,	,,	1.37		,,
42	Kimuraya-no-kyū-yu	,,	,,	,,	,,	,,	3.03		,,
43	Kimuraya-no-sin-yu	,,	,,	,,	,,	,,	2.58		,,
44	Ō-yu	Aone	Aone	Kawasaki	Sibata	,,	0.55		,,
45	Nagō-no-yu	,,	,,	,,	,,	,,	1.39		,,
46	Sin-yu	,,	Nomadaira	,,	,,	,,	0.80		,,

47	Kawara-no-yu	Sakunami	Sakunami	Hirose	Miyagi	Miyagi	0.21	54.0	Aug. 1939
48	Takaku-ryokwan-no-yu	Kawatabi	Kawatabi, Ōguti	Kawatabi	Tamatukuri	,,	0.26		
49	Takatō-ryokwan-no-yu	,,	,,	,,	,,	,,	0.08		
50	Huzisima-ryokwan-no-yu	,,	,,	,,	,,	,,	0.07		
51	Itagaki-ryokwan-no-yu	,,	,,	,,	,,	,,	0.00		
52	Private bath (A. Yosida)	,,	,,	,,	,,	,,	0.00		
53	Tansan-sen	Naruko		Naruko Town	,,	,,	0.06		Aug. 1937
54	Tanaka-ryokwan No. 1	,,		,,	,,	,,	0.23	79.0	Aug. 1939
55	Moto-yu	Kasi	Teradaira, Mafune	Nisigō	Nisi-sirakawa	Hukusima	0.53		
56	Tengu-no-yu	,,	,,	,,	,,	,,	0.41		
57	Yuzin-no-yu	,,	,,	,,	,,	,,	0.75		
58	Mamegara-hudō-no-yu	Sirakawa	Sekikawakubo	Sirakawa Town	,,	,,	1.27		Oct. 1937
59	Moto-yu No. 1	Bobata	Hida, Bobata	Bobata	Isikawa	,,	0.11		Aug. 1937
60	,, No. 2	,,	,,	,,	,,	,,	0.09		,,
61	Naka-no-yu	Takano	Takano	Minowa	Iwaki	,,	0.00		Nov. 1937
62	Moto-yu	Iwasiro-atami	Takatama	Takakawa	Adati	,,	0.00		
63	Itiriki-hotel-no-yu	,,	,,	,,	,,	,,	0.00		
64	Kamiya-ryokwan-no-yu	,,	,,	,,	,,	,,	0.08		
65	Sin-yu	,,	,,	,,	,,	,,	0.00		
66	Private bath (K. Hasimoto)	,,	,,	,,	,,	,,	0.05		
67	,, (T. Matumoto)	,,	,,	,,	,,	,,	0.00		
68	,, (M. Ōgosi)	,,	,,	,,	,,	,,	0.08		
69	Yosino-ya-no-yu	,,	,,	,,	,,	,,	0.02		
70	Takatama-Onsen	Takatama	Akogasima	Marumori	Asaka	,,	0.02	42.5	-
71	Sin-nozi-Onsen	Sin-nozi	Wasikurayama	Tutiyu	Sinobu	,,	0.00		Aug. 1937
72	Spring No. 1	Wasikura	,,	,,	,,	,,	0.00	50.0	,,
73	,, No. 2	,,	,,	,,	,,	,,	0.00	94.0	,,
74	,, No. 3	,,	,,	,,	,,	,,	0.07	48.0	,,
75	Numaziri-Onsen	Numaziri	Numaziriyama	Azuma	Yama	,,	0.84		

Table 3.—(*Continued*)

1. Radium content of the mineral springs of Japan. (*Continued*)

No.	Spring		Section	Village	County	Prefecture	Radium content g./l. $\times 10^{12}$	Temp. of spring °C	Date of sampling
76	Naka-no-yu	Yokomuki	Wakamiya	Azuma	Yama	Hukusima	0.00	24.0	
77	Iwasakiya-no-yu	Ōsio	Ōsio	Ōtaki	Ōnuma	,,	3.72		
78	Kyōdō-no-yu	,,	,,	,,	,,	,,	4.26		
79	Moto-yu	Kawazi	Kawazi	Huziwara Town	Sioya	Totigi	0.10		Aug. 1937
80	Hudō-no-yu	,,	,,	,,	,,	,,	0.61		,,
81	Iwa-no-yu	,,	,,	,,	,,	,,	0.07		,,
82	Yakusi-no-yu	,,	Asamayama, Kawazi	,,	,,	,,	0.00		,,
83	Komoti-no-yu	,,	,,	,,	,,	,,	0.10		,,
84	Kinkei-Kōsen	Kinkei		Huziwara	,,	,,	0.00		
85	Kakkō-Onsen	Nasu	Nasudake, Yumoto	Nasu	Nasu	,,	0.00		Aug. 1937
86	Sika-no-yu	,,	Huruyasiki, ,,	,,	,,	,,	0.55		,,
87	No. 1 Nomi-yu	Ikao	Ikao	Ikao Town	Gumma	Gumma	0.00		,,
88	No. 2	,,	,,	,,	,,	,,	0.00		,,
89	No. 3	,,	,,	,,	,,	,,	0.06		,,
90	No. 4 Ohaguro-no-yu	,,	,,	,,	,,	,,	0.09		,,
91	No. 5 Hukiage-no-yu	,,	,,	,,	,,	,,	0.00		,,
92	No. 6 Ōseki-no-yu	,,	,,	,,	,,	,,	0.00		,,
93	Wasi-no-yu	Kusatu	Takisita, Kusatu	Kusatu Town	Agatuma	,,	0.05		,,
94	Tiyo-no-yu	,,	,,	,,	,,	,,	0.10		,,
95	Nikawa-no-yu	,,	,,	,,	,,	,,	0.03		,,
96	Zizō-no-yu	,,	Zizō, Kusatu	,,	,,	,,	0.01		,,
97	Netu-no-yu	,,	Nisi-mati, ,,	,,	,,	,,	0.15		,,
98	Sirahata-no-yu	,,	,, ,,	,,	,,	,,	0.16		,,

99	Seki-no-yu	Kusatu	Nisi-mati, Kusatu	Kusatu Town	Agatuma	Gumma	0.10		Aug. 1937
100	Matu-no-yu	,,	Higasi-mati, ,,	,,	,,	,,	0.00		,,
101	Nagi-no-yu	,,	Sensui, ,,	,,	,,	,,	0.03		,,
102	Alkali-sen	Isobe	Sionokubo, Nisi-kami-isobe	Isobe Town	Usui	,,	15.53		,,
103	Tansan-sen	,,	,,	,,	,,	,,	11.59		,,
104	Yuhara-Onsen	Yuhara	Suwahara, Yuhara	Minakami	Tone	,,	0.09		,,
105	Minakami-Onsen	Minakami	Ōmidō, Ōana	,,	,,	,,	0.49		,,
106	Tanikawa-Onsen	Tanikawa	Tanikawa	,,	,,	,,	0.10	62.0	,,
107	Yubiso-no-yu No. 1	Yubiso	Yusima, Yubiso	,,	,,	,,	0.00	50.0	,,
108	,, No. 2	,,	Yunosawa, ,,	,,	,,	,,	0.30	66.0	,,
109	Asahi-no-yu	Hōsi	Hōsi, Nagai	Niiharu	,,	,,	0.00	44.0	,,
110	Kotobuki-no-yu	,,	,,	,,	,,	,,	0.00	42.5	,,
111	Taki-no-yu	,,	,,	,,	,,	,,	0.31	43.5	,,
112	Sarugakyō-Onsen	Yusima	Yusima, Sarugakyō	,,	,,	,,	0.55		,,
113	Zizō-Kōsen	Akagi			Seta	,,	1.12		Oct. 1937
114	Nii-sima-Onsen	Setoyama	Setoyama	Hon	Nii-sima	Tokyo	0.08		Aug. 1937
115	Sikine-sima-Onsen	Sikine-sima	Sikine-sima	,,	,,	,,	0.08		,,
116	Asituke-tennen-Onsen	,,	,,	,,	,,	,,	0.06		,,
117	Tinata-tennen-Onsen	,,	,,	,,	,,	,,	0.10		,,
118	Huziya-hotel-no-yu	Sokokura	Sokokura	Onsen	Asigara-simo	Kanagawa	0.01		
119	Private bath (S. Hukuzumi	Yumoto	Yumoto	Yumoto Town	,,	,,	0.00		
120	,, (S. Tanaka)	,,	Tyaya-simogawara	,,	,,	,,	0.00		
121	,, (K. Ōkura)	,,	Sazawa-no-sawa	,,	,,	,,	0.00		
122	,, (S. Nakagawa	Setoyama	Setoyama	Miyagino	,,	,,	0.02		
123	Ubako-no-yu	Ubako	Ubako	Moto-hakone	,,	,,	0.05		Sept. 1938
124	Yoemon-no-yu	Yunohana-zawa	Yunohana-zawa	,,	,,	,,	0.17		
125	Mamane-no-y ı	Yugawara		Yugawara Town	,,	,,	0.00		
126	Simo-no-yu	,,		,,	,,	,,	0.10		
127	Hirokawara-no-yu	,,		,,	,,	,,	0.05		

Table 3.—(*Continued*)

1. Radium content of the mineral springs of Japan. (*Continued*)

No.	Spring		Section	Village	County	Prefecture	Radium content g./l. × 10^{12}	Temp. of spring °C	Date of sampling
128	Moti-no-yu	Yugawara		Yugawara Town	Asigara-simo	Kanagawa	0.28		
129	Monkawa-Onsen	Monkawa	Monkawa	,,	,,	,,	0.41		Sept. 1937
130	Senami-Onsen	Senami	Hamasinden	Senami Town	Iwahune	Niigata	0.11		,,
131	Spring No. 1	Murasugi	Murasugi	Sasaoka	Kita-kambara	,,	0.51		,,
132	,, No. 2	,,	,,	,,	,,	,,	0.04		,,
133	,, No. 3	,,	,,	,,	,,	,,	0.04		,,
134	Netu-no-yu	Matunoyama	Yumoto	Matunoyama	Higasi-kubiki	,,	6.43	68.0	Aug. 1939
135	Hii-no-yu	,,	,,	,,	,,	,,	6.49	83.0	,,
136	Kagami-no-yu	,,	,,	,,	,,	,,	14.30	72.5	,,
137	Usagiguti-Onsen	,,	,,	,,	,,	,,	6.17	36.8	,,
138	Tamago-no-yu	,,	,,	,,	,,	,,	0.29	17.0	,,
139	Komeya-no-yu	,,	,,	,,	,,	,,	2.62		,,
140	Yumoto-Onsen	,,	,,	,,	,,	,,	7.16		Apr. 1938
141	Myōkō-Onsen	Myōkō	Sekikawa	Nakayama	Naka-kubiki	,,	0.24	60.0	Aug. 1939
142	Akakura-Onsen	Akakura	Akakura	,,	,,	,,	0.02		,,
143	Moto-yu	Seki	Seki	Sekiyama	,,	,,	0.09		,,
144	Iwō-no-yu	Tubame	Tubame	,,	,,	,,	0.00		Aug. 1937
145	Moto-yu	,,	,,	,,	,,	,,	0.00		,,
146	Nagae-Kōsen	Nagae	Kōno, Tyōkeizi	Takaoka City		Toyama	0.42		,,
147	Gotairan-no-yu	Wakura	Wakura	Wakura Town	Kasima	Isikawa	6.04		,,
148	Moto-yu	,,	,,	,,	,,	,,	5.89		,,
149	Sin-yu	,,	,,	,,	,,	,,	4.83		,,
150	Hōsi-no-yu	Awazu	Awazu	Awazu	Nomi	,,	0.21		

151	Sō-yu	Awazu	Awazu	Awazu	Nomi	Isikawa	0.15		
152	Hurō-no-yu	,,	,,	,,	,,	,,	0.14		
153	Yamasiro-Onsen	Yamasiro	Yamasiro	Yamasiro Town	Enuma	,,	0.13		Aug. 1937
154	Zinbara-ryokwan-no-yu	Awara	Hunatu	Awara	Sakai	Hukui	0.56		
155	Simbo-Onsen	Simbo	Simbo	Simbo Town	Turuga	,,	0.20	30.0	Oct. 1937
156	Hatu-yu	Sibu	Hirao	Hirao	Simo-takai	Nagano	0.08		Aug. 1937
157	O-yu No. 1	,,	,,	,,	,,	,,	0.11		,,
158	,, No. 2	,,	,,	,,	,,	,,	0.03		,,
159	Yamada-Onsen	Yamada		Yamada	Kami-takai	,,	0.25		Oct. 1937
160	Yamasio-Kōsen	Kasio	Kasio	Ōzika	Simo-ina	,,	18.5	12.1	May 1936
161	Kosibu-no-yu	,,	,,	,,	,,	,,	0.0		,,
162	Hukagasawa-Kōsen	,,	,,	,,	,,	,,	0.0		,,
163	Nasihara-Kōsen	,,	,,	,,	,,	,,	0.0		,,
164	Me-no-yu	Asama	Asama	Hongō	Higasi-tikuma	,,	0.06		June 1939
165	Higasiyama-Onsen	Higasiyama	Iizibora, Asama	,,	,,	,,	0.08		
166	Iriyamabe-Kōsen	Iriyamabe		Iriyamabe	,,	,,	0.43		
167	Itozawa-no-izumi	Itozawa	Itozawa	Yamaguti	Nisi-tikuma	,,	0.0	13.0	May 1936
168	Siogasawa-Kōsen	Tadati		Tadati	,,	,,	0.0	12.0	,,
169	Ōsiba	Masutomi		Masutomi	Kita-koma	Yamanasi	2.68	22 5	Oct. 1936
170	Ginsentō-huru-yu	,,		,,	,,	,,	9.39	31.0	,,
171	Ginsentō-kami-no-yu	,,		,,	,,	,,	7.35	28.5	,,
172	Ginsentō-naka-no-yu	,,		,,	,,	,,	0.43	22.0	,,
173	Ginsentō-simo-no-yu	,,		,,	,,	,,	3.42	17.0	,,
174	Wadegawara No. 1	,,		,,	,,	,,	6.76	18.0	,,
175	,, No. 2	,,		,,	,,	,,	11.62	19.5	,,
176	Hattyōdaira No. 1	,,		,,	,,	,,	0.11	17.0	,,
177	,, No. 2	,,		,,	,,	,,	26.93	14.9	Apr. 1937
178	Yokote-no-yu	,,		,,	,,	,,	0.71	15.0	,,
179	Higasiobi-no-izumi	,,		,,	,,	,,	35.65	25.0	Oct. 1936

Table 3.—(*Continued*)

1. Radium content of the mineral springs of Japan. (*Continued*)

No.	Spring		Section	Village	County	Prefecture	Radium content g./l.×10^{12}	Temp. of spring °C	Date of sampling
180	Nibuzawa	Masutomi		Masutomi	Kita-koma	Yamanasi	6.22	24.0	Oct. 1936
181	Sio-no-sawa-tugane-yu	,,		,,	,,	,,	1.33	20.0	,,
182	Gantō-hunsen	,,		,,	,,	,,	2.46	20.0	,,
183	Kuridaira No. 1	,,		,,	,,	,,	0.77	22.0	,,
184	,, No. 1. B	,,		,,	,,	,,	0.73	17.0	,,
185	Kuridaira-tennenburo	,,		,,	,,	,,	26.05	23.3	,,
186	Yunokubo No. 1	,,		,,	,,	,,	7.56	21.0	,,
187	Kuridaira No. 3	,,		,,	,,	,,	6.56	19.4	,,
188	Tuganerō No. 1	,,		,,	,,	,,	64.53	28.0	,,
189	,, No. 2	,,		,,	,,	,,	29.39	30.8	,,
190	Tuganerō-naka-no-yu	,,		,,	,,	,,	31.61	23.0	Apr. 1937
191	Tuganerō-kasikiri-no-yu	,,		,,	,,	,,	23.95	25.6	,,
192	Tuganerō-simo-no-yu	,,		,,	,,	,,	25.05	22.2	,,
193	Umamiti-zawa	,,		,,	,,	,,	4.93	15.8	Oct. 1936
194	Totikubo No. 1	,,		,,	,,	,,	32.62	27.0	,,
195	Kinsentō	,,		,,	,,	,,	29.30	31.0	,,
196	Hiuke-sui	,,		,,	,,	,,	39.16	11.2	Apr. 1937
197	Iwaana-zawa	,,		,,	,,	,,	0.76	13.8	,,
198	Yuzawa No. 1	,,		,,	,,	,,	71.81	13.7	,,
199	,, No. 2	,,		,,	,,	,,	1.93	17.7	,,
200	,, No. 3	,,		,,	,,	,,	1.76	18.5	,,
201	,, No. 4	,,		,,	,,	,,	0.00	13.7	,,
202	,, No. 5	,,		,,	,,	,,	0.00	11.5	,,

203	Dōbuti-no-yu	Masutomi		Masutomi	Kita-koma	Yamanasi	0.63	9.0	Apr. 1937
204	Siokawa-asiai-no-yu	,,		,,	,,	,,	1.20	17.5	Oct. 1936
205	Siokawa-nisida-no-yu	,,		,,	,,	,,	0.0	17.8	Apr. 1937
206	Siobuti-no-yu	,,		,,	,,	,,	11.06	14.5	Oct. 1936
207	Nakazima-no-izumi	,,		,,	,,	,,	2.94	14.3	Apr. 1937
208	Wada-matuba-Kōsen	,,		,,	,,	,,	82.66	13.0	Oct. 1936
209	Siozawa-kuromori-Kōsen	,,		,,	,,	,,	11.57	12.0	,,
210	Siozawa-kuromori-yosino-kwan	,,		,,	,,	,,	0.00	8.5	Apr. 1937
211	Simo-no-daira-no-yu	,,		,,	,,	,,	13 00	7.8	,,
212	Kamase-hudō-no-yu	,,		,,	,,	,,	0.00	2.2	,,
213	Matudaira-no-yu	,,		,,	,,	,,	1.52	7.0	,,
214	Kōhu-Onsen	Kōhu	Nisiki-mati	Kōhu City		,,	0.08	39.0	Nov. 1939
215	Yukawa-no-yu	Itō	Yukawa	Itō Town	Takata	Sizuoka	0.16	44.0	Sept. 1937
216	Private bath (T. Ōkawa)	,,	,,	,,	,,	,,	0.04	41.9	,,
217	Matubara-no-yu	,,	Matubara	,,	,,	,,	0.16	42.0	,,
218	Tennen-yu	,,	,,	,,	,,	,,	0.19	43.7	,,
219	Tōkyō-kwan-no-yu	,,	,,	,,	,,	,,	0.04	52.0	,,
220	Masuya-ryokwan-no-yu	,,	,,	,,	,,	,,	0.51	48.0	,,
221	Daitō-kwan-no-yu	,,	,,	,,	,,	,,	0.17	45.7	,,
222	Private bath (K. Iizima)	,,	,,	,,	,,	,,	0.27	40.5	,,
223	Hōsen-kwan-no-yu	,,	,,	,,	,,	,,	0.39	42 0	,,
224	Private bath (S. Naruto)	,,	Kusumi	,,	,,	,,	0.14	38.0	,,
225	Tennen-yu	,,	,,	,,	,,	,,	0.11	44.3	,,
226	Enpanrō-no-yu	,,	,,	,,	,,	,,	0.09	47.7	,,
227	Arai-kwan-no-yu	,,	,,	,,	,,	,,	0.26	40.5	,,
228	Yōki-kwan-no-yu	,,	Oka	,,	,,	,,	0.18	51.0	,,
229	Tōkai-kwan-no-yu	,,	,,	,,	,,	,,	0.17	51.2	,,
230	Private bath (K. Satō)	,,	,,	,,	,,	,,	0.27	52.3	,,
231	,, (M. Aoki)	,,	,,	,,	,,	,,	0.20	52.8	,,

Table 3.—(*Continued*)

1. Radium content of the mineral springs of Japan. (*Continued*)

No.	Spring		Section	Village	County	Prefecture	Radium content g./l. $\times 10^{12}$	Temp. of spring °C	Date of sampling
232	Private bath (T. Suzuki)	Itō	Oka	Itō Town	Takata	Sizuoka	0.28	44 8	Sept. 1937
233	,, (Wakatuki)	,,	,,	,,	,,	,,	0.25	45.0	,,
234	,, (Toyosima)	,,	Kamata	,,	,,	,,	0.08	51.0	,,
235	Ō-yu	Yosina	Yosina	Kami-kano	,,	,,	0.10		,,
236	Moto-yu	,,	,,	,,	,,	,,	0.06		,,
237	Higasi-no-yu	,,	,,	,,	,,	,,	0.00		,,
238	Kikusui-Onsen	,,	,,	,,	,,	,,	0.00		,,
239	Hōsen-Onsen	,,	,,	,,	,,	,,	0.02		,,
240	Kane-no-yu	,,	,,	,,	,,	,,	0.06		,,
241	Kan-no-yu	,,	,,	,,	,,	,,	0.00		,,
242	Kiku-no-yu	,,	,,	,,	,,	,,	0.23		,,
243	Seko-no-yu	Yugasima	Yugasima	,,	,,	,,	0.10		Jan. 1939
244	Kidati-no-yu	,,	,,	,,	,,	,,	0.30		,,
245	Sagasawa-Onsen	Sagasawa	Sakasita, Kadonohara	,,	,,	,,	0.21		Aug. 1937
246	Mito-Onsen	Mito	Mito	Utiura	,,	,,	0.50	42 0	
247	Radium-Kōsen	Ena	Yunosima, Takayama	Hukuoka	Ena	Gihu	0.0	13.1	May 1936
248	Itigaku-Kōsen	Itigaku	Momoyama	Nakatu Town	,,	,,	0.0	12.1	,,
249	Yogarasu-Kōsen	Yogarasu	Ōiwa	,,	,,	,,	0.0	14.0	,,
250	Kitatani-no-izumi	Naegi	Kitatani	Naegi Town	,,	,,	0.0	12.3	,,
251	Sika-no-yu	Kasagi	Kerokubo	Kasagi	,,	,,	0.0	15.1	,,
252	Syō-no-yu	,,	,,	,,	,,	,,	0.0	13.6	,,
253	Iwō-sen	Gero	Simoyasiki	Gero Town	Masuda	,,	0.00		Aug. 1937
254	Yunosima-Onsen	,,	Yunosima	,,	,,	,,	0.00		,,

255	Hon-Onsen	Sakakibara	Hamada, Sakakibara	Sakakibara	Issi	Mie	0.17		
256	Makiyama-Kōsen	Makiyama	Makiyama	Tamataki	Ayama	,,	1.80		
257	Yosino-Onsen	Yosino	Yosinoyama	Yosino	Yosino	Nara	0.43		Nov. 1939
258	Sionoha-Onsen	Sionoha			,,	,,	5.52		,,
259	Kowase-no-yu	Katuura		Katuura Town	Higasi-muro	Wakayama	0.06	28.0	,,
260	Kosi-no-yu No. 1	,,		,,	,,	,,	0.19	41.5	,,
261	Obatakeyama-no-yu No. 1	,,		,,	,,	,,	0.13	35.0	,,
262	Yukawarō-no-yu	Yukawa	Yukawa	Nati	,,	,,	0.10	40.2	,,
263	Kawayu-Onsen	Kawayu	Kawayu, Minasegawa	Ukegawa	,,	,,	2.53		
264	Reimei-no-yu	Yuzaki		Sedo-no-kanayama	Nisi-muro	,,	8.44	57.3	Nov. 1939
265	Higasi-sirahama-Onsen	Higasi-sirahama		,,	,,	,,	15.91		,,
266	Sin-tubaki-no-yu	Tubaki	Tubaki	Higasi-tomita	,,	,,	0.26	33.5	
267	Kami-no-yu	Ryūzin	Ryūzin	Ryūzin	Hidaka	,,	0.57	48.0	
268	Simo-no-yu	,,	,,	,,	,,	,,	0.15		
269	Tomiya-ryokwan-no-yu	Yumura		Onsen	Mikata	Hyōgo	0.79		
270	Hon-Onsen	Arima		Arima Town	Arima	,,	64.97	43.0	Aug. 1939
271	Hana-no-bō-no-yu	,,		,,	,,	,,	47.08	34 5	,,
272	Tansan-sen	,,		,,	,,	,,	0.76	16.0	,,
273	Radium-Kōsen	,,		,,	,,	,,	12.68	27.5	,,
274	Katakosi-Kōsen	,,	Karato	Arino	,,	,,	111.05	21.0	,,
275	Yakendo-Kōsen	,,	Yakendo	,,	,,	,,	61.57	22.0	,,
276	Kyū-Onsen	Takarazuka	Yumoto, Takarazuka	Ryōgen	Mukō	,,	26.46		Aug. 1937
277	Iwai-Onsen	Iwai	Iwai	Iwai Town	Iwami	Tottori	0.56	51.0	Oct. 1937
278	Itō-no-yu	,,	,,	,,	,,	,,	0.75	34.0	,,
279	Simo-no-yu	Yosioka	Higasi-sita-mati, Yosioka	Yosioka	Kedaka	,,	0.61		
280	Suzukiya-no-yu	Hamamura	Hamamura	Seizyō	,,	,,	1.63	49.0	Aug. 1939
281	Tabakoya-no-yu	,,	,,	,,	,,	,,	2.40	63.0	,,
282	Hamanoya-no-yu	,,	,,	,,	,,	,,	2.95	61.0	,,
283	Kyōdō-yu	,,	,,	,,	,,	,,	0.60	48.0	,,

Table 3.—(*Continued*)

1. Radium content of the mineral springs of Japan. (*Continued*)

No.	Spring		Section	Village	County	Prefecture	Radium content g./l. × 10^{12}	Temp. of spring °C	Date of sampling
284	Seirei-no-yu	Hamamura	Hukutaziri, Katimi	Seizyō	Kedaka	Tottori	0.23	41.0	Aug. 1939
285	Sagi-no-yu	Katimi	Katimi	,,	,,	,,	1.93	58.0	,,
286	Nakataya-no-yu	,,	,,	,,	,,	,,	0.73	42.0	,,
287	Kabu-yu	Misasa	Misasa	Misasa	Tōhaku	,,	1.14	45.0	,,
288	Otyaya-no-yu	,,	,,	,,	,,	,,	0.28	61.0	,,
289	Private bath (M. Yamamoto)	,,	,,	,,	,,	,,	0.25	52.5	,,
290	Naka-no-yu	,,	,,	,,	,,	,,	0.35	66.0	,,
291	Hanaya-no-yu	,,	,,	,,	,,	,,	0.85	68.0	,,
292	Nakaya-no-yu	,,	,,	,,	,,	,,	0.90	64.0	,,
293	Bun-aburaya-no-yu	,,	,,	,,	,,	,,	2.24	70.5	,,
294	Hasizuya-no-yu	,,	,,	,,	,,	,,	1.00	66.0	,,
295	Yakusidō-no-yu	,,	,,	,,	,,	,,	3.82	74.5	,,
296	Saiki-honkwan-no-yu	,,	,,	,,	,,	,,	2.37	71.0	,,
297	Akazakiya-no-yu	,,	,,	,,	,,	,,	5.25	69.0	,,
298	Ryōyōzyo-gensen	,,	,,	,,	,,	,,	2.89	72.0	,,
299	Iwa-yu (Otoko-yu)	,,	,,	,,	,,	,,	2.95	57.0	,,
300	,, (Onna-yu)	,,	,,	,,	,,	,,	3.61	68.0	,,
301	,, (Makura-yu)	,,	,,	,,	,,	,,	2.90	60.0	,,
302	Eirakuan-no-yu	,,	,,	,,	,,	,,	0.37	66.5	,,
303	Aburaya-uti-yu	,,	,,	,,	,,	,,	0.89	55.0	,,
304	Aburaya-soto-yu (Mitu-yu)	,,	,,	,,	,,	,,	0.21	51.5	,,
305	Iwasaki-no-yu No. 2	,,	,,	,,	,,	,,	2.81	68.0	,,
306	Kiya-kazoku-yu	,,	,,	,,	,,	,,	3.72	70.0	,,

307	Haikyūzyo-no-yu	Misasa	Misasa	Misasa	Tōhaku	Tottori	2.32	62.0	Aug. 1939
308	Seitō-kwan-no-yu	,,	,,	,,	,,	,,	0.14	60.0	,,
309	Iwasaki-no-yu No. 1	,,	,,	,,	,,	,,	0.37	54.5	,,
310	Bansuirō-no-yu	,,	,,	,,	,,	,,	0.26	48.0	,,
311	Onsen-hotel-no-yu	,,	,,	,,	,,	,,	13.48	68.5	,,
312	Tennen-gankutu-no-yu No. 1	,,	,,	,,	,,	,,	10.06	79.5	,,
313	,, No. 2	,,	,,	,,	,,	,,	7.11	69.0	,,
314	Ōhasi-ryokwan-soto-yu	,,	,,	,,	,,	,,	15.95	79.0	,,
315	Yamadaku-no-yu	,,	Yamada	,,	,,	,,	3.65	56.5	,,
316	Kōyōen-no-yu	,,	,,	,,	,,	,,	16.06	64.0	,,
317	Sinsen-ryō-no-yu	,,	,,	,,	,,	,,	2.12	46.0	,,
318	Misasa-kwan-no-yu	,,	,,	,,	,,	,,	15.01	53.0	,,
319	Okayamaidai-ryōyō-zyo-no-yu	,,	,,	,,	,,	,,	17.67	51.5	,,
320	Gunzihogoin-ryōyōzyo-no-yu	,,	,,	,	,,	,,	0.64	46.0	,,
321	Kabu-yu	Sekigane	Sekigane	Yaokuri	,,	,,	0.11	41.0	,,
322	Kami-no-yu	,,	,,	,,	,,	,,	0.13	46.3	,,
323	Kame-no-yu	,,	,,	,,	,,	,,	0.14	46.0	,,
324	Turu-no-yu	,,	,,	,,	,,	,,	0.13	45.5	,,
325	Tokiwa-no-yu	,,	,,	,,	,,	,,	0.28	45.3	,,
326	Tama-no-yu	,,	,,	,,	,,	,,	0.10	39.5	,,
327	Yōzyō-kwan-no-yu	Tōgō	Hikizi	Tōgō	,,	,,	0.84		
328	Tyūsei-kwan-no-yu No. 1	,,	Tyūkōzi	,,	,,	,,	0.46	42.0	
329	,, No. 2	,,	,,	,,	,,	,,	0.88	47.5	
330	Harada-ryokwan-no-yu	,,	,,	,,	,,	,,	0.52		
331	Doi-ryokwan-no-yu	,,	,,	,,	,,	,,	0.63		
332	Watanabe-ryokwan-no-yu	,,	,,	,,	,,	,,	1.34		
333	Kawamoto-ryokwan-no-yu	,,	,,	,,	,,	,,	0.50		
334	Private bath (H. Huzita) No. 1	,,	,,	,,	,,	,,	0.67		

Table 3.—(*Continued*)

1. Radium content of the mineral springs of Japan. (*Continued*)

No.	Spring		Section	Village	County	Prefecture	Radium content g./l. $\times 10^{12}$	Temp. of spring °C	Date of sampling
335	Private bath (H. Huzita) No. 2	Tōgō	Tyūkōzi	Tōgō	Tōhaku	Tottori	2.63		
336	Turuya-no-yu.	,,	,,	,,	,,	,,	0.12		
337	Private bath (Y. Itō)	Matusaki		Matusaki	,,	,,	0.57		
338	Tyōrakuen-no-yu	Tamatukuri	Tamatukuri	Tamayu	Yatuka	Simane	3.17	72.0	Oct. 1937
339	Matu-no-yu	,,	,,	,,	,,	,,	97.14		
340	Ameya-ryokwan-no-yu	Usio		Usio	Ōhara	,,	0.06		
341	Usio-no-ido	Susa		Higasi-susa	Ihisi	,,	0.30		
342	Moto-yu	Sigaku	Sigaku	Sahime	Anno	,,	1.02	45.0	Aug. 1939
343	Abura-yu	,,	,,	,,	,,	,,	0.80	39.5	,,
344	Koyabara-Onsen	Koyabara	Koyabara	,,	,,	,,	6.15	38.3	,,
345	Radium-Kōsen No. 1	Ikeda	Ikeda	,,	,,	,,	36.43	21.7	,,
346	,, No. 2	,,	,,	,,	,,	,,	19.46	17.7	,,
347	Yugakai-Onsen No. 1	Yugakai	Yugakai	Kasubuti	Ōti	,,	35.06	32.3	,,
348	,, No. 2	,,	,,	,,	,,	,,	30.57	27.3	,,
349	,, No. 3	,,	,,	,,	,,	,,	13.45	25.9	,,
350	Sagi-no-yu	Yunogō	Yunogō	Yunogō	Katuda	Okayama	0.62		Aug. 1937
351	Sin-sagi-no-yu	,,	,,	,,	,,	,,	0.50		
352	Yamano-Onsen	Yamano		Yamano	Hukayasu	,,	0.00		
353	Nisimuraya-no-yu	Yuda	Yuda	Yamaguti City		Yamaguti	0.00	62.0	Aug. 1937
354	Mizuno-ryokwan-no-yu	,,	,,	,,		,,	0.22		,,
355	Hisanoya-no-yu	,,	,,	,,		,,	0.00		,,
356	On-yu	Yumoto	Hukagawa, Yumoto	Hukagawa Town	Ōtu	,,	0.68		,,
357	Rei-yu	,,	,,	,,	,,	,,	0.13		,,

358	Mati-no-yu	Tawarayama	Yumati	Tawarayama	Ōtu	Yamaguti	0.00		Sept. 1937
359	Kawa-no-yu	,,	,,	,,	,,	,,	0·00		,,
3€0	Syō-no-yu	,,	,,	,,	,,	,,	0.00		,,
361	Kami-no-yu	Yanai		Yanai Town	Kuga	,,	0.29		
362	Simo-no-yu	,,		,,	,,	,,	0.63		
363	Yuyama-Kōsen	Yuyama		,,	,,	,,	0.34		
364	Gogōtani-Kōsen	Gogōtani		Gogō	Mitoyo	Kagawa	0.10		
365	Kami-no-yu	Dōgo		Dōgo Town	Onsen	Ehime	0.08		Aug. 1937
366	Yōzyō-yu	,,		,,	,,	,,	0.03		,,
367	Donkobori-no-yu	,,		,,	,,	,,	0.03		,,
368	Siunsen	Kaisyo	Kaisyo	Kawakami	,,	,,	0.21	20.0	Nov. 1937
369	Nibukawa-Kōsen	Nibukawa	Nibukawa	Nibukawa	Oti	,,	0.14		Aug. 1937
370	Spring No. 1	Musasi		Hutukaiti Town	Tukusi	Hukuoka	0.15		Sept. 1937
371	,, No. 2	,,		,,	,,	,,	0.17		,,
372	Hunagoya-Kōsen No. 1	Hunagoya	Hunagoya	Mizuta	Yame	,,	0.02		Aug. 1937
373	,, No. 2	,,	,,	,,	,,	,,	0.00	22.0	,,
374	Hyūgami-Onsen			Ōbuti	,,	,,	0.00		Apr. 1939
375	Reityō-sen	Beppu	Beppu	Beppu City		Ōita	0.22	60.0	Aug. 1937
376	Kotobuki-Onsen	,,	,,	,,		,,	0.18	46.5	Sept. 1937
377	Yanagi-Onsen	,,	,,	,,		,,	0.64	49.5	,,
378	Kusunoki-Onsen	,,	,,	,,		,,	0.39	44.5	Aug. 1937
379	Hurō-sen	,,	,,	,,		,,	0.32	59.0	,,
380	Ta-no-yu A	,,	,,	,,		,,	0.18	57.0	,,
381	,, B	,,	,,	,,		,,	0.15	43.0	,,
382	Kaigan-suna-yu	,,	,,	,,		,,	0.04	58.5	Sept. 1937
383	Takegawara-Onsen	,,	,,	,,		,,	0.74	64.5	Aug. 1937
384	Umezono-Onsen	,,	,,	,,		,,	0.75	6I.5	Sept. 1937
385	Kitamati-Onsen	,,	,,	,,		,,	0.30	55.0	,,
386	Kaimonzi-Kōen-Onsen	,,	,,	,,		,,	0.25	62.0	,,

Table 3.—(*Continued*)

1. Radium content of the mineral springs of Japan. (*Continued*)

No.	Spring		Section	Village	County	Prefecture	Radium content g./l. × 10^{12}	Temp. of spring °C	Date of sampling
387	Matogahama-Onsen	Beppu	Beppu	Beppu City		Ōita	0.76	49.5	Sept. 1937
388	Yumimatu-Onsen	,,	,,	,,		,,	0.25	62.0	,,
389	Yumigahama-Onsen	,,	,,	,,		,,	0.59	49.0	,,
390	Unsenzi-Onsen	,,	,,	,,		,,	0.21	51.0	Aug. 1937
391	Hinode-Onsen	,,	,,	,,		,,	0.17	50.5	Sept. 1937
392	Syōzyu-sen	,,	,,	,,		,,	0.48	47.0	,,
393	Nagesi-Onsen	,,	,,	,,		,,	0.72	52.5	,,
394	Kamiya-Onsen	,,	,,	,,		,,	0.11	51.0	,,
395	Hamawaki-Onsen	,,	Hamawaki	,,		,,	0.14	46.5	Aug. 1937
396	Sira-yu	Kwankaizi	Kwankaizi	,,		,,	0.22	43.0	Sept. 1937
397	Kwankaizi-Onsen	,,	,,	,,		,,	0.00	55.5	,,
398	Horita-Onsen	Horita	Horita	,,		,,	0.09	81.0	,,
399	Bōtyō-sen	Kita-isigaki	Kita-isigaki	,,		,,	0.00	63.0	,,
400	Si-no-yu	Kamegawa	Kamegawa	,,		,,	0.00	54.0	,,
401	Kiyō-sen	,,	,,	,,		,,	0.11	56.8	,,
402	Hamada-Onsen	,,	,,	,,		,,	0.24	80.5	,,
403	Gomusō-Onsen	,,	,,	,,		,,	0.04	45.0	,,
404	Suzi-yu	,,	,,	,,		,,	0.00	58.0	,,
405	Sibaseki-Onsen	,,	Sibaseki, Kamegawa	,,		,,	0.00	67.0	,,
406	Netu-no-yu	Kannawa	Kannawa	,,		,,	0.00	48.0	,,
407	Sibu-yu	,,	,,	,,		,,	0.00	91.0	,,
408	Umi-zigoku	,,	,,	,,		,,	0.54	90.5	1939
409	Zizō-Onsen	Myōban	Myōban	,,		,,	0.00	60.0	Sept. 1937

410	Kakuzyu-sen	Myōban	Myōban	Beppu City		Ōita	0.27	70.0	Sept. 1937
411	Yakusi-Onsen	,,	,,	,,		,,	0.27	87.0	,,
412	Tanakaiti-Onsen	Yuhuin	Tanakaiti	Minami-yuhuin	Hayami	,,	0.00		
413	Yunotubo-Onsen	,,		Kita-yuhuin	,,	,,	0.09		
414	Otomaru-Onsen	,,		,,	,,	,,	0.06		
415	Tukahara-Onsen	Tukahara	Tukahara	,,	,,	,,	0.00		
416	Ō-yu (Kin-no-yu)	Yunohira	Tanigawa	Yunohira	Ōita	,,	0.24	92.0	Aug. 1937
417	Hana-no-yu-Onsen	,,	,,	,,	,,	,,	0.19	78.0	,,
418	Sirataki-Onsen	,,	,,	,,	,,	,,	0.56	65.0	,,
419	Kotobuki-Onsen	,,	,,	,,	,,	,,	0.20	69.0	,,
420	Suna-yu	,,	,,	,,	,,	,,	0.13	63.0	,,
421	Simizu-Onsen	,,	,,	,,	,,	,,	0.12	72.5	,,
422	Yunoto-Onsen	,,	,,	,,	,,	,,	0.01	38.0	,,
423	Hidaya-Onsen	,,	,,	,,	,,	,,	0.10		,,
424	Turuya-no-yu	,,	,,	,,	,,	,,	0.07		,,
425	Turuya-intaku-no-yu	,,	,,	,,	,,	,,	0.05		,,
426	Sinya-Onsen	,,	,,	,,	,,	,,	0.00		,,
427	Ōiwa-Onsen	,,	,,	,,	,,	,,	0.04		,,
428	Azumaya-no-yu	,,	,,	,,	,,	,,	0.09		,,
429	Migimaru-ryokwan-no-yu	,,	,,	,,	,,	,,	0.11		,,
430	Kitabeya-Onsen	,,	,,	,,	,,	,,	0.00		,,
431	Private bath (S. Asō)	,,	,,	,,	,,	,,	0.00		,,
432	,, (K. Maki)	,,	,,	,,	,,	,,	0.00		,,
433	,, (I. Asō)	,,	,,	,,	,,	,,	0.08		,,
434	Nagaiki-no-yu	Nagayu	Nagayu	Nagayu	Naori	,,	0.58	42.0	Oct. 1937
435	Kōyōkwan-no-yu	,,	,,	,,	,,	,,	0.47		,,
436	Tōkyōya-no-yu	Takeo	Takeo	Takeo Town	Kinosima	Saga	0.89	52.0	
437	Huruyu-Onsen	Huruyu	Motomura, Huruyu	Minami-yama	Kosiro	,,	0.05		
438	Tosita-Onsen No. 1	Tosita	Tosita, Kawayu	Tyōyō	Aso	Kumamoto	0.04		

Table 3.—(*Continued*)

1. Radium content of the mineral springs of Japan. (*Concluded*)

No.	Spring		Section	Village	County	Prefecture	Radium content g./l.$\times 10^{12}$	Temp. of spring °C	Date of sampling
439	Tosita-Onsen No. 2	Tosita	Tosita, Kawayu	Tyōyō	Aso	Kumamoto	0.10		
440	Gozen-no-yu	Hinaku	Hinaku	Hinaku Town	Asikita	,,	0.08		Aug. 1937
441	Moto-yu-Onsen	Yunoura	Yunoura	Yunoura	,,	,,	0 04	45.0	Oct. 1937
442	Matu-no-yu-Onsen	,,	,,	,,	,,	,,	0.07	42.0	,,
443	Hukamizu-Onsen	,,	,,	,,	,,	,,	0.02	43.0	,,
444	Iwa-no-yu-Onsen	,,	,,	,,	,,	,,	0.06	45.0	,,
445	Kotobuki-Onsen	,,	,,	,,	,,	,,	0.02		,,
446	Sinohara-Onsen	,,	,,	,,	,,	,,	0.06		,,
447	Private bath (H. Yamamoto)	,,	,,	,,	,,	,,	0.00		,,
448	Mura-no-yu-Onsen	Ibusuki		Ibusuki Town	Ibusuki	Kagosima	0.89		Aug. 1937
449	Yaziga-yu-Onsen	,,		,,	,,	,,	0.55		,,
450	Misuzi-no-yu	,,		,,	,,	,,	0.51		,,
451	Sikorogahama-Onsen	,,		,,	,,	,,	0.89		,,
452	Kaisuien-Onsen	,,		,,	,,	,,	1.05		,,
453	Kairakuen-Onsen	,,		,,	,,	,,	1.18		,,
454	Kabusiki-Onsen	Kabusiki	Yunoura	Isaku Town	Hioki	,,	0.00		,,
455	Private bath (K. Huzita)	Miyanozyō	Yuda	Miyanozyō Town	Satuma	,,	0.06		Sept. 1937
456	,, (K. Hayasida)	,,	,,	,,	,,	,,	0.06		,,
457	,, (T. Kamizono)	,,	,,	,,	,,	,,	0.00		,,
458	,, (A. Kamizono)	,,	,,	,,	,,	,,	0.04		,,
459	,, (M. Morita)	,,	,,	,,	,,	,,	0.00		,,
460	Zairai-no-yu	,,	,,	,,	,,	,,	0.06		,,
461	Tansan-sen	Kirisima, Iwōdani	Simo-nakatugawa	Makizono	Aira	,,	0.00		,,

462	Sio-no-yu	Kirisima, Iwōdani	Simo-nakatugawa	Makizono	Aira	Kagosima	0.01		Sept. 1937
463	Myōban-sen	,,	,,	,,	,,	,,	0.01		,,
464	Iwōsen No. 1	,,	,,	,,	,,	,,	0.00		,,
465	,, No. 2	,,	,,	,,	,,	,,	0.00		,,
466	Hayasida-Onsen	Kirisima, Eino-o	Eino, Simo-nakatugawa	,,	,,	,,	0.39	71.0	,,
467	Enrui-sen	,,	,,	,,	,,	,,	3.52		,,
468	Tetubun-sen	,,	,,	,,	,,	,,	0.15		,,
469	Maruya-ryokwan-no-yu	,,	,,	,,	,,	,,	0.31		,,
470	Ramune-Onsen No. 1	Ramune	Tozaki	,,	,,	,,	0.20		Aug. 1937
471	,, No. 2	,,	,,	,,	,,	,,	0.18		,,
472	Taisyō-kwan-no-yu	Hinatayama	Hinatayama	Hayato Town	,,	,,	0.00		
473	Hukuya-no-yu	,,	,,	,,	,,	,,	0.00		
474	Ansintei-no-yu	,,	,,	,,	,,	,,	0.00		
475	Sinkyū-no-yu	,,	,,	,,	,,	,,	0.00		
476	Yuno-o-Onsen	Yuno-o		Hisikari	Isa	,,	1.53		Sept. 1937
477	Kamii-kwan-no-yu	On-yō		Onyō-men	Gazan	Tyūsei-nan-dō, Korea	0.00	46.0	Oct. 1937

2. Radium content of brines from Japanese oil fields and natural gas field.

No.	Oil-field (Natural gas field)*	County	Prefecture	Petroleum well			Radium content g./l.×10^{12}	*p*H**	Date of sampling
				No.	Depth m.	Flow of water 10^3 l./day			
1	Omonogawa	Kawabe	Akita	1	310	4.0	11.05	7.0	July 1937
2	,,	,,	,,	25	226	1.4	7.14	7.9	,,
3	Araya	,,	,,	1	278	2.3	10.02	7.8	,,
4	Aburaden	,,	,,	1	297	21.0	7.02	7 2	,,

Table 3.—(*Concluded*)

2. Radium content of brines from Japanese oil fields and natural gas field. (*Concluded*)

No.	Oil-field (Natural gas field)*	County	Prefecture	Petroleum well			Radium content g./l. × 10^{12}	pH**	Date of sampling
				No.	Depth m.	Flow of water 10^3 l./day			
5	Oguni	Yuri	Akita	R 32	770	0.6	0.0	8.3	Nov. 1935
6	Oguti	Naka-kanbara	Niigata	R 97	381	5.6	0.0	8.5	,,
7	Ōmo	Minami-kanbara	,,	R 4	894	1.8	12.8	7.9	,,
8	Ōguti	,,	,,	C 1	559		1.10	11.1	Oct. 1937
9	Yoita	Santō	,,		426		0.04		Aug. 1939
10	Takamati	Kariha	,,	R 36	1399	7.9	2.2	7.6	Oct. 1935
11	Iriwada	,,	,,	R 5	971	1.1	1.5	7.2	,,
12	Nagamine	,,	,,	C 93	502	0.03	0.0	5.5	Nov. 1935
13	Okano	,,	,,	R 1	78		2.47		Mar. 1939
14	,,	,,	,,	,,	108		2.84		,,
15	Ōtaki*	Isumi	Tiba				0.0		

** The pH value of the water was measured by K. Hosino with the sample he brougnt to the laboratory.

As will be seen from these tables, compared with the mineral springs in other countries, those in Japan are very poor in radium, which may be owing to the poverty in radium of Japanese rocks. Table 6 shows the radium contents of granite and basalt in Japan and those in other countries, from which the mean radium contents of rocks in Japan seem to be somewhat smaller than the mean for those in other countries as obtained by a number of investigators.

Table 4. Springs with the highest concentration of radium in the world.

Spring	Radium content g./l.	Author and date
Pit-holes No. 1/28, 3/28, 1/31. Petroleum district of Grosny. North Caucasus, Russia.	1.8×10^{-8}	B. A. Nikitin[3] (1930)
Pit-hole "Kasennaya No. 1." Petroleum district of Uchta. Northeastern European Russia.	7.4×10^{-9}	V. I. Baranov,[4] I. D. Kurbatov, A. A. Cherepeunikov (1928)
Radium pit-hole of Heidelberg in Germany.	1.8×10^{-9}	A. Becker[5] (1918)
Tzeliutsing, Szechwan, China.	7.3×10^{-10}	S. Gōda[6] (1939)
Pit-hole Stolb Berecley. Petroleum district of Dagestan. Caucasus, Russia.	3.0×10^{-10}	A. A. Cherepeunikov[4] (1928)
Spring Slavyanovsky. Watering place of Gelesnovodsk. North Caucasus, Russia	2.2×10^{-10}	I. D. Kurbatov,[7] V. Baranov (1928)
Bath, England.	1.4×10^{-10}	W. Ramsay[8] (1912)
Saratoga, New York, America.	1.1×10^{-10}	H. Schlundt[9] (1909)
Karlsbad, Germany	1.0×10^{-10}	W. Kolhorster[10] (1912)

Table 5. Springs with the highest concentrations of radium in Japan.

Spring	Location	Prefecture	Radium content g./l. $\times10^{11}$
Katakosi-Kōsen	Arima	Hyōgo	11.11
Matu-no-yu	Tamatukuri	Simane	9.71
Wadamatuba-Kōsen	Masutomi	Yamanasi	8.44
Yuzawa No. 1	,,	,,	7.18

(4) A. Cherepeunikov, Trans. Geol. Survey (Leningrad), No. 4 (1928).
(5) A. Becker, *Z. anorg. allgem. Chem.*, **209** (1923), 131.
(6) S. Gōda, *Bull. Shanghai Nat. Sci. Research Lab.*, **8** (1939), 252.
(7) I. D. Kurbatov, *Compt. rend. acad. sci.*, U.R.S.S., **1930**, 452.
(8) W. Ramsay, *Chem. News*, **134** (1913), 105.
(9) H. Schlundt, *J. Phys. Chem.*, **18** (1914), 662.
(10) W. Kolhorster, *Verhandl. deut. physik. Ges.*, **14** (1912), 356.

Table 5.—(*Concluded*)

Spring	Location	Prefecture	Radium content g./l. × 10^{11}
Hon-Onsen	Arima	Hyōgo	7.13
Hana-no-bō-no-yu	,,	,,	6.85
Tuganerō No. 1	Masutomi	Yamanasi	6.45
Yakendo-Kōsen	Arima	Hyōgo	6.16
Hiuke-sui	Masutomi	Yamanasi	3.92
Radium-Kōsen No. 1	Ikeda	Simane	3.64
Higasiobi-no-izumi	Masutomi	Yamanasi	3.57
Yugakai-Onsen No. 1	Yugakai	Simane	3.51
Totikubo No. 1	Masutomi	Yamanasi	3.26
Tuganerō-naka-no-yu	,,	,,	3.16
Yugakai-Onsen No. 2	Yugakai	Simane	3.06
Tuganerō No. 2	Masutomi	Yamanasi	2.94
Kinsentō	,,	,,	2.93
Hattyōdaira No. 2	,,	,,	2.69
Kyū-Onsen	Takarazuka	Hyōgo	2.65
Kuridaira-tennen-buro	Masutomi	Yamanasi	2.61
Tuganerō-simo-no-yu	,,	,,	2.51
Tuganerō-kasikiri-no-yu	,,	,,	2.40
Radium-Kōsen No. 2	Ikeda	Simane	1.95
Yamasio-Kōsen	Kasio	Nagano	1.85
Okayamaidai-ryōyōzyo-no-yu	Misasa	Tottori	1.77
Kōyōen-no-yu	,,	,,	1.61
Ōhasi-ryokwan-soto-yu	,,	,,	1.60
Higasi-sirahama-Onsen	Higasi-sirahama	Wakayama	1.59
Alkali-sen	Isobe	Gumma	1.55
Misasa-kwan-no-yu	Misasa	Tottori	1.50
Kagami-no-yu	Matunoyama	Niigata	1.43
Onsen-hotel-no-yu	Misasa	Tottori	1.35
Yugakai-Onsen No. 3	Yugakai	Simane	1.35
Simono-daira-no-yu	Masutomi	Yamanasi	1.30
Petroleum well No. R4	Ōmo oil field	Niigata	1.28
Tansan-sen	Isobe	Gumma	1.16
Wadegawara No. 2	Masutomi	Yamanasi	1.16
Siozawa-kuromori-Kōsen	,,	,,	1.16
Siobuti-no-yu	,,	,,	1.11
Petroleum well No. 1	Omonogawa oil field	Akita	1.11
Toyotomi-Onsen	Toyotomi	Hokkaidō	1.01
Tennen-gankutu-no-yu No. 1	Misasa	Tottori	1.01
Petroleum well No. 1	Araya oil field	Akita	1.00

Table 6. Comparison of mean radium contents of granite and basalt in Japan and those in other countries.

Author	Granite		Basalt	
	Number of specimens	Radium content g./g.×10^{12}	Number of specimens	Radium content g./g.×10^{12}
	Japan			
Z. Hatuda,(11) T. Asayama(12) H. Hamaguti,(13) T. Nakai(13)	41	1.41	6	0.12
	Europe and America			
J. Joly(14)	86	3.01	18	1.40
R. J. Strutt(15)	10	2.79	8	0.44
E. H. Büchner(16)	5	3.70	1	0.50
C. S. Piggot(17)	24	1.75	13	0.96
R. D. Evans(18)			2	0.33

IV. The Mineral Springs of Masutomi, Yamanasi Prefecture. The Masutomi Mineral Springs surpass all other Japanese cold springs in their radon content.(19) Compared with those of strongest radioactivity in Europe, they are inferior only to Oberschlema, Brambach, Lurisia and Joachimsthal, but superior to Gastein, Landeck, Baden-Baden, etc., so that they no doubt hold the fifth position in the world.

With the object of studying these radioactive mineral springs, the writer stayed at this spa from October 15 to 19, 1936, and again from April 24 to 30, 1937. The temperature, the radon content(20) and the flow of the waters were measured at 45 springs in this region, direct at the source, and water samples for radium determination were also drawn direct from the source.

All these determinations are given in Table 7. The pH values of these waters were determined by K. Kuroda in July 1939. The amount of total evaporated residue was determined by S. Okauti for the samples taken in October 1936.

(11) Z. Hatuda, *Mem. Coll. Sci.*, Kyōto Imp. Univ., B, **10** (1934), 63; **12** (1936), 1.
(12) T. Asayama, *Jap. J. Astronomy* and *Geophysics*, **14** (1936), 19.
(13) T. Nakai, *J. Chem. Soc. Japan*, **61** (1940), 149.
(14) J. Joly, *Phil. Mag.*, (6) **24** (1912), 694.
(15) R. J. Strutt, *Proc. Roy. Soc*, **76** A (1905), 88.
(16) E. H. Büchner, *Jahr. f. Radioakt.*, **10** (1913), 516.
(17) C. S Piggot, *Am. J. Sci.*, **35**, A (1938), 227; **25** (1933), 229.
(18) R. D. Evans, *Am. J. Sci.*, **29** (1935), 445.
(19) In October 1939, S. Matuura and I. Iwasaki of the Kyūsyū Imperial University found a very strong radioactive mineral spring at Ikeda, Simane Prefecture, the radon content of which is almost equal to that of Masutomi (v. Table 18).
(20) For measuring the radon content of water, the IM Fontactoscope, made by the Institute of Physical and Chemical Research, Tokyo, was used.

Table 7. Mineral springs of Masutomi and neighbourhood.

No.	Spring	Temp. of spring °C	Temp. of air °C	Radium content g./l. × 10^{12}	Radon per liter of water in Mache's units	pH	Total residue g./l.	Flow of water hl./day	Radium discharge 10^{-3} g. (mg.)/year
1	Ōsiba	22.5	18.0	2.68	362	6.4	9.40	49	0.005
2	Ginsentō-huru-yu	31.0	18.3	9.39	153	6.5	10.30	22	0.008
3	Ginsentō-kami-no-yu	28.5	16.5	7.35	104	6.5	9.64	13	0.004
4	Ginsentō-naka-no-yu	22.0	16.5	0.43	11.6	6.7	9.72	19	0.000
5	Ginsentō-simo-no-yu	17.0	14.0	3.42	42.2		9.78	16	0.002
6	Wadegawara No. 1	18.0	15.6	6.76	588	6.5	3.25		
7	Wadegawara No 2	19.5	16.5	11.62	1343	6.2	7.12	2	0.001
8	Hattyō-daira No. 1	17.0	17.0	0.11	17.5		3.76	49	0.000
9	Hattyō-daira No. 2	14.9	15.7	26.93*				4	0.004
10	Yokote-no-yu	15.0	15.7	0.71*				20	0 001
11	Higasiobi-no-izumi	25.0	17.0	35.65	8.9	6.3	9.69	22	0.029
12	Nibuzawa	24.0	20.5	6.22	330		7.25	77	0.018
13	Sio-no-sawa-tugane-yu	20.0	17.0	1.33	193		4.89	45	0.002
14	Gantō-hunsen	20.0	17.0	2.46	10.9		2.92		
15	Kuridaira No. 1	22.0	11.5	0.77	576	6.0	3.62		
16	Kuridaira No. 1. B	17.0	12.5	0.73	304		2.32		
17	Kuridaira-tennen-buro	23.3	17.0	26.05	7.4	6.4	8.56	32	0.030
18	Yunokubo No. 1	21.0	18.0	7.56	202		3.08		
19	Kuridaira No. 3	19.4	11.9	6.56	355*		7.65		
20	Tuganerō No. 1	28.0	16.5	64.53	4.9	6.4	8.53	175	0.412
21	Tuganerō No. 2	30.8	15.6	29.39	4.0	6.4	8.99	31	0.033
22	Tuganerō-naka-no-yu	23.0	16.3	31.61*				9	0 010
23	Tuganerō-kasikiri-no-yu	25.6	14.4	23.95*	6.5*			36	0.032
24	Tuganerō-simo-no-yu	22.2	14.0	25.05*				1	0.001
25	Umamiti-zawa	15.8	14.5	4.93	6.4		3.63		
26	Totikubo No. 1	27.0	13.3	32.62	25.6	6.3	9.42	81	0.096

Table 7.—(*Concluded*)

No.	Spring	Temp. of spring °C	Temp. of air °C	Radium content g./l. $\times 10^{12}$	Radon per liter of water in Mache's units	pH	Total residue g./l.	Flow of water hl./day	Radium discharge 10^{-3} g. (mg.)/year
27	Kinsentō	31.0	16.0	29.30	3.3	6.3	7.49	410	0.460
28	Hiuke-sui	11.2	15.7	39.16*			4.05	2	0.003
29	Iwaana-zawa	13.8	12.3	0.76*	0.0*		0.17		
30	Yuzawa No. 1	13.7	12 8	71.81*	16.1*			59	0.155
31	,, No. 2	17.7	12.8	1.93*					
32	,, No. 3	18.5	12 8	1.76*					
33	,, No. 4	13.7	12.8	0.00*					
34	,, No. 5	11.5	13.8	0.00*					
35	Dōbuti-no-yu	9.0	15.7	0.63*			2.45		
36	Siokawa-asiai-no-yu	17 5	16.5	1.20	1.5*		3.81	64	0.003
37	Siokawa-nisida-no-yu	17.8	20.6	0.0*	0.4*				
38	Siobuti-no-yu	14.5	16.5	11.06	3.9*		5.10	32	0.013
39	Nakazima-no-izumi	14.3	18.3	2.94*	1.7*				
40	Wada-matuba-Kōsen	13.0	16.0	82.66	6 1*	6.4	13.34	2	0.006
41	Siozawa-kuromori-Kōsen	12.0	16.0	11.57	1.8*		4.46	8	0.003
42	Siozawa-kuromori-yosi-no-kwan	8.5	11.0	0.00*	1.2*				
43	Simo-no-daira-no-yu	7.8	15.0	13.00*	2.3*			26	0.012
44	Kamase-hudō-no-yu	2.2	17.8	0.00*	1.0*				
45	Matudaira-no-yu	7.0	15.5	1.52*					
								Total:	1.343

The orifice temperature was measured on the same sample as that which was determined for the radium content.

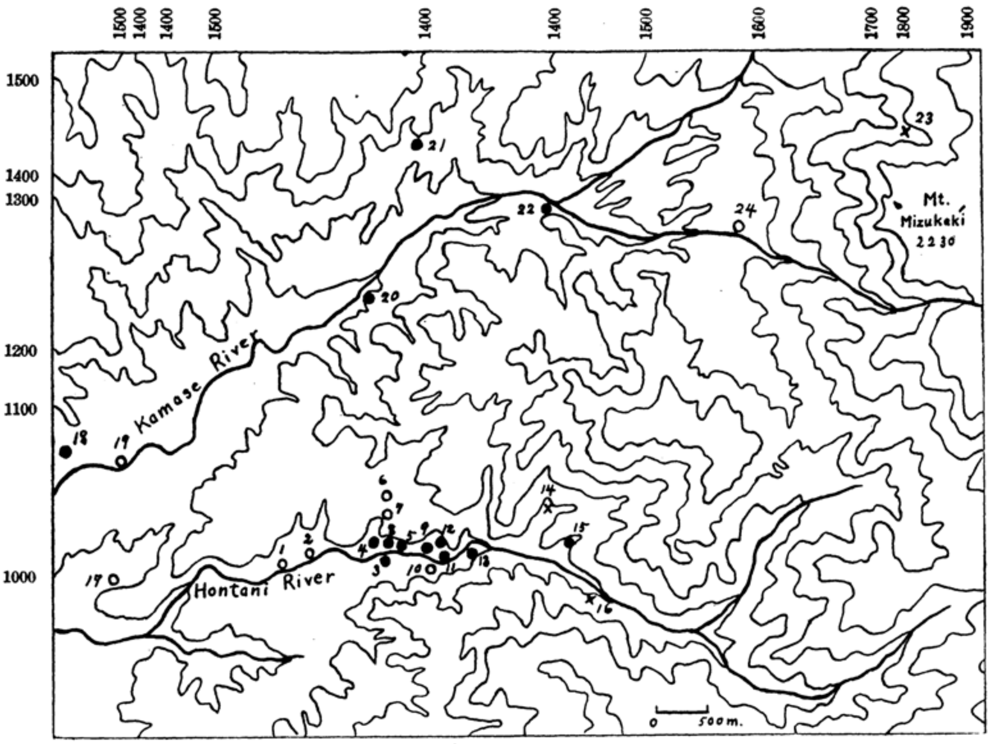
The radium content of the water was determined with the sample taken in October, 1936. Sampling of the waters starred was made in April, 1937.

The radon content of the water was measured in October, 1936 and that of the sample starred in April, 1937.

The flow of water was measured in April, 1937.

The pH value was determined by Kuroda in July, 1939.

The total residue was determined by Okauti from sample drawn October, 1936.

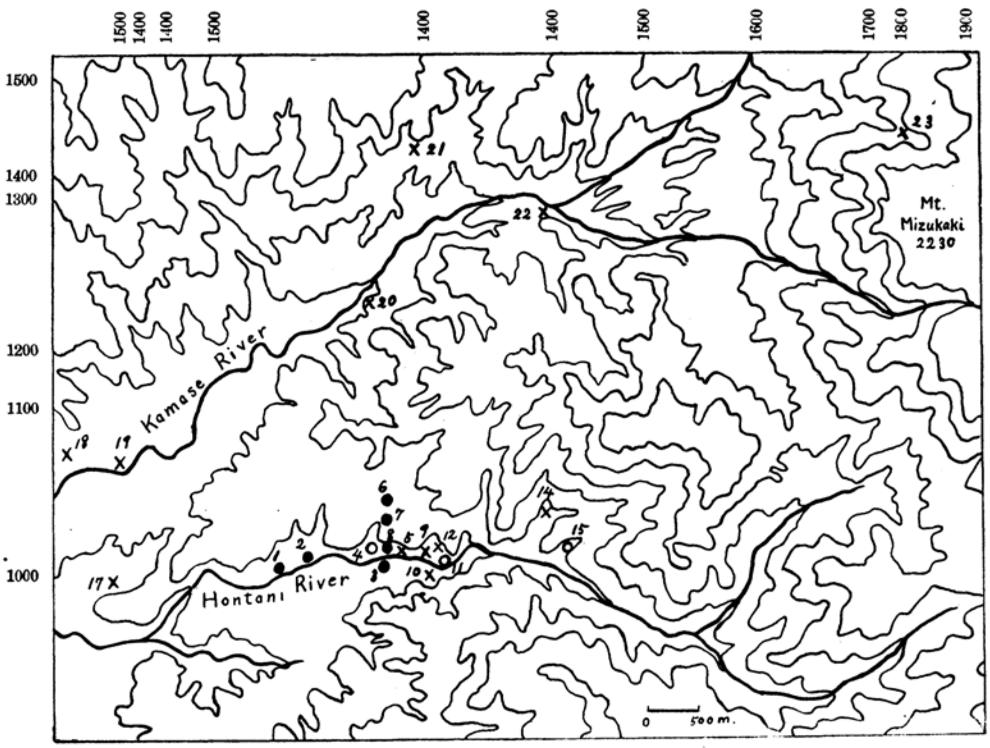


● Mineral spring containing more than 1×10^{-11} g. Ra/l.
○ Mineral spring containing 1×10^{-12} g. Ra/l.$\sim 1 \times 10^{-11}$ g. Ra/l.
× Mineral spring containing less than 1×10^{-12} g. Ra/l.

1. Ōsiba
2. Ginsentō
3. Wadegawara
4. Hattyōdaira
5. Higasiobi-no-izumi
6. Nibuzawa
7. Sio-no-sawa-tugane-yu
8. Kuridaira
9. Tuganerō
10. Umamiti-zawa
11. Totikubo
12. Kinsentō
13. Hiukesui
14. Iwaanazawa
15. Yuzawa
16. Dōbuti-no-yu
17. Siokawa-no-yu
18. Siobuti-no-yu
19. Nakazima-no-izumi
20. Wada-matuba-Kōsen
21. Siozawa-kuromori-no-yu
22. Simono-daira-no-yu
23. Kamase-hudō-no-yu
24. Matudaira-no-yu

Fig. 3. Geographical distribution of the amount of radium in the Masutomi mineral springs.

Geographical distribution of radium and radon contents. The geographical distribution of radium and radon contents is shown on the accompanying maps, Figs. 3 and 4, from which it will be seen that almost all the springs, especially those with high radium and radon contents, are distributed along the two rivers, Hontani and Kamase. It shows that these spring waters flow out along these weak points of the earth's crust. It is also interesting to note as will be seen from these figures that, whereas those springs that are situated along the Hontani River are rich in both radium and radon, those along the Kamase River show compara-



The numerals against the black circle, open circle or cross are the mineral spring numbers in the Fig. 3.

● Mineral spring containing more than 100 Mache's units of radon.
○ Mineral spring containing 10~100 Mache's units of radon.
× Mineral spring containing less than 10 Mache's units of radon.

Fig. 4. Geographical distribution of the amount of radon in the Masutomi mineral springs.

tively low radio-activity notwithstanding their extraordinary high radium concentration.

Relation between the radium content and the radon content of spring waters. As will be seen from Table 7, the amount of radium in all the mineral waters was found to be much less than that required for radioactive equilibrium with the radon that is present in them. Even in the mineral waters at Wadamatuba, which shows the highest radium content, the amount of radium found was 0.83×10^{-10} g. per liter of water and the radon content 22.2×10^{-10} curie per liter, showing that the radon in the water is about 27 times greater than what the radium present in the water can produce.

According to Mache and Schweidler, the radioactivity of spring water is mainly due to the geological structure of the strata through which they last passed. The probable sources of the radium contained in spring waters are also largely the rocks where the waters originate and through which they pass, from which consideration the above results, showing that the amount of radium in spring waters is remarkably small compared

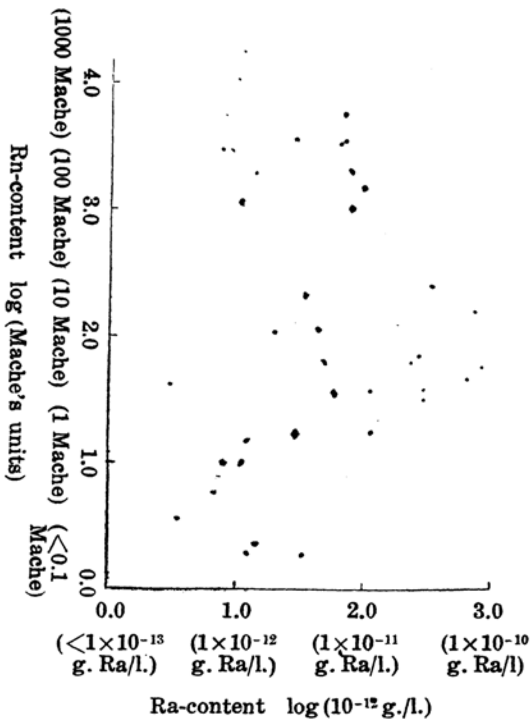


Fig. 5. Relation between the Ra-content and the Rn-content of the Masutomi mineral springs.

with that of radon, may be simply explained by the difference in their solubilities in water. It indicates that spring waters usually contain an excess of free radon which they adsorb from rocks independently of radium, besides that which have their origin in radium dissolved in waters.

In Fig. 5, the radium content of spring waters is plotted against their radon content. No correlation can be established between them. Mineral waters with high radium content do not always show a corresponding high radon concentration, while those with high radon content do not always contain a large amount of radium. It also indicates that the radon from mineral waters is not due to the dissolved radium alone, as stated above.

Variation in radon and radium contents. Radon content. The radon content of some of the springs was measured at intervals of several years, namely, first in August, 1914, by R. Ishizu, then in October, 1936, and in April, 1937, by the writer, and most recently, in July, 1939, by Oana and Kuroda, the results of which are shown in Table 8. Most of them varied but little during these intervals of time, although a few showed remarkable variations, especially the mineral waters of Wadegawara No. 2.

The radon concentration in spring waters may be affected by various factors, such as the temperature and volume output of water, weather conditions, especially the amount of rainfall, and also by the alterations in the underground water and changes in the rocks through which the waters pass. K. Noguti,[21] after measuring the seasonal changes in radioactivity, temperature and flow of spring waters at Zigokudani near Volcano Asama, continuously during the period from June 1936 to June 1937, reported that their radioactivity increases with increase in flow of the water and decreases with rise in temperature of the water. S. Gōda[22] also, upon measuring the radioactivity, temperature, pH value, turbidity, and flow of water of a well in Shanghai, China, and the amount of rainfall there continuously from February 1933 to December 1936, reported that its radon concentration reaches maximum in summer and

(21) K. Noguti, *J. Chem. Soc. Japan*, **60** (1939), 7.
(22) S. Gōda, *Bull. Shanghai Nat. Sci. Research Lab.*, **8** (1939), 269.

Table 8. Variations in the radioactivity of springs in Masutomi.

No.*	Spring	Radon per liter of water in Mache's units							
		August 1914 (R. Ishizu)		October 1936		April 1937		July 1939 (S. Oana and K. Kuroda)	
		Temp. of Spring °C	Mache	Temp. of Spring °C	Mache	Temp. of Spring °C	Mache	Temp. of Spring °C	Mache
1	Ōsiba	19.0	277	22.5	362				
2	Ginsentō-huru-yu	29.5	103	31.0	153				
6	Wadegawara No. 1	14.0	657	18.0	588				
7	,, No. 2	21.5	828	19.5	1343	16.0	708	20.5	1085
11	Higasiobi-no-izumi			25.0	8.9			24.5	9.0
12	Nibuzawa	23.5	394	24.0	330				
13	Sio-no-sawa-tugane-yu	17.5	236	20.0	193				
15	Kuridaira No. 1	16.0	590	22.0	576				
16	,, No. 1. B.	20.0	318	17.0	304				
18	Yunokubo No. 1	17.0	248	21.0	202				
19	Kuridaira No. 3					19.4	355	21.4	233
20	Tuganerō No. 1			28.0	4.9			30.1	4.3
21	,, No. 2			30.8	4.0			29.6	3.5
25	Umamiti-zawa	12.0	2.9	15.8	6.4				
26	Totikubo No. 1			27.0	25.6			28.0	16.1
27	Kinsentō	31.5	2.2	31.0	3.3			31.0	2.1

* Numbers correspond to those used in Table 7.

minimum in winter and furthermore, that the variation in radioactivity runs parallel to that of the pH value, turbidity, and flow of water, from all which he concluded that the variation in radon concentration is due to the activity of underground water movement as the result of the amount of rainfall.

The variation in the radioactivity of spring water at Masutomi also can be explained only after much data of such factors as have just been mentioned have been obtained, so that, for the present, we shall leave this problem merely as a suggestion and wait the accumulation of further data.

Radium content. The radium content of some of the mineral springs in Masutomi was determined in October, 1936, by the writer and again in July, 1939, by Kuroda, the results being given in Table 9. Some of them show remarkable variation. In this case their radium contents also are considered to be affected by numerous factors, as in the case of radon.

There seems to be no relation between the variation in radium content of these mineral waters and that of their temperature and the amount of solid matter held, although it is worthy of note that, as will be seen from Tables 8 and 9, the variation in radium content of these waters runs parallel to that of their radon content. In every case, the radium content was higher in October, 1936, than in July, 1939. Parallel with this result, the radon concentration of the former is also higher than that of the latter.

Table 9. Variations in radium content of the Masutomi springs.

No.**	Spring	October 1936			July 1939 (K. Kuroda)		
		Temp. of spring °C	Ra content g./l.×10¹²	Total residue g./l.	Temp. of spring °C	Ra content g./l.×10¹²	Total residue g./l.
1	Ōsiba	22.5	2.68	9.40	22.6	1.93	
2	Ginsentō-huru-yu	31.0	9.39	10.30	30.0	6.50	11.18
3	Ginsentō-kami-no-yu	28.5	7.35	9.64	28.5	5.86	
4	Ginsentō-naka-no-yu	22.0	0.43	9.72	27.0	0.58	10.61
5	Ginsentō-simo-no-yu	17.0	3.42	9.78		2.05	11.33
6	Wadegawara No. 1	18.0	6.76	3.25	22.0	8.20	5.63
7	,, No. 2	19.5	11.62	7.12	20.5	8.20	7.32
11	Higasiobi-no-izumi	25.0	35.65	9.69	24.5	18.89	10.17
12	Nibuzawa	24.0	6.22	7.25	24.0	7.73	16.20
13	Sio-no-sawa-tugane-yu	20.0	1.33	4.89	20.9	0.58	4.84
14	Gantō-hunsen	20.0	2.46	2.92	26.0	1.74	4.11
15	Kuridaira No. 1	22.0	0.77	3.62	21.2	1.46	3.72
16	,, No. 1. B.	17.0	0.73	2.32		0.58	10.00
17	Kuridaira-tennen-buro	23.3	26.05	8.56	23.6	14.06	8.95
19	Kuridaira No. 3	19.4	6.56	7.65	21.4	5.56	6.67
20	Tuganerō No. 1	28.0	64.53	8.53	30.1	24.61	9.39
21	,, No. 2	30.8	29.39	8.99	29.6	30.31	9.66
26	Totikubo No. 1	27.0	32.62	9.42	28.0	26.95	3.07
27	Kinsentō	31.0	29.30	7.49	31.0	21.97	8.33
28	Hiuke-sui	11.2*	39.16*		14.7	28.71	4.05
29	Iwaana-zawa	13.8*	0.76*		13.0	0.41	0.17
35	Dōbuti-no-yu	9.0*	0.63*		16.0	1.93	2.45
40	Wada-matuba-Kōsen	13.0	82.66	13.34	11.5	84.36	15.06

* Sampling in April 1937. ** Numbers correspond to those used in Table 7.

Relation between the radium and radon contents and the orifice temperature of spring waters. From the results thus far obtained we shall consider the relation between the radon and radium contents of the springs in Masutomi and their temperature. Looked at as a whole, the quantities of both the radium and radon in the waters seem to bear no general relation to the temperature of the waters.

We shall next attempt to compare springs that are situated close together within a distance of about 100 meters. The radium and radon contents and the temperature of these spring waters are given in Table 10.

A tendency will be seen from this table (excluding the case of Group IV and Group V) for the radium content of the waters to increase with

Table 10. Relation between the radium and radon contents and the temperature of springs.

Spring	Temp. of spring °C	Radium content g./l.×10^{12}	Radon per liter of water in Mache's units
		Group I	
Ginsentō-huru-yu	31.0	9.39	153
Ginsentō-kami-no-yu	28.5	7.35	104
Ginsentō-naka-no-yu	22.0	0.43	11.6
Ginsentō-simo-no-yu	17.0	3.42	42.2
		Group II	
Wadegawara No. 2	19.5	11.62	1343
,, No. 1	18.0	6.76	588
		Group III	
Kuridaira-tennen-buro	23.3	26.05	7.4
Kuridaira No. 1	22.0	0.77	576
,, No. 1. B.	17.0	0.73	304
		Group IV	
Tuganerō No. 2	30.8	29.39	4.0
,, No. 1	28.0	64.53	4.9
Tuganerō-kasikiri-no-yu	25.6	23.95	6.5
Tuganerō naka-no-yu	23.0	31.61	
Tuganerō-simo-no-yu	22.2	25.05	
		Group V	
Yuzawa No. 3	18.5	1.76	
,, No. 2	17.7	1.93	
,, No. 1	13.7	71.81	16.1
,, No. 4	13.7	0.00	
,, No. 5	11.5	0.00	
		Group VI	
Siozawa-kuromori-Kōsen	12.0	11.57	1.8
Siozawa-kuromori-yosinokwan	8.5	0.00	1.2

rise in their temperature. In the spring waters of Group IV, their radioactivity increases with fall in temperature, which may naturally be expected from the variation in the solubility of radium and radon with temperature.

Relation between the radium and radon contents and the pH value of spring waters. As will be seen from Table 7, all the spring waters in this region are weakly acidic, showing a *p*H value of 6.0~6.7. The maximum is 6.7 in the water of Ginsentō-naka-no-yu and the minimum 6.0 in that of Kuridaira No. 1. The difference in *p*H values for each spring water is consequently too small for comparison with the radium and radon contents.

Relation between the radium content and the amount of total solid matter in spring waters. The radium content and the amount of total evaporated residue (determined by Okauti) of the spring waters in Masutomi are given in Table 7. Plotting the amount of total residue as abscissae and the radium content as ordinates, we get Fig. 6. There seems to be a tendency here for the samples of high radium content to have fairly large proportions of solid matter.

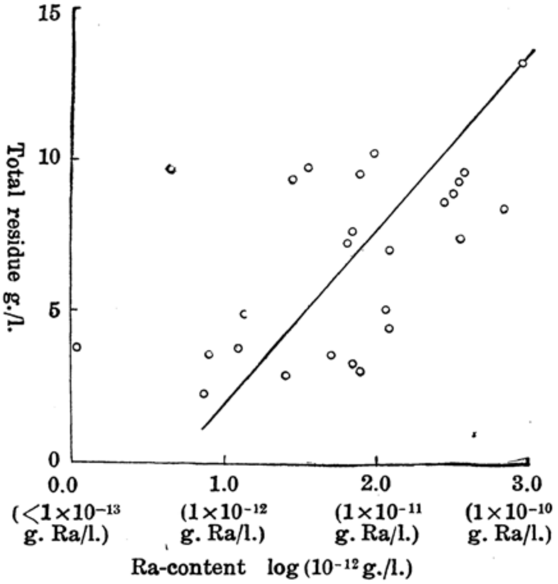
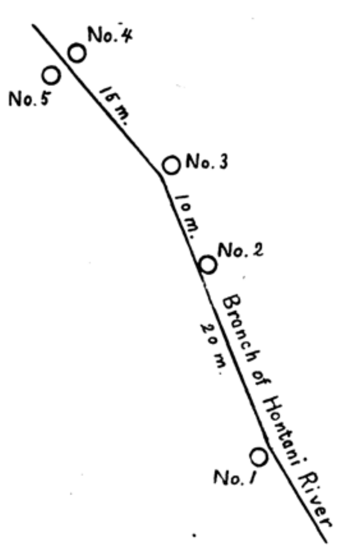


Fig. 6. Relation between the Ra-content and the amount of total residue in the Masutomi mineral springs.



Fg. 7. Mineral springs of Yuzawa (Masutomi).

The mineral springs of Yuzawa. The five mineral springs of Yuzawa are located along a branch of the Hontani River, close together, being separated by distances of only 10 or 20 meters (V. Fig. 7), but as will be seen from Table 10, the radium content varies remarkably in this group of closely related springs. Notwithstanding that the radium content of the water at Spring No. 1 exceeds 7×10^{-11} g. radium per liter of water, no radium is detected in that of Springs Nos. 4–5, showing that they may pass through different strata, although they gush out to the surface of the earth within only a few meters of each other.

On the other hand, B. A. Nikitin(23), after studying the waters of the Ribi-Eibat oil field, reported that although water obtained from the same strata are closely related chemically, they contain different quantities of radium, in view of which it is not certain whether the waters in Yuzawa do not also come from different strata. This question can be answered only after a sufficient number of investigations have been made on the mineral waters and of the geological formations through which they pass.

Similar examples are found in the other groups of springs in this region (V. Table 10). Although the three springs of Kuridaira lie at

(23) B. A. Nikitin, *Tras. inst. ètat radium* (U.S.S.R.), **2** (1933), 160.

distances of less than 10 meters from one another, the radium content of the waters at Tennen-buro is about 40 times those of the other two, whereas the radon content of the latter is much higher than that of the former.

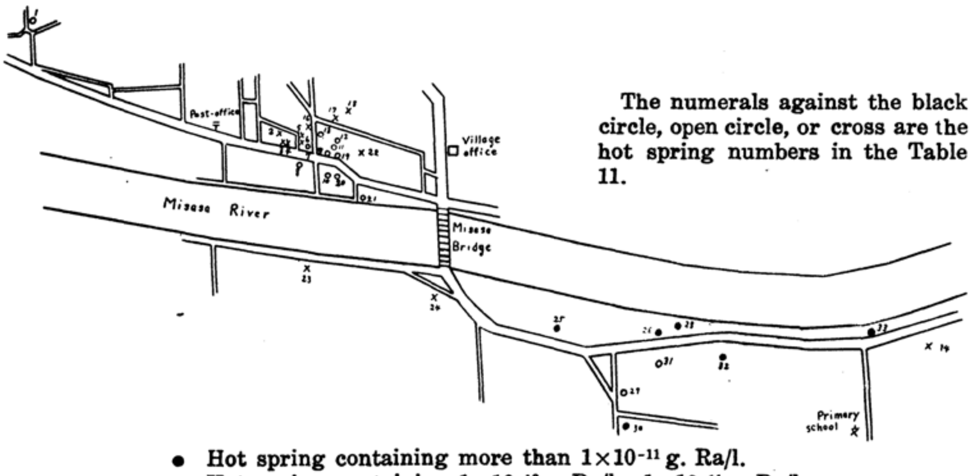
While the distance between the sources of two springs in Wadegawara is only one or two meters, the radium and radon contents of Spring No. 2 are nearly twice that of Spring No. 1.

Radium discharge. The radium discharge is calculated for each spring from its radium content and daily flow of water, the results of which are shown in Table 7. The radium brought up by a group of springs in this district amounts to about one milligram per year.

V. Hot Springs of Misasa, Tottori Prefecture. The Misasa Hot Springs, the most radioactive in Japan, ranks second only to Ischia in Italy. They are almost equal to the most radioactive hot spring in Gastein, which surpasses all the other hot springs of the world in radon content.

It is situated on the western and eastern banks of the Misasa River (V. Fig. 8). The region itself is flat with only a slight inclination, and of limited area. In this small space gush out in series a large number of hot springs of high radon content and high temperature, their sources being located mostly near the stream. (A large number of hot springs gush out in the stream too.) No less than 50 springs are found in this district, not very distant from one another.

The writer, who spent about a weak here in August, 1939, observed the temperature, pH value, and the flow of water of the 34 springs direct at the source and took samples of water for radium determination direct from the source, the results of all which are given in Table 11. For some of them the radioactivity was measured by Ishizu and Hattori and the total evaporated residues were determined by Kuroda and the Imperial Hygienic Laboratory of Tokyo, the results of which are also given in this table for reference.



● Hot spring containing more than 1×10^{-11} g. Ra/l.
○ Hot spring containing 1×10^{-12} g. Ra/l.$\sim1\times10^{-11}$ g. Ra/l.
× Hot spring containing less than 1×10^{-12} g. Ra/l.

Fig. 8. Geographical distribution of the amount of radium in the Misasa hot springs.

Table 11. Hot springs of Misasa.

No.	Spring	Temp. of spring °C	Temp. of air °C	Radium content g./l.×10^{12}	Radon per liter of water in Mache's units (R. Ishizu, Y. Hattori)	pH	Flow of water hl./day	Radium discharge 10^{-3} g. (mg.)/year	Total residue g./l. (K. Kuroda)
	Misasa Group								
1	Kabu-yu (Public bath)	45.0	24.4	1.14	10.2*	6.9	1967	0.082	
2	Otyaya-no-yu	61.0	26.1	0.28	27.9*	6.9			
3	Private bath (M. Yamamoto)	52.5	26.9	0.25		6.7	98	0.001	
4	Naka-no-yu (Public bath)	66.0	28.3	0.35	11.0*	7.0	625	0.008	1.24**
5	Hanaya-no-yu	68.0	25.9	0.85	74.4	6.8	567	0.018	
6	Nakaya-no-yu	64.0	27.2	0.90	24.5*	6.8	407	0.013	
7	Bun-aburaya-no-yu	70.5	28.9	2.24	24.5*	7.0	44	0.004	
8	Hasizuya-no-yu	66.0	26.4	1.00	15.5*	6.9			
9	Yakusidō-no-yu	74.5	28.6	3.82	14.5	7.2	75	0.010	0.86**
10	Saiki-honkwan-no-yu	71.0	26.1	2.37		7.5	1832	0.158	
11	Akazakiya-no-yu	69.0	26.8	5.25	53.4	7.0	861	0.165	
12	Ryōyōzyo-gensen	72.0	28.7	2.89	130.5	6.8	1094	0.115	1.31**
13	Iwa-yu (Otoko-yu)	57.0	25.0	2.95	36.0*	6.8	825	0.035	
14	,, (Onna-yu)	68.0	28.7	3.61	34.5	6.9	536	0.071	1.23**
15	,, (Makura-yu)	60.0	25.0	2.90		6.9	62	0.007	
16	Eirakuan-no-yu	66.5	25.3	0.37	48.8	7.1	475	0.006	
17	Aburaya-uti-yu	55.0	29.0	0.89	19.2*	6.9	176	0.006	

18	Aburaya-soto-yu (Mitu-yu)	51.5	29.0	0.21		6.8	152	0.001	
19	Iwasaki-no-yu No. 2	68.0	27.2	2.81	19.3	7.2	496	0.051	1.16**
20	Kiya-kazoku-yu	70.0	26.7	3.72	28.4*	7.0	172	0.023	
21	Haikyūzyo-no-yu	62.0	28.8	2.32		6.8	684	0.058	0.92
22	Seitō-kwan-no-yu	60.0	26.3	0.14	85.6	7.0	610	0.003	
23	Iwasaki-no-yu No. 1	54.5	26.5	0.37	15.1	7.1	88	0.001	0.64
24	Bansuirō-no-yu	48.0	26.3	0.26		7.2	87	0.001	1.57
				Yamada Group					
25	Onsen-hotel-no-yu	68.5	26.8	13.48		6.5			0.72
26	Tennen-gankutu-no-yu No. 1	79.5	26.8	10.06	42.9	6.4	142	0.052	
27	,, No. 2	69.0	26.8	7.11		6.3			
28	Ōhasi-ryokwan-soto-yu	79.0	30.7	15.95		6.6			
29	Yamadaku-no-yu (Public bath)	56.5	25.4	3.65	168.6	6.6	392	0.052	
30	Kōyōen-no-yu	64.0	28.5	16.06		6.2	47	0.028	
31	Sinsen-ryō-no-yu	46.0	32.0	2.12		6.3			
32	Misasa-kwan-no-yu	53.0	26.8	15.01		6.4			
33	Okayamaidai-ryōyōzyo-no-yu	51.5	27.4	17.67		6.8			1.53
34	Gunzihogoin-ryōyōzyo-no-yu	46.0	27.4	0.64		7.4	276	0.006	1.21
							Total:	0.975	

Data on radon content of the water obtained by Y. Hattori[24] in October, 1935; that starred by R. Ishizu[25] in February and March, 1914.
** Determined by the Imperial Hygienic Laboratory of Tokyo.

(24) Y. Hattori, *Bull. Imp. Hygienic Lab.*, **52** (1939), 142. (25) R. Ishizu, "*The Mineral springs of Japan,*" (1916), 135.

Geographical distribution of radium content. The geographical distribution of radium content is shown on the accompanying map, Fig. 8. The springs in this district may be devided into two groups, the Misasa and Yamada. The 24 springs, Nos. 1–24, belong to the former and the 10 springs Nos. 25–34 to the latter.

As will be seen from Fig. 8, all the waters of the Yamada group have fairly high radium content compared with that of the Misasa group, with the exception of spring No. 34, the chemical quality of which also differs from the others of this group, as will be shown later.

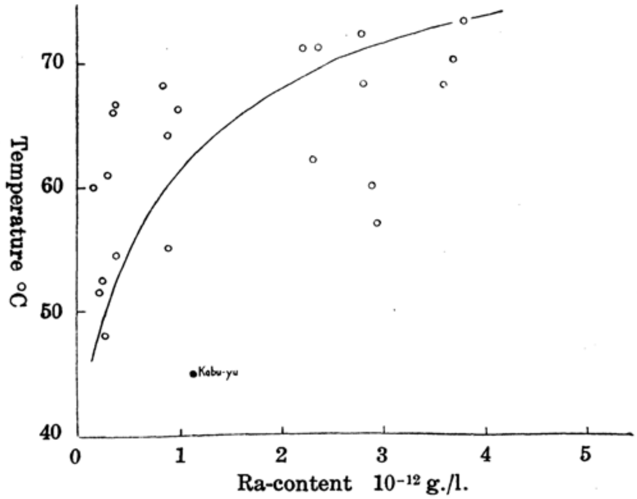
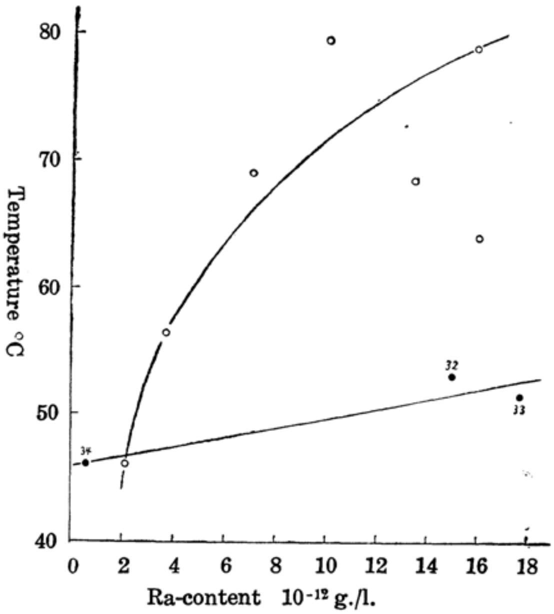


Fig. 9. Relation between the Ra-content and the orifice temperature of the springs of the Misasa group.

Relation between the radium content and the orifice temperature of spring waters. We shall consider the 24 springs of the Misasa group, the radium contents of which are plotted in Fig. 9 against their orifice temperatures, excluding the spring of Kabu-yu, seeing that it is situated far away from the rest. Here we find that the general tendency is for the radium content to increase with the rise in their orifice temperature.

We shall next take the ten springs of the Yamada group, the radium contents of which are plotted as ordinates and their orifice temperature as abscissae, resulting in Fig. 10. Since the three springs, Nos. 32–34, lie some distance away from the others, these three are again separated from this group, so that in each group the points lie nearly close to a straight line. There is again a tendency for the radium content to increase with the



The numerals against the black circle are the hot spring numbers in the Table 11.

Fig. 10. Relation between the Ra-content and the orifice temperature of the springs of the Yamada group.

rise in their orifice temperature, which results correspond well with the solubility of radium salts in water.

Relation between the radium content and the pH value of spring waters. As will be seen from Table 11, the *p*H value of the waters of the Yamada group is somewhat lower than that of the Misasa group, while the radium content of the former is higher than that of the latter, which may be due to differences in the geological formations through which the waters pass. It is also worthy of note that the *p*H value of Spring No. 34 differs from the others in the same group, besides having a lower radium content than the others.

Radium discharge. The radium discharge is calculated for each spring from its radium content and daily flow of water, the result of which is given in Table 11. The radium brought up by a group of springs in this district amounts to about 1 milligram per year.

VI. Thermal Springs of Itō, Sizuoka Prefecture. The water samples of the Itō Thermal Springs were obtained in September, 1937, by the Itō Hot Spring Owners' Association. The orifice temperature and the *p*H value of these springs were measured by Kuroda in December, 1939, with results as shown in Table 12.

Table 12. Thermal springs of Itō.

No.	Spring	Temp. of spring °C	Radium content g./l. × 10^{12}	*p*H (Kuroda)	No.*	l:m:n
1	Yukawa, Yukawa-no-yu (Komoti-yu)	44.0	0.16	7.8	145	6:58:35
2	,, Private bath (T. Ōkawa)	41.9	0.04	8.2	140	4:51:46
3	Matubara, Matubara-no-yu (Ō-yu)	42.0	0.16	8.0	117	5:54:41
4	,, Tennen-yu	43.7	0.19	8.0	118	6:58:36
5	,, Tōkyō-kwan-no-yu	52.0	0.04	8.0	129	3:63:34
6	,, Masuya-ryokwan-no-yu	48.0	0.51	7.5	112	45:43:12
7	,, Daitō-kwan-no-yu	45.7	0.17	7.5	—	—
8	,, Private bath (K. Iizima)	40.5	0.27	7.3	93	25:41:34
9	,, Hōsen-kwan-no-yu	42.0	0.39	7.5	83	27:49:24
10	Kusumi, Private bath (S. Naruto)	38.0	0.14	7.4	28	11:48:42
11	,, Tennen-yu	44.3	0.11	8.0	32	7:25:67
12	,, Enpanrō-no-yu	47.7	0.09	7.5	39	10:47:43
13	,, Arai-kwan-no-yu	40.5	0.26	7.4	6	21:30:50
14	Oka, Yōki-kwan-no-yu	51.0	0.18	8.0	178	2:90: 9
15	,, Tōkai-kwan-no-yu	51.2	0.17	7.9	155	6:86: 8
16	,, Private bath (K. Satō)	52.3	0.27	8.1	—	—
17	,, ,, (M. Aoki)	52.8	0.20	8.0	159	0:95: 5
18	,, ,, (T. Suzuki)	44.8	0.28	7.6	151	10:68:22
19	,, ,, (Wakatuki)	45.0	0.25	7.1	149	3:61:36
20	Kamata, Private bath (Toyosima)	51.0	0.08	8.0	210	0:76:24

* Numbers correspond to those used in the preceding paper by T. Fukutomi.(26)

After investigating the physical and chemical properties of the water of 207 of the thermal springs in the town of Itō and its neighbourhood, T. Fukutomi and Z. Huzii[26] concluded as follows:

There is no doubt of the existence in the town of Itō and neighbourhood of 3 primary springs of entirely different character, namely, the one (B) characterized by high temperature, small amounts of chemical constituents, and the other (A), differing from the preceding in its relatively low temperature, moderate quantity of chloride, sulphate, calcium, and the last one (C), the cold underground water. The primary hot spring A is a weak common salt spring, its orifice temperature being 47.4°C and the total amount of chemical constituents 6.6 g./l. The fact that springs containing a large quantity of this primary spring water are all located at the seaside, indicates some relation between the water of this primary spring and sea water. The primary hot spring B is a simple thermal, its orifice temperature being 54.8°C and the total amount of chemical constituents 0.085 g./l. The cold underground water has a temperature of 28°C, the total amount of its chemical constituents being 0.02 g./l. The percentage ratio l:m:n in the mixing of three primary springs A, B and C in each spring water, as calculated by T. Fukutomi and Z. Huzii, is shown in Table 12.

Assuming now the radium content of the underground water to be 0.0×10^{-12} g. Ra./l. and the percentage ratio l:m:n of each sample of water taken for radium determination to be as that given by Fukutomi, the writer calculated the radium content of the primary hot springs A and B from the data in Table 12 and obtained the following results.

Primary hot springs A contains 0.9×10^{-12} g. and B 0.2×10^{-12} g. of radium per liter of water.

Table 13.

Primary hot spring	Temp. of spring °C	Total chemical constituents g./l.	Radium content g./l.$\times10^{12}$
A	47.4	6.6	0.9
B	54.8	0.085	0.2
C (underground water)	28	0.02	0.0

Table 13 shows a general tendency for the radium content to increase with increasing amounts of the chemical constituents.

The writer next calculated the radium content of each spring water from their percentage ratio l:m:n, in the mixing of the 3 primary springs A, B and C, and their radium contents, with results as shown in Table 14, which calculated values agree fairly well with the observed values in each sample. This result, therefore, supports the theory of Fukutomi and Huzii.

(26) T. Fukutomi and Z. Huzii, *Bull. Earthq. Res. Inst.*, **15** (1937), 506.

Table 14. Comparison of the calculated and observed values for radium content of spring waters.

No.	Radium content g./l.×10^{12}		No.	Radium content g./l.×10^{12}	
	Calculated	Observed		Calculated	Observed
1	0.2	0.16	11	0.1	0.11
2	0.1	0.04	12	0.2	0.09
3	0.2	0.16	13	0.3	0.26
4	0.2	0.19	14	0.2	0.18
5	0.2	0.04	15	0.2	0.17
6	0.6	0.51	17	0.2	0.20
8	0.3	0.27	18	0.2	0.28
9	0.3	0.39	19	0.2	0.25
10	0.2	0.14	20	0.2	0.08

VII. Thermal and Mineral Springs of Arima, Hyōgo Prefecture. The spring waters from Arima were sampled by Y. Nemoto in November, 1936. The radium content of these waters are shown in Table 15, which also gives the total evaporated residue determined by him.

The next sampling was made by K. Yamasaki in August, 1939, the results of which are shown in Table 16. Their total residues were determined by Kurorda.

The geographical distribution of these springs is shown in Figs. 11 and 12, from which distribution the four springs in Arima are regarded as one group. The radium contents of these springs are plotted against their

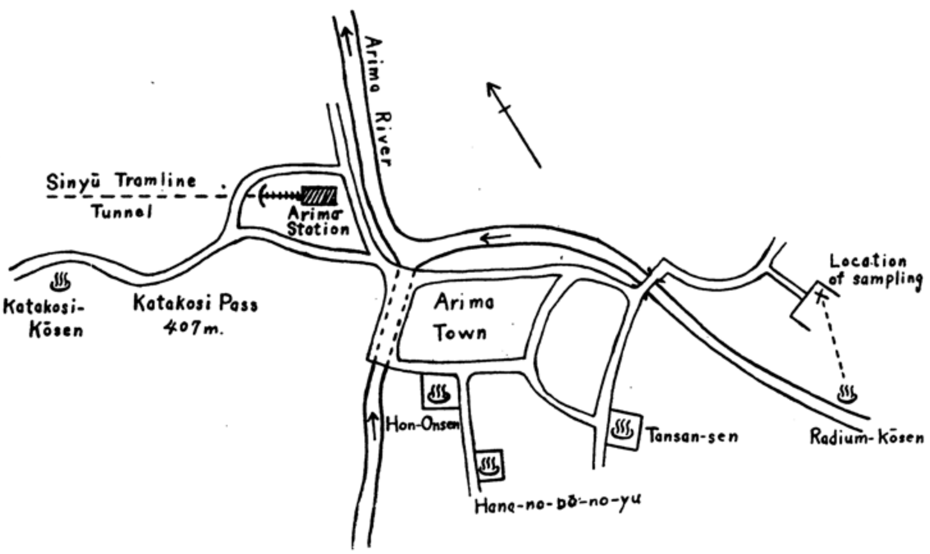


Fig. 11. Distribution of the Arima thermal and mineral springs. I.

Table 15. Thermal and mineral springs of Arima.

(Sampled November, 1936)

No.	Spring	Temp. of spring °C	Radium content g./l. ×10[12]	Total residue g./l. (Y. Nemoto)	Flow of water hl./day (Y. Nemoto)	Radium discharge 10^{-3} g. (mg.)/year
1	Arima, Hon-Onsen	—	71.29	18.43	1263	3.286
2	,, Hana-no-bō-no-yu	37.6	68.50	20.23	270	0.675
5	Arino, Katakosi-Kōsen	—	90.82	16.08	4	0.013
6	,, Yakendo-Kōsen	20.0	57.53	9.57	130	0.273

Table 16. Thermal and mineral springs of Arima.

(Sampled August, 1939)

No.	Spring	Temp. of spring °C	Temp. of air °C	Radium content g./l. ×10^{12}	Total residue g./l. (K. Kuroda)
1	Arima, Hon-Onsen	43.0	22.0	64.97	16.75
2	,, Hanano-bō-no-yu	34.5	22.0	47.08	17.38
3	,, Tansan-sen	16.0	22.0	0.76	
4	,, Radium-Kōsen*	27.5	22.0	12.68	
5	Arino, Katakosi-Kōsen	21.0	26.0	111.05	23.63
6	,, Yakendo-Kōsen	22.0	28.5	61.57	10.91

* Since the source of this spring was buried in the ground by a flood in July of 1938, water conveyed from the spring by means of a bamboo conduit pipe was sampled at Tanaka's villa, situated approximately 100 meters distance from the source. (V. Fig. 11)

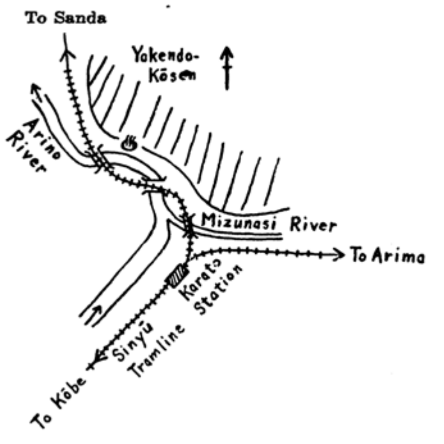


Fig. 12. Arima mineral springs. II.

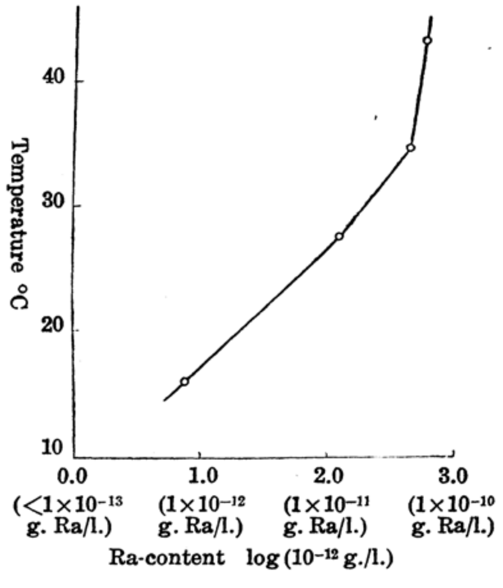


Fig. 13. Relation between the Ra-content and the orifice temperature of the Arima thermal and mineral springs.

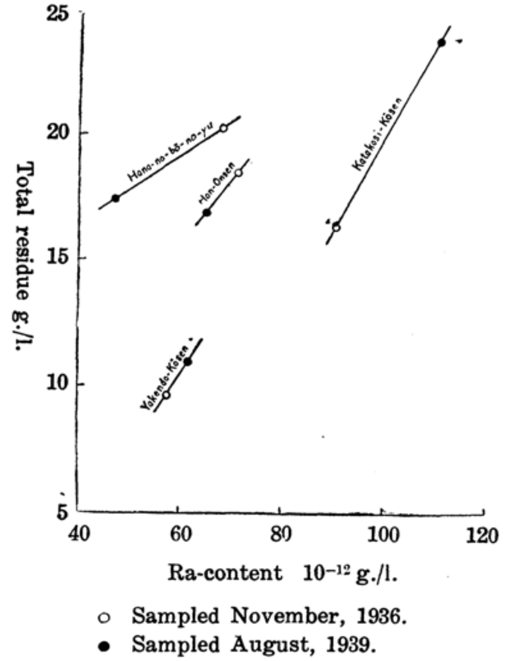


Fig. 14. Relation between the differences in the Ra-content and the amount of total residue in the Arima thermal and mineral springs.

orifice temperature in Fig 13. The relation between them appears to be simple linear.

Considering then the difference in the radium content of the four springs between the first and second determinations, it is interesting to note that, as will be seen from Tables 15 and 16, the differences run parallel to that of the amounts of total solid matter (V. Fig. 14).

From the daily flow of water measured by Nemoto in November, 1936, and their radium concentration, the radium discharge of the four springs was calculated, with results as given in Table 15. Since the springs of Arima Hon-Onsen have considerably high radium concentration and the flow of water is abundant, the quantity of radium brought up by this single spring to the surface of the earth amounts to more than 3 mg. per year.

VIII. Hot Springs of Beppu, Ōita Prefecture. The water samples of Beppu Hot Springs for radium determination were drawn in August and September, 1937, by the city office of Beppu. The orifice temperatures of these springs were measured at the time of sampling, with results as shown in Table 17. For some of them the pH value was determined by the Beppu Geophysical Research Laboratory[27] in November, 1927, and in February, 1928, the results of which are also given in this table for reference.

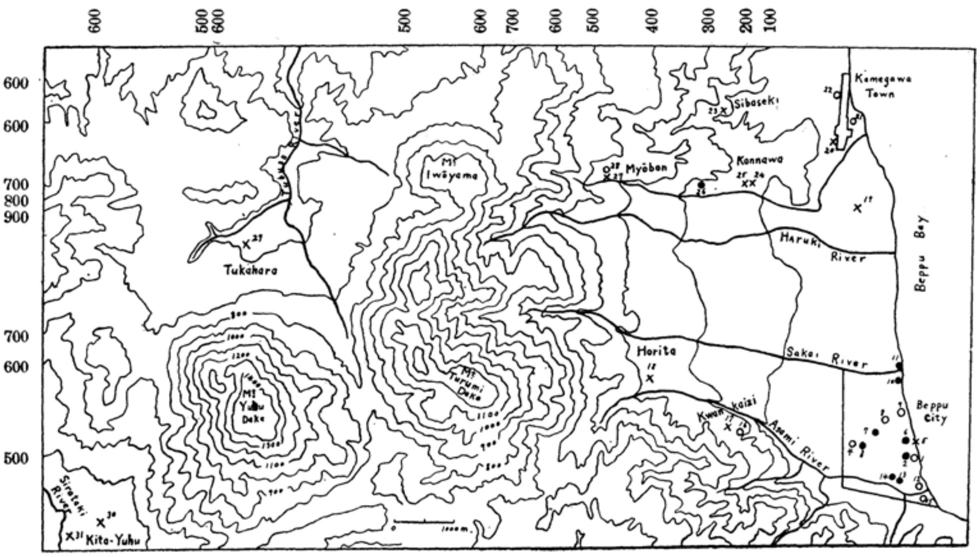
The geographical distribution of these 41 springs is shown in Fig. 15. According to S. Suzuki,[28] the hot springs of Beppu are, from their geographical and geological distributions, divided into three groups. The first is the group of the hot springs of Beppu, Kwankwaizi and Horita, the second that of Kamegawa, Kannawa and Myōban, and the third that of the hot springs in the basin of Yuhuin.

As will be seen from Table 17, the springs of the first group, which are all very weak acidic or very weak alkaline, show a somewhat high radium content compared with that of other groups, that of the second group, excluding the springs at Kamegawa, which are all strongly acidic

(27) *Repts. Beppu Geophys. Research Lab.*, **1** (1937), 78.
(28) S. Suzuki, *Repts. Beppu Geophys. Research Lab.*, **1** (1937), 6.

and usually contain small quantities of radium, and that of the third which shows the lowest radium concentration.

The fact that in each group of springs, their radium contents appear to bear no general relation to their orifice temperature, is believed to be due to other yet unknown factors.



• Hot spring containing more than 3×10^{-13} g. Ra/l.
○ Hot spring containing $1 \sim 3 \times 10^{-13}$ g. Ra/l.
× Hot spring containing less than 1×10^{-13} g. Ra/l.

1.	Reityō-sen	Beppu	15.	Hamawaki-Onsen	Beppu
2.	Kotobuki-Onsen	,,	16.	Sira-yu	Kwankaizi
	Yanagi-Onsen	,,	17.	Kwankaizi-Onsen	,,
	Kusunoki-Onsen	,,	18.	Horita-Onsen	Horita
3.	Hurō-sen	,,	19.	Bōtyō-sen	Kita-isigaki
4.	Ta-no-yu	,,	20.	Si-no-yu	Kamegawa
5.	Kaigan-suna-yu	,,	21.	Kiyō-sen	,,
6.	Takegawara-Onsen	,,	22.	Hamada-Onsen	,,
	Umezono-Onsen	,,	23.	Sibaseki-Onsen	Sibaseki
7.	Kitamati-Onsen	,,	24.	Netu-no-yu	Kannawa
8.	Kaimonzi-Kōen-Onsen	,,	25.	Sibu-yu	,,
9.	Yumimatu-Onsen	,,	26.	Umi-zigoku	,,
10.	Matogahama-Onsen	,,	27.	Zizō-Onsen	Myōban
11.	Yumigahama-Onsen	,,	28.	Kakuzyu-sen	,,
12.	Hinode-Onsen	,,		Yakusi-Onsen	,,
13.	Syōzyu-sen	,,	29.	Tukahara-Onsen	Tukahara
14.	Nagesi-Onsen	,,	30.	Yunotubo-Onsen	Yuhuin
	Kamiya-Onsen	,,	31.	Otomaru-Onsen	,,

Fig. 15. Geographical distribution of the amount of radium in the Beppu hot springs.

Table 17. Hot springs of Beppu.

No.	Location	Spring	Temp. of spring °C	Radium content g./l.×10¹²	Reaction pH	Chemical classification
			Group I			
1	Beppu	Reityō-sen	60.0	0.22	Very weak alkaline	Simple
2	,,	Kotobuki-Onsen	46.5	0.18	Weakly acidic	,,
3	,,	Yanagi-Onsen	49.5	0.64	Very weak alkaline	,,
4	,,	Kusunoki-Onsen	44.5	0.39	Very weak acidic	,,
5	,,	Hurō-sen	59.0	0.32	Weakly acidic	Iron carbonate
6	,,	Ta-no-yu A	57.0	0.18	,,	Simple
7	,,	,, B	43.0	0.15		,,
8	,,	Kaigan-suna-yu	58.5	0.04	Weakly acidic	Iron carbonate
9	,,	Takegawara-Onsen	64.5	0.74	,,	,,
10	,,	Umezono-Onsen	61.5	0.75	Very weak alkaline	Simple
11	,,	Kitamati-Onsen	55.0	0.30		
12	,,	Kaimonzi-Kōen-Onsen	62.0	0.25	Very weak alkaline	Simple
13	,,	Matogahama-Onsen	49.5	0.76	,,	,,
14	,,	Yumimatu-Onsen	62.0	0.25	,,	Alkaline
15	,,	Yumigahama-Onsen	49.0	0.59		
16	,,	Unsenzi-Onsen	51.0	0.21		
17	,,	Hinode-Onsen	50.5	0.17	Very weak alkaline	Simple
18	,,	Syōzyu-sen	47.0	0.48		
19	,,	Nagesi-Onsen	52.5	0.72	Very weak alkaline	Simple
20	,,	Kamiya-Onsen	51.0	0.11		Simple carbon dioxated
21	,,	Hamawaki-Onsen	46.5	0.14	Very weak alkaline	Common salt
22	Kwankaizi	Sira-yu	43.0	0.22		
23	,,	Kwankaizi-Onsen	55.5	0.00	7.4	Simple carbon dioxated
24	Horita	Horita-Onsen	81.0	0.09	7.6	Sulphur
			Group II			
25	Kita-isigaki	Bōtyō-sen	63.0	0.00		
26	Kamegawa	Si-no-yu	54.0	0.00	Very weak alkaline	Weak common salt
27	,,	Kiyō-sen	56.8	0.11	Weakly acidic	,,
28	,,	Hamada-Onsen	80.5	0.24	Neutral	Carbonated
29	,,	Gomusō-Onsen	45 0	0.04		
30	,,	Suzi-yu	58.0	0.00		
31	Sibaseki	Sibaseki-Onsen	67.0	0.00	3.2	Iron carbonate
32	Kannawa	Netu-no-yu	48.0	0.00	5.2	Simple
33	,,	Sibu-yu	91.0	0.00	Acidic	Acid vitriol

Table 17.—(*Concluded*)

No.	Location	Spring	Temp. of spring °C	Radium content g./l. × 10¹²	Reaction pH	Chemical classification
34	Kannawa	Umi-zigoku	90.5	0.54	1.6	Muriated acid vitriol
35	Myōban	Zizō-Onsen	60.0	0.00	Strong acidic	Sulphurated acid
36	,,	Kakuzyu-sen	70.0	0.27	,,	,,
37	,,	Yakusi-Onsen	87.0	0.27	,,	Acid alum vitriol
		Group III				
38	Yuhuin	Tanakaiti-Onsen	58.0	0.00		Simple
39	,,	Yunotubo-Onsen	58.0	0.09		,,
40	,,	Otomaru-Onsen		0.06		
41	Tukahara	Tukahara-Onsen	48.0	0.00		

IX. Relation between the Radium and the Radon Contents of Spring Waters. In this study, samples of water from 477 different mineral springs were tested for radium concentration. Some of them have been also measured for radon content by the present writer and other investigators. The radium and radon contents of these springs are shown in Table 18, arranged in the order of their radon contents.

The radon content of the mineral waters at Masutomi in Yamanasi Prefecture; Kasio, Itozawa and Tadati in Nagano Prefecture; and Ena, Itigaku, Yogarasu, Naegi and Kasagi in Gihu Prefecture, were measured by the writer by means of the IM Fontactoscope. The data of the other springs were obtained by R. Ishizu, Y. Kinugasa, H. Kibezaki(1); S. Matuura(29) of the Kyūsyū Imperial University; K. Siratori(30) of the Tōhoku Imperial University; Y. Hattori(31) of the Tokyo Imperial Hygienic Laboratory and others.

Although the radon contents obtained by the writer were measured from the same sample that was used for radium determination, in the other cases some time elapsed between the date when the waters were tested for radon content and that when the samples were drawn for radium determination. Since the radon concentration may vary more or less with lapse of time, the old measurements are likely to differ from what they are today, although we shall not be very far out in estimating approximately the rough values for the radon content of each sample from these data.

According to Carl Genser(32), the radioactive natural waters are divided into three types.

(1) The Radium-salt-water type, containing radium and radon in their radioactive equilibrium proportions (e.g. Heidelberg).

(2) The Radium-emanation-water type, containing no dissolved

(29) S. Matuura, I. Iwasaki and R. Hukusima, *J. Chem. Soc. Japan*, **61**(1940), 225.
(30) K. Siratori, *Science Repts. Tōhoku Imp. Univ.*, **16** (1925).
(31) Y. Hattori, *Bull. Imp. Hygienic Lab.*, **52** (1939), 141.
(32) C. Genser, Z. deutsch. geol. Gesell. **1933**, 482.

Table 18. Relation between the radium and the radon contents of spring waters.

No.*	Spring	Location	Prefecture	Radium content g./l.×10^{12}	Radon per liter of water			
					in 10^{-10} curies	in Mache's units	Examined by	Date
346	Radium-Kōsen No. 2	Ikeda	Simane	19.46	5384	1479	S. Matuura	Oct. 1939
175	Wadegawara No. 2	Masutomi	Yamanasi	11.62	4889	1343	T. Nakai	Oct. 1936
174	,, No. 1	,,	,,	6.76	2139	588	,,	,,
183	Kuridaira No. 1	,,	,,	0.77	2097	576	,,	,,
169	Ōsiba	,,	,,	2.68	1318	362	,,	,,
187	Kuridaira No. 3	,,	,,	6.56	1290	355	,,	Apr. 1937
180	Nibuzawa	,,	,,	6.22	1203	330	,,	Oct. 1936
184	Kuridaira No. 1. B.	,	,,	0.73	1108	304	,,	,,
247	Radium-Kōsen	Ena	Gihu	0.0	808	222	,,	May 1936
186	Yunokubo No. 1	Masutomi	Yamanasi	7.56	736	202	,,	Oct. 1936
345	Radium-Kōsen No. 1	Ikeda	Simane	36.43	721	198	S. Matuura	Oct. 1939
181	Sio-no-sawa-tugane-yu	Masutomi	Yamanasi	1.33	701	193	T. Nakai	Oct. 1936
315	Yamadaku-no-yu	Misasa	Tottori	3.65	613	169	Y. Hattori	Oct. 1935
170	Ginsentō-huru-yu	Masutomi	Yamanasi	9.39	556	153	T. Nakai	Oct. 1936
298	Ryōyōzyo-gensen	Misasa	Tottori	2.89	473	130	Y. Hattori	Oct. 1935
273	Radium-Kōsen	Arima	Hyōgo	12.68	440	121	R. Nomitu of the Imp. Univ. of Kyōto	Sept. 1931
171	Ginsentō-kami-no-yu	Masutomi	Yamanasi	7.35	378	104	T. Nakai	Oct. 1936
308	Seitō-kwan-no-yu	Misasa	Tottori	0.14	311	85.6	Y. Hattori	Oct. 1935
291	Hanaya-no-yu	,,	,,	0.85	271	74.4	,,	,,
304	Aburaya-soto-yu	,,	,,	0.21	236	64.7	K. Shiratori	Aug. 1925
297	Akazakiya-no-yu	,,	,,	5.25	194	53.4	Y. Hattori	Oct. 1935

* Numbers correspond to those used in Table 3.

Table 18.—(*Continued*)

No.*	Spring	Location	Prefecture	Radium content g./l.×10^{12}	Radon per liter of water			
					in 10^{-10} curies	in Mache's units	Examined by	Date
131	Spring No. 1	Murasugi	Niigata	0.51	180	49.6	Y. Kinugasa.	Aug. 1914
302	Eirakuan-no-yu	Misasa	Tottori	0.37	177	48.8	Y. Hattori	Oct. 1935
132	Spring No. 2	Murasugi	Niigata	0.04	177	48.6	Y. Kinugasa	Aug. 1914
312	Tennen-gankutu-no-yu No. 1	Misasa	Tottori	10.06	156	42.9	Y. Hattori	Oct. 1935
173	Ginsentō-simo-no-yu	Masutomi	Yamanasi	3.42	153	42.2	T. Nakai	Oct. 1936
133	Spring No. 3	Murasugi	Niigata	0.04	147	40.3	Y. Kinugasa	Aug. 1914
326	Tama-no-yu	Sekigane	Tottori	0.10	141	38.6	Y. Hattori	Sept. 1935
348	Yugakai-Onsen No. 2	Yugakai	Simane	30.57	140	38.5	S. Matuura	Oct. 1939
289	Private bath (M. Yamamoto)	Misasa	Tottori	0.25	132	36.3	K. Shiratori	Aug. 1925
299	Iwa-yu (Otoko-yu)	,,	,,	2.95	131	36.0	R. Ishizu	Mar. 1914
347	Yugakai-Onsen No. 1	Yugakai	Simane	35.06	128	35.3	S. Matuura	Oct. 1939
300	Iwa-yu (Onna-yu)	Misasa	Tottori	3.61	126	34.5	Y. Hattori	Oct. 1935
284	Seirei-no-yu	Katimi	,,	0.23	122	33.6	,,	Mar. 1937
321	Kabu-yu	Sekigane	,,	0.11	121	33.1	,,	Sept. 1935
323	Kame-no-yu	,,	,,	0.14	120	33.0	,,	,,
322	Kami-no-yu	,,	,,	0.13	113	31.1	,,	,,
306	Kiya-kazoku-yu	Misasa	,,	3.72	103	28.4	R. Ishizu	Mar. 1914
288	Otyaya-no-yu	,,	,,	0.28	102	27.9	,,	Feb. 1914
194	Totikubo No. 1	Masutomi	Yamanasi	32.62	93.2	25.6	T. Nakai	Oct. 1936
58	Mamegara-hudō-no-yu	Sirakawa	Hukusima	1.27	89.2	24.5	Ishikawa Middle school	Nov. 1930
292	Nakaya-no-yu	Misasa	Tottori	0.90	89.2	24.5	R. Ishizu	Mar. 1914
293	Bun-aburaya-no-yu	,,	,,	2.24	89.2	24.5	,,	,,

252	Syō-no-yu	Kasagi	Gihu	0.0	70.0	19.3	T. Nakai	May 1936
305	Iwasaki-no-yu No. 2	Misasa	Tottori	2.81	70.0	19.3	Y. Hattori	Oct. 1935
303	Aburaya-uti-yu	”	”	0.89	69.9	19.2	R. Ishizu	Mar. 1914
176	Hattyōdaira No. 1	Masutomi	Yamanasi	0.11	63.7	17.5	T. Nakai	Oct. 1936
248	Itigaku-Kōsen	Itigaku	Gihu	0.0	60.1	16.5	”	May 1936
251	Sika-no-yu	Kasagi	”	0.0	59.6	16.4	”	”
198	Yuzawa No. 1	Masutomi	Yamanasi	71.81	58.4	16.1	”	Apr. 1937
296	Saiki-honkwan-no-yu	Misasa	Tottori	2.37	57.9	15.9	K. Shiratori	Aug. 1925
294	Hasizuya-no-yu	”	”	1.00	56.4	15.5	R. Ishizu	Mar. 1914
309	Iwasaki-no-yu No. 1	”	”	0.37	54.7	15.1	Y. Hattori	Oct. 1935
295	Yakusidō-no-yu	”	”	3.82	52.7	14.5	”	”
325	Tokiwa-no-yu	Sekigane	”	0.28	46.2	12.7	”	Sept. 1935
59	Moto-yu No. 1	Bobata	Hukusima	0.11	43.5	12.0	S. Hanzawa	Sept. 1914
172	Ginsentō-naka-no-yu	Masutomi	Yamanasi	0.43	42.0	11.6	T. Nākai	Oct. 1936
290	Naka-no-yu	Misasa	Tottori	0.35	40.0	11.0	R. Ishizu	Feb. 1914
182	Gantō-hunsen	Masutomi	Yamanasi	2.46	39.5	10.9	T. Nakai	Oct. 1936
250	Kitatani-no-izumi	Naegi	Gihu	0.0	37.9	10.4	”	May 1936
287	Kabu-yu	Misasa	Tottori	1.14	37.1	10.2	R. Ishizu	Feb. 1914
179	Higasiobi-no-izumi	Masutomi	Yamanasi	35.65	32.4	8.90	T. Nakai	Oct. 1936
286	Nakataya-no-yu	Katimi	Tottori	0.73	31.2	8.57	R. Ishizu	Feb. 1914
167	Itozawa-no-izumi	Itozawa	Nagano	0.0	29.4	8.00	T. Nakai	May 1936
285	Sagi-no-yu	Katimi	Tottori	1.93	29.0	7.98	Y. Hattori	Mar. 1937
185	Kuridaira-tennen-buro	Masutomi	Yamanasi	26.05	27.1	7.40	T. Nakai	Oct. 1936
191	Tuganerō-kasikiri-no-yu	”	”	23.95	23.7	6.50	”	Apr. 1937
193	Umamiti-zawa	”	”	4.93	23.3	6.40	”	Oct. 1936
208	Wada-matuba-Kōsen	”	”	82.66	22.2	6.10	”	Apr. 1937
280	Suzukiya-no-yu	Hamamura	Tottori	1.63	19.5	5.35	R. Ishizu	Feb. 1914
188	Tuganerō No. 1	Masutomi	Yamanasi	64.53	17.8	4.90	T. Nakai	Oct. 1936
55	Moto-yu	Kasi	Hukusima	0.53	16.7	4.59	Y. Kinugasa	Nov. 1913

Table 18.—(*Continued*)

No.*	Spring	Location	Prefecture	Radium content g./l.×10^{12}	Radon per liter of water: in 10^{-10} curies	Radon per liter of water: in Mache's units	Radon per liter of water: Examined by	Radon per liter of water: Date
366	Yōzyō-yu	Dōgō	Ehime	0.03	16.1	4.42	H. Kibezaki	July 1913
189	Tuganerō No. 2	Masutomi	Yamanasi	29.39	14.6	4.00	T. Nakai	Oct. 1936
365	Kami-no-yu	Dōgo	Ehime	0.08	14.5	3.98	H. Kibezaki	July 1913
206	Siobuti-no-yu	Masutomi	Yamanasi	11.06	14.2	3.90	T. Nakai	Oct. 1936
281	Tabakoya-no-yu	Hamamura	Tottori	2.40	14.2	3.89	R. Ishizu	Feb. 1914
57	Yuzin-no-yu	Kasi	Hukusima	0.75	12.3	3.38	Y. Kinugasa	Nov. 1913
195	Kinsentō	Masutomi	Yamanasi	29.30	12.0	3.30	T. Nakai	Oct. 1936
168	Siogasawa-Kōsen	Tadati	Nagano	0.0	11.9	3.28	,,	Nov. 1936
154	Zinbara-ryokwan-no-yu	Awara	Hukui	0.56	11.7	3.21	Y. Kinugasa	Dec. 1913
327	Yōzyō-kwan-no-yu	Tōgō	Tottori	0.84	11.2	3.07	R. Ishizu	Feb. 1914
56	Tengu-no-yu	Kasi	Hukusima	0.41	10.4	2.86	Y. Kinugasa	Nov. 1913
344	Koyabara-Onsen	Koyabara	Simane	6.15	8.88	2.44	S. Matuura	Oct. 1939
211	Simo-no-daira-no-yu	Masutomi	Yamanasi	13.00	8.37	2.30	T. Nakai	Apr. 1937
164	Me-no-yu	Asama	Nagano	0.06	8.26	2.27	Y. Kinugasa	Aug. 1914
8	Toyotomi-Onsen	Toyotomi	Hokkaidō	10.10	8.08	2.22	Hokkaidō Ind. Research Lab.	July 1934
275	Yakendo-Kōsen	Arima	Hyōgo	57.53	8.01	2.20	Y. Nemoto	Nov. 1936
279	Simo-no-yu	Yosioka	Tottori	0.61	7.72	2.12	Y. Hattori	Feb. 1937
108	Yubiso-no-yu No. 2	Yubiso	Gumma	0.30	7.17	1.97	M. Komori	Apr. 1913

243	Seko-no-yu	Yugasima	Sizuoka	0.10	7.03	1.93	Y. Kinugasa	Mar. 1913
283	Kyōdō-yu	Hamamura	Tottori	0.60	6.92	1.90	R. Ishizu	Feb. 1914
209	Siozawa-kuromori-Kōsen	Masutomi	Yamanasi	11.57	6.55	1.80	T. Nakai	Apr. 1937
272	Tansan-sen	Arima	Hyōgo	0.76	6.34	1.75	H. Kibezaki	May 1913
359	Kawa-no-yu	Tawarayama	Yamaguti	0.00	6.21	1.71	,,	Aug. 1914
207	Nakazima-no-izumi	Masutomi	Yamanasi	2.94	6.19	1.70	T. Nakai	Apr. 1937
204	Siokawa-asiai-no-yu	,,	,,	1.20	5.46	1.50	,,	,,
210	Siozawa-kuromori-yosino-kwan	,,	,,	0.00	4.37	1.20	,,	,,
249	Yogarasu-Kōsen	Yogarasu	Gihu	0.0	3.93	1.08	,,	May 1936
38	Kyōdō-yokuzyō-no-yu	Atumi	Yamagata	0.58	3.68	1.01	H. Kakehi	June 1914
53	Tansan-sen	Naruko	Miyagi	0.06	3.64	1.00	Phys. Institute of Tōhoku Imp. Univ.	
212	Kamase-hudō-no-yu	Masutomi	Yamanasi	0.00	3.64	1.00	T. Nakai	Apr. 1937
373	Hunagoya-Kōsen	Hunagoya	Hukuoka	0.00	3.43	0.94	H. Kibezaki	Aug. 1914
351	Sin-sagi-no-yu	Yunogō	Okayama	0.50	3.09	0.85	R. Ishizu	July 1913
350	Sagi-no-yu	,,	,,	0.62	3.06	0.84	,,	Mar. 1914
277	Iwai-Onsen	Iwai	Tottori	0.56	2.87	0.79	Y. Hattori	Dec. 1937
276	Kyū-Onsen	Takarazuka	Hyōgo	26.46	2.66	0.73	Ōsaka Imp. Hyg. Lab.	Sept. 1913
87	Nomi-yu	Ikao	Gumma	0.00	2.44	0.67	Y. Kinugasa	Nov. 1913
90	Ohaguro-no-yu	,,	,,	0.09	1.86	0.51	,,	,,
260	Kosi-no-yu No. 1	Katuura	Wakayama	0.19	1.53	0.42	T. Iwasaki and K. Kuroda	Nov. 1939
205	Siokawa-nisida-no-yu	Masutomi	Yamanasi	0.0	1.46	0.40	T. Nakai	Apr. 1937
92	Ōseki-no-yu	Ikao	Gumma	0.00	1.35	0.37	Y. Kinugasa	Nov. 1913

Table 18.—(*Concluded*)

No.*	Spring	Location	Prefecture	Radium content g./l.×10^{12}	Radon per liter of water in 10^{-10} curies	in Mache's units	Examined by	Date
340	Ameya-ryokwan-no-yu	Usio	Simane	0.06	1.19	0.33	R. Ishizu	Mar. 1914
401	Kiyō-sen	Kamegawa	Ōita	0.11	1.16	0.32	,,	Oct. 1913
405	Sibaseki-Onsen	,,	,,	0.00	1.13	0.31	,,	,,
235	Ō-yu	Yosina	Sizuoka	0.10	1.09	0.30	Y. Kinugasa	Mar. 1913
217	Matubara-no-yu	Itō	,,	0.16	1.06	0.29	R. Ishizu	Feb. 1913
160	Yamasio-Kōsen	Kasio	Nagano	18.5	0.93	0.26	T. Nakai	May 1936
342	Moto-yu	Sigaku	Simane	1.02	0.93	0.26	S. Matuura	Oct. 1939
270	Hon-Onsen	Arima	Hyōgo	71.29	0.92	0.25	H. Kibezaki	May 1913
98	Sirahata-no-yu	Kusatu	Gumma	0.16	0.80	0.22	R. Ishizu	Apr. 1913
400	Si-no-yu	Kamegawa	Ōita	0.00	0.80	0.22	,,	Oct. 1913
379	Hurō-sen	Beppu	,,	0.32	0.66	0.18	H. Kibezaki	Mar. 1913
94	Tíyò-no-yu	Kusatu	Gumma	0.10	0.62	0.17	R. Ishizu	Apr. 1913
97	Netu-no-yu	,,	,,	0.15	0.58	0.16	,,	,,
244	Kidati-no-yu	Yugasima	Sizuoka	0.30	0.55	0.15	Y. Kinugasa	Mar. 1913
96	Zizō-no-yu	Kusatu	Gumma	0.01	0.51	0.14	R. Ishizu	Apr. 1913
93	Wasi-no-yu	,,	,,	0.05	0.47	0.13	,,	,,
409	Zizō-Onsen	Myōban	Ōita	0.00	0.47	0.13	,,	Oct. 1913
253	Iwō-sen	Gero	Gihu	0.00	0.44	0.12	Tokyo Imp. Hyg. Lab.	Nov. 1934
402	Hamada-Onsen	Kamegawa	Ōita	0.24	0.40	0.11	R. Ishizu	Oct. 1913
254	Yunosima-Onsen	Gero	Gihu	0.00	0.36	0.10	Tokyo Imp. Hyg. Lab.	Sept. 1934
130	Senami-Onsen	Senami	Niigata	0.11	0.22	0.06	Y. Kinugasa	Aug. 1914
397	Kwankaizi-Onsen	Kwankaizi	Ōita	0.00	0.07	0.02	H. Kibezaki	Mar. 1913

radium, only gaseous emanation (most of the famous radioactive springs in Germany belong to this type).

(3) The Radium-emanation-water type, containing radium-salts in which the amount of dissolved radium is less than that required for radioactive equilibrium with the radon that is present in this water (e.g. Oberschlema, Brambach, Kreuznach, etc.).

The hot springs of Arima Hon-Onsen show a radium content of 0.71×10^{-10} g. per liter of water and a radon content of 0.92×10^{-10} curie per liter. The radium is almost in radioactive equilibrium with radon, but since the radon content of this spring was determined in 1913 and the radium content in 1936, there is more than a twenty years internal between the two determinations, with the result that it may be unsafe to compare such data. Assuming, however, that its radon content did not vary much during this interval of time, this spring would belong to the Radium-salt-water type.

The Ena spring contains 8.08×10^{-10} curie of radon per liter, although no dissolved radium can be detected in this water. This spring is a typical Radium-emanation-water.

As will be seen from Table 18, in almost every case the amount of radium in the spring waters was found to be much less than that required for radioactive equilibrium with the radon that is present in them. Even in the water from the Wadamatuba spring in Masutomi, which shows the highest radium content,(33) the radon content was found to be 22.2×10^{-10} curie and the amount of radium found was 0.83×10^{-10} g. per liter of water, showing that the radon in the water is about 27 times greater than that which the radium present in the water is able to produce. In other words, the permanent activity of this water is only 4 per cent of its temporary activity.

Thus all the radioactive natural waters in Japan, with only one exception, belong to the class of Radium-emanation-water or Radium-emanation-water with radium-salt, which shows that although a part of the radon in the water of mineral springs is formed directly from their parent radium dissolved in them, the greater part of them might have been extracted independently of the radium from the rocks where the waters originate and through which they pass. The difference between the amount of radon and that of radium in natural waters may be due to the difference between the solubility of gaseous radon in water and that of the radium salts.

The correlation between the radium and radon contents is shown in Fig. 16, in which the radium contents of the spring waters that were examined by the writer are plotted against their radon contents. Here we see that spring waters of high radium content do not always show a high radon content, while those of high radon concentration do not always contain a large amount of radium. The amounts of radium and of radon in spring waters do not always run parallel with each other. For example, the waters of the Wadamatuba spring in Masutomi, which shows the

(33) Although the Katakosi spring in Arima and the Matu-no-yu in Tamatukuri show higher radium content than this spring, they have never been tested for radon.

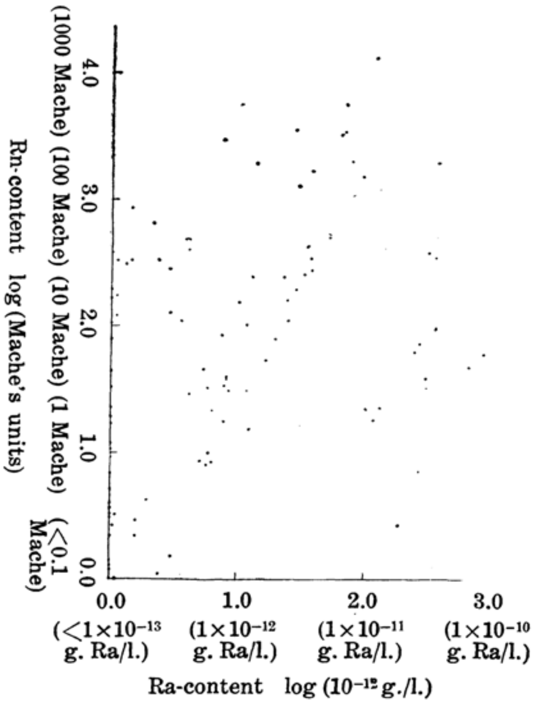


Fig. 16. Relation between the Ra-content and the Rn-content of Japanese mineral springs.

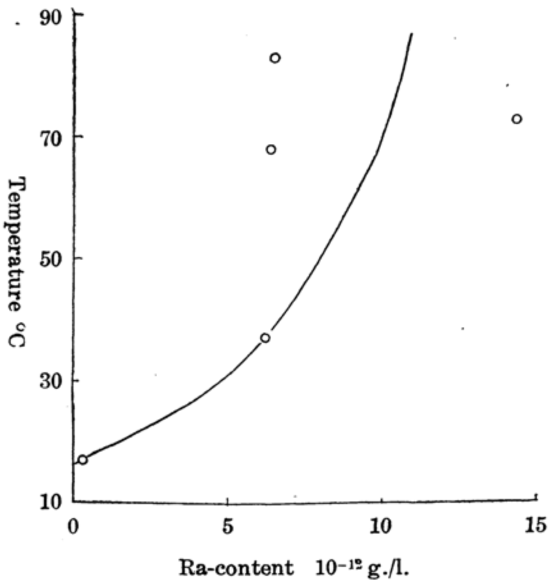


Fig. 17. Relation between the Ra-content and the orifice temperature of the Matunoyama hot springs.

highest radium content of 82.66×10^{-12} g. per liter of water, contain only 6.1 Mache's units of radon, while the waters of Ikeda spring No. 2 and that of Wadegawara No. 2, both show an unusually high concentration of radon, do not contain so much radium as the others, which shows, as already stated, that radon from spring waters does not originate in dissolved radium alone, but also from other sources.

X. Relation between the Radium Content and the Orifice Temperature of Mineral Springs. We shall now take some groups of springs and study the relation between their radium content and their orifice temperature.

The correlation between the radium content and the orifice temperature of the Masutomi springs in Yamanasi Prefecture, that of the Misasa springs in Tottori Prefecture, and that of the Arima springs in Hyōgo Prefecture, has been shown in the preceding figures (V. Table 10, Figs. 9, 10, 14). In Figs. 17-21, the radium content of the spring waters is plotted against their orifice temperature for the Matunoyama springs in Niigata Prefecture; Katuura in Wakayama Prefecture; Hamamura (including Katimi) in Tottori Prcfecture; and Ikeda and Yugakai in Simane Prefecture.

As will be seen from these figures, there seems

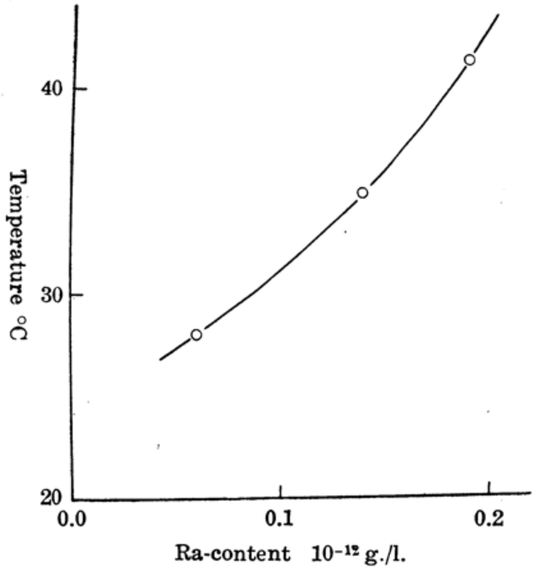


Fig. 18. Relation between the Ra-content and the orifice temperature of the Katuura thermal springs.

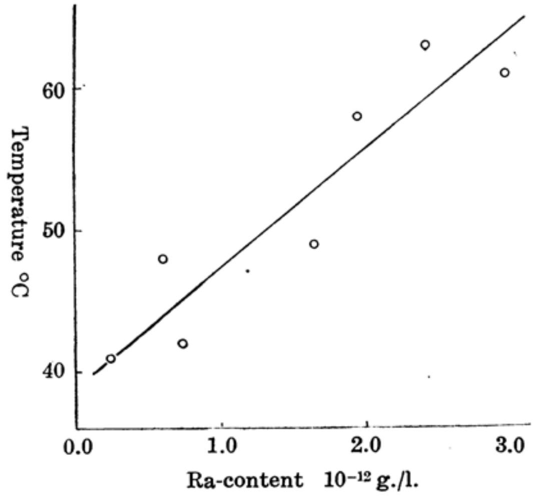


Fig. 19. Relation between the Ra-content and the orifice temperature of the Hamamura (Katimi) hot springs.

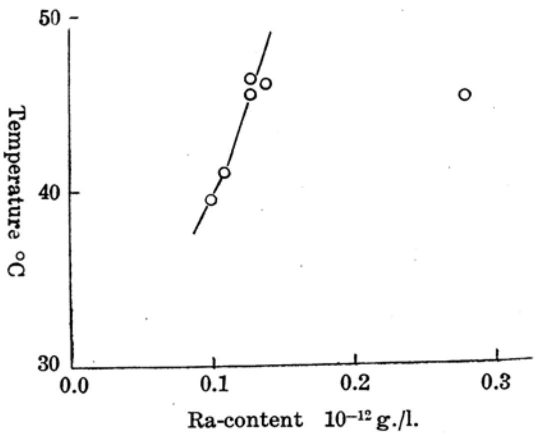


Fig. 20. Relation between the Ra-content and the orifice temperature of the Sekigane hot springs.

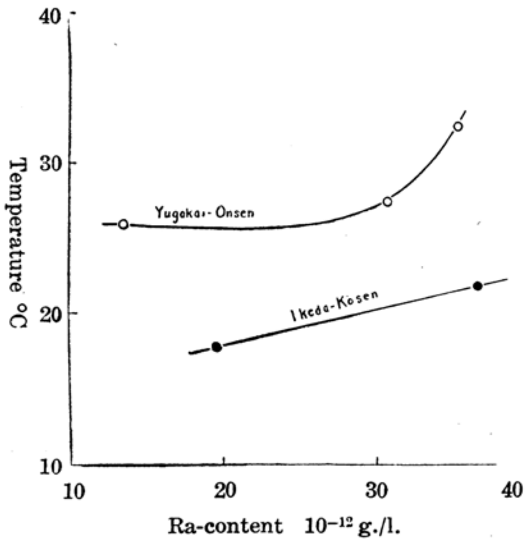


Fig. 21. Relation between the Ra-content and the orifice temperature of the Yugakai thermal springs and the Ikeda mineral springs.

to be a linear relation between them, the radium content increasing with rise in orifice temperature in almost every group, which may be explained by the difference in the solubility of radium by temperature, assuming that the radium contained in spring waters comes mostly from rocks where the waters originate and through which they pass.

As stated in the previous chapter, however, in the case of the Beppu springs in Oita Prefecture, no such correlation exists between their radium contents and orifice temperature. It may be affected by other unknown factors more pronounced than that of temperature.

XI. Relation between the Radium Content and the Chemical Qualities of Spring Waters. *Chemical classification of mineral springs and their radium contents.* According to the system arranged by the Pharmaceutical Society of Japan, the mineral springs are classified as below.

1. Simple cold.
2. Simple.
3. Simple carbondioxated.
4. Earthy carbondioxated.
5. Alkaline.
6. Common salt.
7. Bitter.
8. Iron carbonate.
9. Vitriol.
10. Alum.
11. Alum vitriol.
12. Acid.
13. Sulphur.

In Table 19, the mineral springs are arranged according to their chemical composition.

Chemical analyses of these mineral springs were made by the Imperial Hygienic Laboratories of Tokyo and Ōsaka.(34) The data of the springs starred were obtained by the Hygienic Laboratories of the respective Prefectures.

Although some of these data are old, it is believed that the main chemical constituents of the springs do not vary much so far as no great changes occur at their origins, so that we may presume from these results the chemical classification to which any water sample taken for radium determination belongs to. The total evaporated residues of these waters and those of some of their essential chemical constituents are also given in this table for reference.

As will be seen from this table, almost all the simple cold springs contain no radium. The simple thermals also show a low radium content, being lower than 1×10^{-12} g. radium per liter of water, excluding the waters at Yakusidō-no-yu in Misasa, which is a radioactive spring containing 14.50 Mache units of radon. Thus the simple springs in which the quantity of dissolved solid constituents is less than 1 gram per kilogram, are all poor in radium. As will be described in the next section, there appears to be a linear relation between the radium content of spring waters and their total solid constituents, with the result that spring waters containing small quantities of solid matter are usually poor in radium.

All sulphur springs show comparatively low radium content. It is well known that sulphur springs are closely related to volcanos and it is also evident from numerous measurements that volcanic rocks are generally very poor in radium compared with other igneous rocks, such as granite, etc., from all which one naturally expects sulphur springs to be poor in radium.

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(34) A Table of Composition of Mineral Waters in Japan, Bull. Imp. Hygienic Lab., **34** (1929); **54** (1940) and other reports of the Imperial Hygienic Laboratories.

Table 19. Mineral springs arranged according to chemical composition.

1. Simple cold springs.

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content g./l. × 10^{12}	Total residue g./Kg.	Radio-activity	Mache's units
76	Naka-no-yu*	Yokomuki	Hukusima	24.0	0.00	0.64		
133	Spring No. 3*	Murasugi	Niigata	13.5	0.04		Radioactive	40.3
167	Itozawa-no-izumi*	Itozawa	Nagano	13.0	0.0			
168	Siogasawa-Kōsen	Tadati	,,	12.0	0.0			
247	Radium-Kōsen	Ena	Gihu	13.1	0.0	0.12	Radioactive	22.2
248	Itigaku-Kōsen*	Itigaku	,,	12.1	0.0		,,	16.5
249	Yogarasu-Kōsen*	Yogarasu	,,	14.0	0.0			
250	Kitatani-no-izumi*	Naegi	,,	12.3	0.0		Radioactive	10.4

2. Simple thermals.

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content g./l. × 10^{12}	Total residue g./Kg.	Radio-activity	Mache's units
19	Tuta-Onsen*	Tuta	Aomori		0.24			
25	Yuze-Onsen*	Yuze	Akita		0.00			
44	Ō-yu*	Aone	Miyagi	54.0	0.55	0.54		
55	Moto-yu	Kasi	Hukusima	51.0	0.53			
56	Tengu-no-yu	,,	,,	48.5	0.41			
57	Yuzin-no-yu	,,	,,	50.0	0.75			
66	Private bath* (K. Hasimoto)	Iwasiro-atami	,,		0.05	0.39		

** Numbers correspond to those used in Table 3.

Table 19.—(*Continued*)

2. Simple thermals. (*Concluded*)

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content g./l.×10^{12}	Total residue g./Kg.	Radio-activity	Mache's units
68	Private bath* (M. Ōgosi)	Iwasiro-atami	Hukusima	35.0	0.08	0.38		
79	Moto-yu	Kawazi	Totigi	46.0	0.10	0.33		
105	Minakami-Onsen	Minakami	Gumma	40.0	0.49	0.42		
107	Yubiso-Onsen No. 1	Yubiso	,,	50.0	0.00			
108	,, No. 2	,,	,,	66.0	0.30	0.55		
123	Ubako-no-yu	Ubako	Kanagawa	39.7	0.05	0.65		
131	Spring No. 1*	Murasugi	Niigata	25.6	0.51	0.35	Radioactive	49.6
132	,, No. 2*	,,	,,	26.0	0.04		,,	48.6
164	Me-no-yu	Asama	Nagano	36.5	0.06	0.44		
165	Higasiyama-Onsen*	Higasiyama	,,	42.0	0.08			
215	Yukawa-no-yu	Itō	Sizuoka	44.0	0.16	0.92		
217	Matubara-no-yu	,,	,,	42.0	0.16	0.98		
236	Moto-yu	Yosina	,,	39.0	0.06	0.48		
239	Hōsen-Onsen	,,	,,	47.5	0.02	0.65		
241	Kan-no-yu	,,	,,	51.0	0.00	0.82		
295	Yakusidō-no-yu	Misasa	Tottori	74.5	3.82	0.86	Radioactive	14.5
309	Iwasaki-no-yu No. 1	,,	,,	54.5	0.37	0.64	,,	15.1
322	Kami-no-yu	Sekigane	,,	46.3	0.13	0.50	,,	31.1
323	Kame-no-yu	,,	,,	46.0	0.14	0.58	,,	33.0
325	Tokiwa-no-yu	,,	,,	45.3	0.28	0.65	,,	12.7
326	Tama-no-yu	,,	,,	39.5	0.10	0.69	,,	38.6
327	Yōzyō-kwan-no-yu	Tōgō	,,	50.0	0.84	0.93		
354	Mizuno-ryokwan-no-yu*	Yuda	Yamaguti		0.22			
358	Mati-no-yu	Tawarayama	,,	40.0	0.00	0.18		
359	Kawa-no-yu	,,	,,	40.0	0.00	0.18		
360	Syō-no-yu	,,	,,	38.0	0.00	0.19		

365	Kami-no-yu*	Dōgo	Ehime	46.8	0.08	0.78		
366	Yōzyō-yu*	,,	,,	45.4	0.03	0.83		
367	Donkobori-no-yu*	,,	,,		0.03			
371	Spring No. 2*	Musasi	Hukuoka		0.17			
375	Reityō-sen*	Beppu	Ōita	60.0	0.22	0.97		
376	Kotobuki-Onsen*	,,	,,	46.5	0.18	1.35		
377	Yanagi-Onsen*	,,	,,	49.5	0.64	0.72		
378	Kusunoki-Onsen*	,,	,,	44.5	0.39	0.89		
380	Ta-no-yu A*	,,	,,	57.0	0.18	0.70		
381	,, B*	,,	,,	43.0	0.15			
384	Umezono-Onsen*	,,	,,	61.5	0.75	1.00		
386	Kaimonzi-Kōen-Onsen*	,,	,,	62.0	0.25	1.17		
387	Matogahama-Onsen*	,,	,,	49.5	0.76	1.62		
391	Hinode-Onsen*	,,	,,	50.5	0.17	0.77		
393	Nagesi-Onsen*	,,	,,	52.5	0.72			
406	Netu-no-yu*	Kannawa	,,	48.0	0.00	0.42		
436	Tokyoya-no-yu	Takeo	Saga	52.0	0.89			
437	Huruyu-Onsen*	Huruyu	,,		0.05			
477	Kamii-kwan-no-yu	On-yō	Korea	46.0	0.00	0.21		

3. Simple carbondioxated springs.

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content g./l.×10^{12}	Total residue g./Kg.	Free CO_2 g./Kg.	
53	Tansan-sen	Naruko	Miyagi	39.0	0.06			
272	Tansan-sen*	Arima	Hyōgo	16.0	0.76			
394	Kamiya-Onsen*	Beppu	Ōita	51.0	0.11	0.72		
397	Kwankaizi-Onsen*	Kwankaizi	,,	55.5	0.00	0.77	0.59	
441	Moto-yu-Onsen*	Yunoura	Kumamoto	45.0	0.04	0.32	0.05	
470	Ramune-Onsen No. 1*	Ramune	Kagosima		0.20	2.45	1.22	
471	,, No. 2*	,,	,,		0.18	1.08	1.62	

Table 19.—(*Continued*)

4. Earthy carbondioxated springs.

5. Alkaline springs.

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content g./l.×10^{12}	Total residue g./Kg.	Free CO_2 g./Kg.	NaCl g./Kg.
			a. Alkaline springs.					
262	Yukawarō-no-yu*	Yukawa	Wakayama	40.2	0.10			
263	Kawayu-Onsen*	Kawayu	,,	56.0	2.53			
267	Kami-no-yu*	Ryūzin	,,	48.0	0.57			
268	Simo-no-yu*	,,	,,		0.15			
269	Tomiya-ryokwan-no-yu*	Yumura	Hyōgo	88.0	0.79			
312	Tennen-gankutu-no-yu No. 1	Misasa	Tottori	79.5	10.06	2.27		
369	Nibuzawa-Kōsen	Nibuzawa	Ehime		0.14			
388	Yumimatu-Onsen*	Beppu	Ōita	62.0	0.25			
			b. Alkaline carbondioxated spring.					
444	Iwa-no-yu-Onsen*	Yunoura	Kumamoto	45.0	0.06	0.35	0.79	
			c. Alkaline muriated springs.					
7	Ponyu-Onsen	Ponyu	Hokkaidō	37.0	0.00	1.87		0.57
264	Reimei-no-yu*	Yuzaki	Wakayama	57.3	8.44			
443	Hukamizu-Onsen*	Yunoura	Kumamoto	42.0	0.02	0.36		

6. Common salt springs.

No.**	Spring	Location	Prefecture	Temp. of spring °C	Rndium content g./l.×10¹²	Total residue g./Kg.	NaCl g./Kg.
			a. Weak common salt springs.				
125	Mamane-no-yu	Yugawara	Kanagawa	75.0	0.00	2.00	1.22
126	Simo-no-yu	,,	,,	63.0	0.10	1.99	1.20
130	Senami-Onsen	Senami	Niigata	100.0	0.11	4.19	3.37
183	Kuridaira No. 1	Masutomi	Yamanasi	22.0	0.77	3.62	1.40
184	,, No. 1. B.	,,	,,	17.0	0.73	2.32	
298	Ryōyōzyo-gensen	Misasa	Tottori	72.0	2.89	1.31	0.61
300	Iwa-yu (Onna-yu)	,,	,,	68.0	3.61	1.23	0.57
305	Iwasaki-no-yu No. 2	,,	,,	68.0	2.81	1.16	0.71
342	Moto-yu*	Sigaku	Simane	45.0	1.02	3.14	1.52
395	Hamawaki-Onsen*	Beppu	Ōita	46.5	0.14		3.13
400	Si-no-yu	Kamegawa	,,	54.0	0.00	1.22	0.68
401	Kiyō-sen*	,,	,,	56.8	0.11	2.65	
416	Ō-yu*	Yunohira	,,	92.0	0.24	1.58	0.89
417	Hana-no-yu-Onsen*	,,	,,	78.0	0.19	1.58	0.97
			b. Simple common salt spring.				
			c. Concentrated common salt springs.				
160	Yamasio-Kōsen	Kasio	Nagano	12.1	18.5	29.45	26.81
270	Hon-Ȯnsen	Arima	Hyōgo	43.0	71.29	18.43	
			d. Carbondioxated common salt springs.				
146	Nagae-Kōsen*	Nagae	Toyama	21.0	0.42	2.32	1.01
345	Radium-Kōsen No. 1	Ikeda	Simane	21.7	36.43	9.83	5.60

Table 19.—(*Continued*)

6. Common salt springs. (*Concluded*)

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content $g./l. \times 10^{12}$	Total residue g./Kg.	NaCl g./Kg.	SO_4'' g./Kg.
			d. Carbondioxated common salt springs. (*Concluded*)					
346	Radium-Kōsen No. 2*	Ikeda	Simane	17.7	19.46			
476	Yuno-o-Onsen*	Yuno-o	Kagosima	74.0	1.53	2.27	0.58	
			e. Alkaline common salt springs					
8	Toyotomi-Onsen	Toyotomi	Hokkaidō	43.5	10.10	11.70	9.82	
102	Alkali-sen	Isobe	Gumma	15.5	15.53	26.91	21.35	
347	Yugakai-Onsen No. 1	Yugakai	Simane	32.3	35.06	10.35	6.66	
440	Gozen-no-yu*	Hinaku	Kumamoto	48.0	0.08	0.70	0.37	
445	Kotobuki-Onsen*	Yunoura	,,	41.0	0.02	0.67	0.42	
			f. Saline common salt springs.					
40	Kamasaki-no-yu	Kamasaki	Miyagi	47.0	1.00	5.19	2.47	1.37
41	Mogamiya-no-yu	,,	,,	38.5	1.37	5.03	2.41	
42	Kimuraya-no-kyū-yu	,,	,,	39.0	3.03	5.15	2.42	1.37
43	Kimuraya-no-sin-yu	,,	,,		2.58	5.14	2.42	1.35
77•	Iwasakiya-no-yu	Ōsio	Hukusima		3.72	4.56	2.88	0.52
			g. Earth-muriated common salt springs.					
26	Onogawa-Onsen	Onogawa	Yamagata	76.0	8.37	5.67	3.90	
27	Kyokuato-gensen	Akayu	,,	62.0	0.09			
28	Matusima-kwan-no-yu	,,	,,	54.0	0.09			
129	Monkawa-Onsen	Monkawa	Kanagawa	45.0	0.41	21.97	15.84	

134	Netu-no-yu	Matunoyama	Niigata	68.0	6.43	15.08	8.68	
135	Hii-no-yu	,,	,,	83.0	6.49	13.87	7.93	
136	Kagami-no-yu	,,	,,	72.5	14.30	15.50	7.97	
147	Gotairan-no-yu	Wakura	Isikawa	82.0	6.04	21.00	11.41	
148	Moto-yu*	,,	,,		5.89			
149	Sin-yu*	,,	,,		4.83			
271	Hana-no-bō-no-yu	Arima	Hyōgo	37.6	68.50	20.23		
274	Katakosi-Kōsen	,,	,,	21.0	111.05	23.63		
350	Sagi-no-yu	Yunogō	Okayama	38.0	0.62	2.27	1.18	
351	Sin-sagi-no-yu	,,	,,	37.7	0.50	2.25	1.17	
448	Mura-no-yu-Onsen*	Ibusuki	Kagosima	65.0	0.89	5.11	3.34	
449	Yaziga-yu-Onsen*	,,	,,	56.0	0.55	4.08	2.74	
	h. Earthy common salt springs.							
174	Wadegawara No. 1	Masutomi	Yamanasi	18.0	6.76	3.25		
175	,, No. 2	,,	,,	19.5	11.62	7.12		
180	Nibuzawa	,,	,,	24.0	6.22	7.25	4.98	
181	Sio-no-sawa-tugane-yu	,,	,,	20.0	1.33	4.89	5.29	
194	Totikubo No. 1	,,	,,	27.0	32.62	9.42		
195	Kinsentō*	,,	,,	31.0	29.30	7.49	3.51	
	i. Sulphated common salt springs							
38	Kyōdō-yokuzyō-no-yu	Atumi	Yamagata		0.58	3.51	2.35	0.57
128	Moti-no-yu	Yugawara	Kanagawa	59.0	0.28	2.28	1.29	0.49
154	Zinbara-ryokwan-no-yu	Awara	Hukui	76.0	0.56	10.03	5.90	0.39
156	Hatu-yu	Sibu	Nagano	73.5	0.08			
157	Ō-yu No. 1	,,	,,	77.0	0.11	1.21	0.53	0.37
158	,, No. 2	,,	,,		0.03			

Table 19.—(*Continued*)

7. Bitter springs.

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content g./1.×10¹²	Total residue g./Kg.	SO_4'' g./Kg.	
			a. Bitter springs.					
438	Spring No. 1*	Tosita	Kumamoto		0.04			
439	,, No. 2*	,,	,,		0.10			
			b Saline bitter springs.					
22	Usi-no-yu*	Yumoto	Iwate		0.08	1.44		
153	Yamasiro-Onsen*	Yamasiro	Isikawa	66.0	0.13			
235	Ō-yu	Yosina	Sizuoka	50.0	0.10	1.24	0.64	
245	Sagasawa-Onsen	Sagasawa	,,	58.0	0.21	1.62	0.78	
340	Ameya-ryokwan-no-yu*	Usio	Simane	43.0	0.06	1.10		
			c. Sulphated bitter springs.					
15	Sin-yu*	Sinyu	Aomori	60.0	0.05	1.64		
34	Gensen*	Akakura	Yamagata	65.0	0.12			
47	Kawara-no-yu*	Sakunami	Miyagi	54.0	0.21			
106	Tanikawa-Onsen	Tanikawa	Gumma	62.0	0.10	1.02	0.49	
109	Asahi-no-yu	Hōsi	,,	44.0	0.00	1.20	0.72	
112	Sarugakyō-Onsen	Yuzima	,,	59.0	0.55	1.79		
277	Iwai-Onsen	Iwai	Tottori	51.0	0.56	1.22	0.96	
285	Sagi-no-yu	Katimi	,,	58.0	1.93	0.98	0.41	
			d. Earthy sulphated bitter springs.					
87	Nomi-yu	Ikao	Gumma	44.0	0.00	1.16	0.29	
88	Spring No. 2	,,	,,	49.0	0.00	1.08	0.21	
90	Ohaguro-no-yu	,,	,,	50.0	0.09	1.46	0.32	
91	Hukiage-no-yu	,,	,,	41.0	0.00	1.65	0.39	

92	Ōseki-no-yu	Ikao	Gumma	47.0	0.00	1.03	0.27	
104	Yuhara-Onsen	Yuhara	,,	55.0	0.09	1.47	0.57	
			e. Muriated saline bitter springs.					
280	Suzukiya-no-yu	Hamamura	Tottori	49.0	1.63	1.07		
286	Nakataya-no-yu	Katimi	,,	42.0	0.73	1.36	0.46	
			f. Muriated sulphated bitter springs.					
127	Hirokawara-no-yu*	Yugawara	Kanagawa	68.0	0.05	4.79		
284	Seirei-no-yu	Hamamura	Tottori	41.0	0.23	1.64	0.63	

8. Iron carbonate springs.

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content $g./l.\times 10^{12}$	Total residue g./Kg.	NaCl g./Kg.	CO_2 g./Kg.
			a. Iron carbonate springs.					
379	Hurō-sen*	Beppu	Ōita	59.0	0.32	0.80		
382	Kaigan-suna-yu*	,,	,,	58.5	0.04	0.99		
383	Takegawara-Onsen*	,,	,,	64.5	0.74	0.97		
405	Sibaseki-Onsen*	Kamegawa	,,	67.0	0.00	1.59		
			b. Earthy iron carbonate spring.					
89	Spring No. 3	Ikao	Gumma	47.5	0.06	0.82		
			c. Iron carbonate common salt springs.					
143	Moto-yu	Seki	Niigata	41.0	0.09	1.06	0.98	
344	Koyabara-Onsen*	Koyabara	Simane	38.3	6.15	8.03	3.91	
450	Misuzi-no-yu*	Ibusuki	Kagosima	58.0	0.51	3.69	2.36	

Table 19.—(*Continued*)

8. Iron carbonate springs. (*Concluded*)

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content g./1.×10^{12}	Total residue g./Kg.	NaCl g./Kg.	CO_2 g./Kg.
	d. Iron carbondioxated common salt spring.							
276	Kyū-Onsen	Takarazuka	Hyōgo	18.5	26.46	18.80	15.26	
	e. Iron carbondioxated springs.							
257	Yosino-Onsen	Yosino	Nara		0.43	0.83		1.38
373	Hunagoya-Kōsen No. 2*	Hunagoya	Hukuoka	22.0	0.00	0.36		1.69
434	Nagaiki-no-yu*	Nagayu	Ōita	42.0	0.58	2.70		2.55

9. Vitriol springs.

10. Alum springs.

11. Alum vitriol springs.

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content g./1.×10^{12}	Total residue g./Kg.		
72	Spring No. 1*	Wasikura	Hukusima	50.0	0.00			

12. Acid springs.

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content g./1.×10^{12}	Total residue g./Kg.	H_2SO_4 (free) g./Kg.	HCl (free) g./Kg.	H_2S g./Kg.
	a. Acid springs								
6	Kawayu-Onsen*	Kawayu	Hokkaidō		0.00				
11	Netu-no-yu*	Sukayu	Aomori	50.0	0.32	1.09	0.10		
12	Hie-no-yu*	,,	,,	48.0	0.15	0.95	0.13		

			b. Acid vitriol springs.						
93	Wasi-no-yu	Kusatu	Gumma	60.0	0.05	4.09			
407	Sibu-yu*	Kannawa	Ōita	91.0	0.00	1.71		0.08	
			c. Acid alum vitriol spring.						
98	Sirahata-no-yu	Kusatu	Gumma	61.0	0.16		2.51		
			d. Muriated acid alum vitriol spring.						
408	Umi-zigoku	Kannawa	Ōita	90.5	0.54	3.51		0.22	
			e. Sulphureted acid springs.						
24	Okama-no-yu*	Tamagawa	Akita		0.60				
75	Numaziri-Onsen	Numaziri	Hukusima	63.0	0.84	1.94	0.17	0.52	0.13
86	Sika-no-yu	Nasu	Totigi	76.0	0.55	2.14	1.07		0.04
409	Zizō-Onsen*	Myōban	Ōita	60.0	0.00	1.02	0.59	0.00	0.01
410	Kakuzyu-sen*	,,	,,	70.0	0.27	5.88	2.21	0.02	0.002
			f. Sulphureted acid alum vitriol spring.						
33	Kawara-no-yu*	Takayu	Yamagata	42.0	0.23	4.02	1.68		

13. Sulphur springs.

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content g./l.×10^{12}	Total residue g./Kg.	H_2S g./Kg.	
			a. Sulphur springs.					
59	Moto-yu No. 1*	Bōbata	Hukusima	14.0	0.11	0.18		
150	Hōsi-no-yu*	Awazu	Isikawa	64.0	0.21	2.35	0.02	
251	Sika-no-yu*	Kasagi	Gihu	15.1	0.0			
253	Iwō-sen	Gero	,,	52.0	0.00	0.32	0.002	
254	Yunosima-Onsen*	,,	,,	65.0	0.00			
370	Spring No. 1*	Musasi	Hukuoka		0.15			
398	Horita-Onsen*	Horita	Ōita	81.0	0.09	0.36	0.01	

Table 19.—(*Concluded*)

13. Sulphur springs. (*Concluded*)

No.**	Spring	Location	Prefecture	Temp. of spring °C	Radium content g./l. $\times 10^{12}$	Total residue g./Kg.	H_2S g./Kg.	
			b. Hydrogen sulphide springs.					
21	Hanamaki-Onsen	Hanamaki	Iwate	72.0	0.00	0.96	0.002	
141	Myōkō-Onsen*	Myōkō	Niigata	60.0	0.24			
145	Moto-yu*	Tubame	,,		0.00			
266	Sin-tubaki-no-yu*	Tubaki	Wakayama	33.5	0.26	0.39	0.05	
454	Kabusiki-Onsen*	Kabusiki	Kagosima	52.0	0.00	0.36	0.04	
455	Private bath* (K. Huzita)	Miyanozyō	,,	52.0	0.06	0.39	0.002	
466	Hayasida-Onsen*	Kirisima-Eino-o	,,	71.0	0.39			
			c. Alkaline sulphur springs.					
138	Tamago-no-yu*	Matunoyama	Niigata	17.0	0.29			
259	Kowase-no-yu*	Katuura	Wakayama	28.0	0.06			
260	Kosi-no-yu No. 1*	,,	,,	41.5	0.19			
261	Obatakeyama-no-yu*	,,	,,	35.0	0.13			
			d. Muriated sulphur springs.					
20	Nanasigure-Kōsen	Nanasigure	Iwate		0.60	13.33	0.01	
38	Kyōdōyokuzyō-no-yu	Atumi	Yamagata		0.58	3.51		
159	Yamada-Onsen*	Yamada	Nagano	60.0	0.25	4.75		
290	Naka-no-yu	Misasa	Tottori	66.0	0.35	1.24	0.002	
			e. Earthy sulphur spring.					
85	Kakkō-Onsen*	Nasu	Totigi	60.0	0.00	1.79	0.02	

We shall next consider springs containing comparatively large amounts of sulphate ions, such as bitter, alum, vitriol and acid springs, some of which show a fairly high radium content. As for the relation between the radium and sulphate ions in mineral waters, I. D. Kurbatov,[2] who studied a number of salt waters from the petroleum district in North Caucasus, Russia, reported that the amount of sulphate ions in the water is not an indicator of the presence or absence of radium and its isotopes and that brines of unusually high radium concentration contain also some sulphate ions. According to the analyses of S. Gōda, brines from Tzeliutsing in Szechwan, China, notwithstanding its unusually high radium content of 2.5×10^{-10} g. per liter of water, contains 0.147% of sulphate ions. Table 20 gives the amount of sulphate ions[34] in some Japanese springs containing fairly large quantities of radium. As will be seen from this table, springs with high radium content are not always poor in sulphate ions.

Finally, common salt springs are generally rich in radium, especially spring waters containing a large quantity of sodium chloride, all of which, with a few exceptions, show high radium concentration. This cannot be easily explained, because the origin of the sodium chloride in spring waters is not yet clearly understood.

Relation between the radium content and the quantities of dissolved solid constituents of spring waters. The radium content and the quantities of dissolved solid constituents in spring waters were shown in Tables 7, 11, 16, 19.

Table 20. Sulphate ion content of spring waters of high radium content.

No.*	Spring	Location	Prefecture	Radium content g./l. $\times 10^{12}$	SO_4'' g./Kg.
8	Toyotomi-Onsen	Toyotomi	Hokkaidō	10.10	0.001
26	Onogawa-Onsen	Onogawa	Yamagata	8.37	0.093
40	Kamasaki-no-yu	Kamasaki	Miyagi	1.00	1.371
42	Kimuraya-no-kyū-yu	,,	,,	3.03	1.374
43	Kimuraya-no-sin-yu	,,	,,	2.58	1.348
77	Iwasakiya-no-yu	Ōsio	Hukusima	3.72	0.516
102	Alkali-sen	Isobe	Gumma	15.53	0.000
134	Netu-no-yu	Matunoyama	Niigata	6.43	0.094
136	Kagami-no-yu	,,	,,	14.30	0.241
147	Gotairan-no-yu	Wakura	Isikawa	6.04	0.191
160	Yamasio-Kōsen	Kasio	Nagano	18.5	0.000
180	Nibuzawa	Masutomi	Yamanasi	6.22	0.470
181	Sio-no-sawa-tugane-yu	,,	,,	1.33	0.472
276	Kyū-Onsen	Takarazuka	Hyōgo	26.46	0.000
285	Sagi-no-yu	Katimi	Tottori	1.93	0.408
298	Ryōyōzyo-gensen	Misasa	,,	2.89	0.092
347	Yugakai-Onsen No. 1	Yugakai	Simane	35.06	0.442

* Numbers correspond to those used in Table 3.

Although the amount of total evaporated residues determined by Okauti, Nemoto and Kuroda were obtained for the same samples that were used for radium determination, the other data obtained by the Tokyo and Ōsaka Imperial Hygienic Laboratories and the Hygienic Laboratories of the various Prefectures, are all old results. We can presume, however, from these data the approximate quantities of dissolved solid constituents for each water sample. In Fig. 22, the radium contents of the springs are plotted against their total residues.

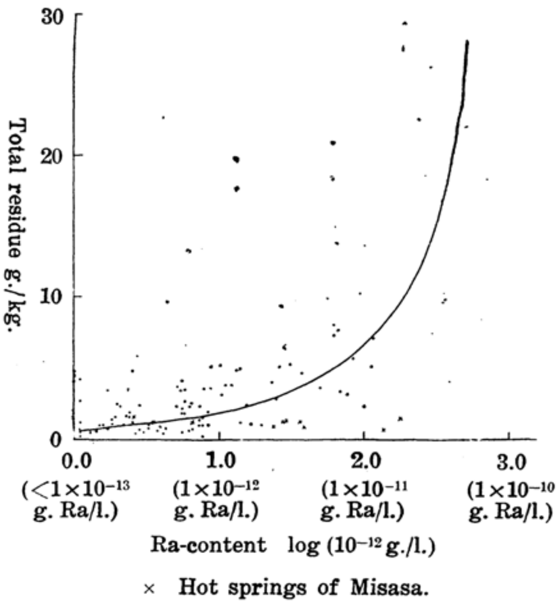

Fig. 22. Relation between the Ra-content and the amount of total residue of Japanese springs.

As will be seen from this figure, the radium content seems to increase with increasing quantities of total residues. All the concentrated mineral waters, for example, which contain large quantities of dissolved solid constituents exceeding 5 grams per kilogram of water with a few exceptions show a fairly high radium content, while all the dilute ones, which contain only small qualities of dissolved solid constituents, less than 1 gram per kilogram of water, show a low radium content of less than 1×10^{-12} g. per liter of water. It is, however, notable that the waters at Misasa have a high radium content notwithstanding the small quantities of dissolved solid constituents.

Relation between the radium content and the p*H value of spring waters.* The radium content and the pH value of spring waters are given in Table 21, arranged according to their pH values, which latter were determined by Kuroda[35] and the writer from the samples that were used for radium determination with the colorimetric method. Some old results obtained by Hattori,[31] the Beppu Geophysics Research Laboratory[27] and the Imperial Hygienic Laboratory of Tokyo[36] are given in the table for reference. Besides these data, the chemical reactions of spring waters of comparatively high radium content, are shown in Table 22.

As shown in these tables, all spring waters of high radium content seem to belong to the weak acidic, neutral or weak alkaline waters, while the strong acidic and the strong alkaline waters show a fairly low radium content.

In the case of strong acidic spring waters, it may be presumed that

(35) These unpublished data are reported here by permission of Mr. K. Kuroda, for which courtesy the writer tenders his sincere thanks.
(36) Reports of the Imperial Hygienic Laboratory of Tokyo.

acidic gases, derived from magma very deep in the earth, ascend in the state of vapour and reach the superficial crustal layer, where some of it turn liquid owing to the effect of underground water and then gush out to the surface of the earth. Should they pass through the rocks as solution from the beginning, the waters might act on the rocks with the result that the waters would lose their strong acidity. Assuming then that the largest part of the radium contained in spring waters comes from the rocks where the waters originate and through which they pass, as stated in the previous section, the above fact that strong acidic spring waters show a fairly low radium content is easily understood.

As to the strong alkaline spring waters, the strong alkalinity is due to many reasons, so that the low radium concentration of strong alkaline spring waters can not be explained simply.

Table 21. The radium content and the *p*H value of spring waters.

No.†	Spring	Location	Prefecture	Radium content g./l. $\times 10^{12}$	*p*H
98	Sirahata-no-yu	Kusatu	Gumma	0.16	1.55‡
408	Umi-zigoku	Kannawa	Ōita	0.54	1.6**
124	Yoemon-no-yu	Yunohana-zawa	Kanagawa	0.17	2.3*
	Kōbō-no-yu	,,	,,	0.19*	2.8*
	Gongen-no-yu	,,	,,	0.13*	2.9*
405	Sibaseki-Onsen	Kamegawa	Ōita	0.00	3.2**
123	Ubako-no-yu	Ubako	Kanagawa	0.05	3.3‡
	Huezuka-no-yu	Yunohana-zawa	,,	0.00*	3.5*
88	Spring No. 2	Ikao	Gumma	0.00	5.2***
406	Netu-no-yu	Kannawa	Ōita	0.00	5.2**
91	Hukiage-no-yu	Ikao	,,	0.00	5.4***
87	Nomi-yu	,,	,,	0.00	5.6***
92	Ōseki-no-yu	,,	,,	0.00	5.6***
343	Abura-yu	Sigaku	Simane	0.80	5.9
183	Kuridaira No. 1	Masutomi	Yamanasi	0.77	6.0*
342	Moto-yu	Sigaku	Simane	1.02	6.0
345	Radium-Kōsen No. 1	Ikeda	,,	36.43	6.0
89	Spring No. 3	Ikao	Gumma	0.06	6.1***
344	Koyabara-Onsen	Koyabara	Simane	6.15	6.1
90	Ohaguro-no-yu	Ikao	Gumma	0.09	6.2***
175	Wadegawara No. 2	Masutomi	Yamanasi	11.62	6.2*
316	Kōyōen-no-yu	Misasa	Tottori	16.06	6.2
346	Radium-Kōsen No. 2	Ikeda	Simane	19.46	6.2
348	Yugakai-Onsen No. 2	Yugakai	,,	30.57	6.2
349	,, No. 3	,,	,,	13.45	6.2
179	Higasiobi-no-izumi	Masutomi	Yamanasi	35.65	6.3*

† Numbers correspond to those used in Table 3.
‡ Determined by the Imperial Hygienic Laboratory of Tokyo.
* Determined by Kuroda.
** Determined by the Beppu Geophysics Research Laboratory.
*** Determined by Hattori.

Table 21. – (*Continued*)

No.†	Spring	Location	Prefecture	Radium content g./l.×10^{12}	*p*H
194	Totikubo No. 1	Masutomi	Yamanasi	32.62	6.3*
195	Kinsentō	,,	,,	29.30	6.3*
313	Tennen-gankutu-no-yu No. 2	Misasa	Tottori	7.11	6.3
317	Sinsen-ryō-no-yu	,,	,,	2.12	6.3
169	Ōsiba	Masutomi	Yamanasi	2.68	6.4*
185	Kuridaira-tennenburo	,,	,,	26.05	6.4*
188	Tuganerō No. 1	,,	,,	64.53	6.4*
189	,, No. 2	,,	,,	29.39	6.4*
208	Wada-matuba-Kōsen	,,	,,	82.66	6.4*
312	Tennen-gankutu-no-yu	Misasa	Tottori	10.06	6.4
318	Misasa-kwan-no-yu	,,	,,	15.01	6.4
347	Yugakai-Onsen No. 1	Yugakai	Simane	35.06	6.4
170	Ginsentō-huru-yu	Masutomi	Yamanasi	9.39	6.5*
171	Ginsentō-kami-no-yu	,,	,,	7.35	6.5*
174	Wadegawara No. 1	,,	,,	6.76	6.5*
311	Onsen-hotel-no-yu	Misasa	Tottori	13.48	6.5
314	Ōhasi-ryokwan-soto-yu	,,	,,	15.95	6.6
315	Yamadaku-no-yu	,,	,,	3.65	6.6
172	Ginsentō-naka-no-yu	Masutomi	Yamanasi	0.43	6.7*
289	Private bath (M. Yamamoto)	Misasa	Tottori	0.25	6.7
291	Hanaya-no-yu	,,	,,	0.85	6.8
292	Nakaya-no-yu	,,	,,	0.90	6.8
298	Ryōyōzyo-gensen	,,	,,	2.89	6.8
299	Iwa-yu (Otoko-yu)	,,	,,	2.95	6.8
304	Aburaya-soto-yu	,,	,,	0.21	6.8
307	Haikyū-zyo-no-yu	,,	,,	2.32	6.8
319	Okayamaidai-ryōyōzyo-no-yu	,,	,,	17.67	6.8
287	Kabu-yu	,,	,,	1.14	6.9
288	Otyaya-no-yu	,,	,,	0.28	6.9
294	Hasizuya-no-yu	,,	,,	1.00	6.9
300	Iwa-yu (Onna-yu)	,,	,,	3.61	6.9
301	Iwa-yu (Makura-yu)	,,	,,	2.90	6.9
303	Aburaya-uti-yu	,,	,,	0.89	6.9
134	Netu-no-yu	Matunoyama	Niigata	6.43	7.0*
290	Naka-no-yu	Misasa	Tottori	0.35	7.0
293	Bun-aburaya-no-yu	,,	,,	2.24	7.0
297	Akazakiya-no-yu	,,	,,	5.25	7.0
306	Kiya-kazoku-yu	,,	,,	3.72	7.0
308	Seitō-kwan-no-yu	,,	,,	0.14	7.0
104	Spring No. 1	Minakami	Gumma	0.09	7.05‡

Table 21.—(*Continued*)

No.†	Spring	Location	Prefecture	Radium content g./l. × 10^{12}	*p*H
135	Hii-no-yu	Matunoyama	Niigata	6.49	7.1*
233	Private bath (Wakatuki)	Itō	Sizuoka	0.25	7.1*
286	Nakataya-no-yu	Katimi	Tottori	0.73	7.1
302	Eirakuan-no-yu	Misasa	,,	0.37	7.1
309	Iwasaki-no-yu No. 1	,,	,,	0.37	7.1
21	Hanamaki-Onsen	Hanamaki	Iwate	0.00	7.15‡
137	Usagiguti-Onsen	Matunoyama	Niigata	6.17	7.2*
136	Kagami-no-yu	,,	,,	14.30	7.2*
295	Yakusidō-no-yu	Misasa	Tottori	3.82	7.2
305	Iwasaki-no-yu No. 2	,,	,,	2.81	7.2
310	Bansuirō-no-yu	,,	,,	0.26	7.2
277	Iwai-Onsen	Iwai	,,	0.56	7.25***
222	Private bath (K. Iizima)	Itō	Sizuoka	0.27	7.3*
281	Tabakoya-no-yu	Hamamura	Tottori	2.40	7.3
284	Seirei-no-yu	Katimi	,,	0.23	7.3
224	Private bath (S. Naruto)	Itō	Sizuoka	0.14	7.4*
227	Arai-kwan-no-yu	,,	,,	0.26	7.4*
264	Reimei-no-yu	Yuzaki	Wakayama	8.44	7.4*
285	Sagi-no-yu	Katimi	Tottori	1.93	7.4
320	Gunzihogoin-ryōyōzyo-no-yu	Misasa	,,	0.64	7.4
321	Kabu-yu	Sekigane	,,	0.11	7.4
324	Turu-no-yu	,,	,,	0.13	7.4
397	Kwankaizi-Onsen	Kwankaizi	Ōita	0.00	7.4**
279	Simo-no-yu	Yosioka	Tottori	0.61	7.45***
129	Monkawa-Onsen	Monkawa	Kanagawa	0.41	7.5‡
220	Masuya-ryokwan-no-yu	Itō	Sizuoka	0.51	7.5*
221	Daitō-kwan-no-yu	,,	,,	0.17	7.5*
223	Hōsen-kwan-no-yu	,,	,,	0.39	7.5*
226	Enpanrō-no-yu	,,	,,	0.09	7.5*
296	Saiki-honkwan-no-yu	Misasa	Tottori	2.37	7.5
322	Kami-no-yu	Sekigane	,,	0.13	7.5
323	Kame-no-yu	,,	,,	0.14	7.5
326	Tama-no-yu	,,	,,	0.10	7.5
107	Yubiso-no-yu No. 1	Yubiso	Gumma	0.00	7.5‡
105	Spring No. 2	Minakami	,,	0.49	7.6‡
138	Tamago-no-yu	Matunoyama	Niigata	0.29	7.6*
232	Private bath (T. Suzuki)	Itō	Sizuoka	0.28	7.6*
325	Tokiwa-no-yu	Sekigane	Tottori	0.28	7.6
398	Horita-Onsen	Beppu	Ōita	0.09	7.6*
215	Yukawa-no-yu	Itō	Sizuoka	0.16	7.8*
236	Moto-yu	Yosina	,,	0.06	7.8‡
229	Tōkai-kwan-no-yu	,,	,,	0.17	7.9*

Table 21.—(*Concluded*)

No.†	Spring	Location	Prefecture	Radium content g./l.×10^12	pH
267	Kami-no-yu	Ryūzin	Wakayama	0.57	7.9†
217	Matubara-no-yu	Itō	Sizuoka	0.16	8.0*
218	Tennen-yu (Matubara)	,,	,,	0.19	8.0*
219	Tōkyō-kwan-no-yu	,,	,,	0.04	8.0*
225	Tennen-yu (Kusumi)	,,	,,	0.11	8.0*
228	Yōki-kwan-no-yu	,,	,,	0.18	8.0*
231	Private bath (M. Aoki)	,,	,,	0.20	8.0*
234	,, (Toyosima)	,,	,,	0.08	8.0*
230	,, (K. Satō)	,,	,,	0.27	8.1*
216	,, (T. Ōkawa)	,,	,,	0.04	8.2*
108	Yubiso-no-yu No. 2	Yubiso	,,	0.30	8.45‡
109	Asahi-no-yu	Hōsi	,,	0.00	8.45‡
260	Kosi-no-yu No. 1	Katuura	Wakayama	0.19	8.6*
259	Kowase-no-yu	,,	,,	0.06	9.1*
261	Obatakeyama-no-yu	,,	,,	0.13	9.1*
262	Yukawarō-no-yu	Yukawa	,,	0.10	9.2*
266	Sin-tubaki-no-yu	Tubaki	,,	0.26	9.2*

Table 22. Chemical reactions of spring waters of high radium content.

No.*	Spring	Location	Prefecture	Radium content g./l.×10^12	Reaction
8	Toyotomi-Onsen	Toyotomi	Hokkaidō	10.10	Very weak alkaline
40	Kamasaki-no-yu	Kamasaki	Miyagi	1.00	,,
42	Kimuraya-no-kyū-yu	,,	,,	3.03	,,
43	Kimuraya-no-sin-yu	,,	,,	2.58	Neutral
77	Iwasakiya-no-yu	Ōsio	Hukusima	3.72	,,
102	Alkali-sen	Isobe	Gumma	15.53	Very weak alkaline
147	Gotairan-no-yu	Wakura	Isikawa	6.04	,,
160	Yamasio-Kōsen	Kasio	Nagano	18.5	,,
276	Kyū-Onsen	Takarazuka	Hyōgo	26.46	Acidic

* Numbers correspond to those used in Table 3.

XII. Radium Content of Brines from Oil Fields. *Brines from oil fields.* The radium content of brines from the oil fields in Akita and Niigata Prefectures and that from the natural gas field with indications of oil, in Tiba Prefecture, is shown in Table 23. The results of chemical analysis of K. Hosino[37] of the Tokyo Imperial Industrial Research Institute are also shown in this table, where the composition is given in molar ratio, taking sodium as 100. The last column shows, for reference, the absolute concentration of the sodium (gram per liter) in each brine.

On the origin of the brine in oil fields, several theories have so far been published by a number of investigators.[38] Hunt,[39] in 1875, before any other investigator, suggested, from the fact that the brine water from Ontario has an unusually high content of calcium chloride and magnesium chloride, that it comes from ancient sea water that was embedded simultaneously with the formation of the petroleum deposit. In 1905, Lane[40] extended this idea more generally and published a well-known theory called the connate or fossil water theory, according to which the brine from an oil field is nothing else but the ancient sea water separated from its surroundings at the time of deposition of the sedimentary rocks, and now held fast in deeply buried rocks. On the other hand, the opinions of Washburne[41] and Richardson[42] differ somewhat in that their belief is that the salts contained in brine water did not originate from sea water as the forementioned investigators believed, but from some other salts derived from readily soluble basic magmas or salt deposits. These opinions, however, do not seem to be adequate for Japan, where geological conditions differ from those in other countries. It may therefore be mentioned here that the acceptable theories at present are those which are based on sea water as the origin, which, however, differs in the details.

With the assumption that Japanese brine waters have been derived from ancient sea waters, the radium content of these brines is compared with that of sea waters (V. Table 24). As will be seen from Tables 23 and 24, the former is much higher than the latter, some of these brine samples showing an unusually high radium content compared with spring waters. Is it to be considered that it is entirely due to radium dissolved out from contiguous soft rocks?

Comparing now the composition of sea waters and that of Japanese brines, marked similarities are found between them in the ratios of sodium, potassium, calcium and bromine, but much smaller quantities are observed in the magnesium and sulphate and much larger quantities in the iodine in the case of brine. As to the small content of sulphate in the brine from the oil fields, which is true not only for Japan but also for any other countries, many explanations have been offered by investigators, abroad, such as Höfner,[43] Rogers,[44] Palmer,[45] Mills and

(37) These unpublished data are reported here by permission of Mr. K. Hosino, to whom the author desires to express his sincere thanks for his courtesy.
(38) T. Ishikawa and T. Baba, this Bulletin, **53** (1932), 362.
(39) T. S. Hunt, Chemical and Geological Essays, Boston, (1875), 117.
(40) A. C. Lane, *J. Geol.*, **14** (1905), 221; *Geol. Soc. Am. Bull.*, **19** (1908), 501.
(41) C. H. Washburne, *Am. Inst. Eng. Trans.*, **48** (1904), 687.
(42) G. B. Richardson, *Econ. Geol.*, **12** (1917), 37.
(43) H. V. Höfner, Das Erdöl, II (1909), 28.
(44) G. S. Rogers, U. S. Geol. Surv. Bull., (1917), 653; U. S Geol. Surv., Prof. Paper, (1919), 117.
(45) C. Palmer, *Econ. Geol.*, (1924), No. 7.

Table 23. Radium content and chemical composition of Japanese brines.

No.	Oil field (Natural gas field)	Prefecture	No. of the petroleum well	Radium content g./l.×10^{12}	Total solid matter* g./Kg.	Composition of water*								
						Na	K	Ca	Mg	Cl	SO_4	Br	I	Na g./Kg.
1	Omonogawa	Akita	1	11.05	21.45	100	1.42	0.62	0.26	56.2	0.00	0.11	0.04	12.77
2	,,	,,	25	7.14	24.87	100	1.33	0.23	0.13	51.0	0.00	0.08	0.04	14.83
3	Araya	,,	1	10.02	22.44	100	1.40	0.46	0.65	54.3	0.00	0.08	0.02	13.14
4	Aburaden	,,	1	7.02	24.11	100	1.33	0.44	0.71	55.2	0.00	0.09	0.03	14.41
5	Oguni	Niigata	R 32	0.0	14.68	100	0.00	0.29	1.54	77.9	0.02	0.10	0.04	5.51
6	Oguti	,,	R 97	0.0	11.11	100	0.00	0.23	1.84	17.9	0.03	0.13	0.03	4.06
7	Ōmo	,,	R 4	12.8	29.17	100	1.36	0.16	0.15	105.8	0.00	0.16	0.04	10.46
8	Ōguti	,,	C 1	1.10	2.45	100	4.26	5.02	0.83	57.4	0.00	0.09	0.00	1.24
9	Yoita	,,		0.04	9.37	100	1.13	0.50	1.98	108.2	0.03	0.27	0.01	3.43
10	Takamati	,,	R 36	2.2	15.69	100	0.00	0.54	0.22	85.2	0.02	0.22	0.04	5.75
11	Iriwada	,,	R 5	1.5	22.91	100	0.00	3.81	0.56	115.2	0.01	0.21	0.06	7.94
12	Nagamine	,,	C 93	0.0	35.19	100	0.00	3.65	6.15	76.8	0.00	0.30	0.06	11.05
13	Okano	,,	R 1 (78 m.)	2.47	18.59	100	0.15	10.84	2.42	109.1	0.17	0.43	0.08	5.63
14	,,	,,	R 1 (108 m.)	2.84	23.35	100	0.00	9.60	4.48	138.8	0.02	0.38	0.10	6.96
15	Ōtaki	Tiba		0.0	12.45	100	1.05	0.63	2.14	72.2	0.03	0.19	5.20	4.48

* Analysed by K. Hosino.

Wells,(46) and Bastin.(47) Little attention, however, has hitherto been given to the comparatively small content of mangesium in the brine. Recently, T. Ishikawa and T. Baba,(38) after studying the compositions of Japanese brine and that of fish blood, especially from the marked similarity in their ratios of sodium, potassium, magnesium and iodine, suggested that the origin of the brine from oil fields is the body fluids of these fishes whose oils have been known to most plausibly explain the origin of petroleum.

From the high concentration of radium in ocean-bottom sediments, R. D. Evans, A. F. Kip and E. G. Moberg,(57) after studying the radium content of marine life, concluded that it must be partly dut to biochemical processes, namely, that various forms of marine life have radium concentrated in their bodies, die and settle to the bottom.

These two theories contribute vitally important suggestions to the fact that brine waters have comparatively high radium content. At the

Table 24. Radium content of ocean water as reported by various observers.

Author and Date	General Locality	Radium content $g./l. \times 10^{12}$	Author and Date	General Locality	Radium content $g./l. \times 10^{12}$
Eve(48) (1907)	N. Atlantic	0.6	Wright, Heise(54) (1918)	South China Sea	0.1–0.2
Joly(49) (1908)	Ireland	11.2-34.6*			
	Mediterranean	11.5-28.2*	Devaputra, Thompson, Utterback(55) (1932)	Pacific	3.3–6.9*
	Arabia	27.8*		Atlantic	38–47*
Eve(50) (1909)	N. Atlantic	0.47–1.50	Evans(56) (1934)	Pacific	0.03
Joly(51) (1909)	Atlantic	8 -17*		Atlantic	0.15
	Mediterranean	2 -14*	Evans, Kip, Moberg(57) (1937)	Pacific	0.02–0.06
	Black Sea	7*	Föyu, Karlik, Petterson, Rona(58) (1939)	Baltic Sea	0.03–0.2
	Indian Ocean	4 - 7*		Sweden	
Satterly(52) (1911)	S.E. England	0.2–1.6	S. Gōda(59) (1939)	Yellow Sea	0.04–0.15
Lloyd(53) (1915)	Gulf of Mexico	1.7		East China Sea South China Sea	

* According to Evans,(57) the high radium concentration of sea water obtained by Joly and Devaputra is owing to experimental errors.

(46) V. R. Mills and R. C. Wells, U. S. Geol. Surv. Bull., (1919), No. 693.
(47) E. S. Bastin, *Bull. Am. Assoc. Petr. Geologists*, **10** (1926), No. 12.
(48) A. S. Eve, *Phil. Mag.*, (6), **13** (1907), 248.
(49) J. Joly, *Phil. Mag.*, (6), **15** (1908), 385.
(50) A. S. Eve, *Phil. Mag.*, (6), **18** (1909), 18.
(51) J. Joly, *Phil. Mag.*, (6), **18** (1909), 396.
(52) J. Satterly, *Proc. Cambridge Phil. Soc.*, **16** (1910–12), 360.
(53) S. J. Lloyd, *Am. J. Sci.*, **39** (1915), 580.
(54) J. R. Wright and G. W. Heise, *Philippine J. Sci.*, **13**A (1918), 49.
(55) D. Devaputra, T G. Thompson, C. L. Utterback, *J. conseil intern. exploration mer*, **7** (1932), 358; *C. A.* **29** (1935), 2076.
(56) R. D. Evans, *Phys. Res.*, **46** (1934), 328.
(57) R. D. Evans, A. F. Kip and E. G. Moberg, *Am. J. Sci.*, **36** (1938), 241.
(58) E. Föyu, B. Karlik, H. Petterson and E. Rona, *Nature*, **143** (1939), 276.
(59) S. Gōda, *Bull. Shanghai Natural Sci. Research*, **9** (1939), 111.

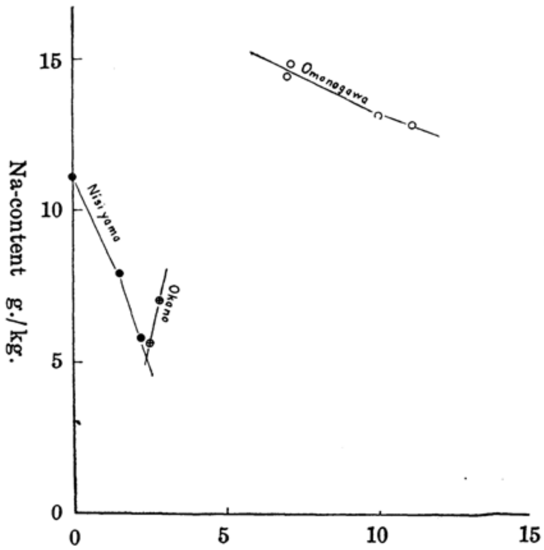

Fig. 23. Relation between the Ra-content and the Na-content of brines.

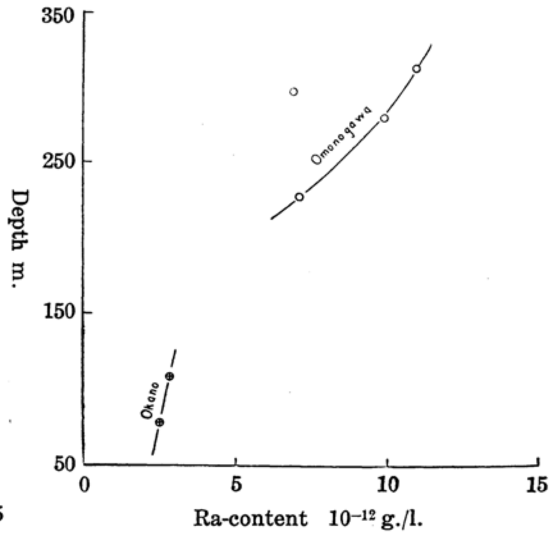

Fig. 24. Relation between the Ra-content of brine waters from the petroleum well and the depth of the well. I.

same time, we may naturally assume that the presence of a certain part of the radium content is due to radium dissolved from contiguous soft rocks.

The radium content of brine waters from oil fields in other countries has been determined by a number of investigators, all of which show high values, especially brines from the oil fields in Caucasus, Russia; that from Tzeliutsing in Szechwan, China, having an extraordinary high content of radium, as described in a previous chapter. Although it is not certain whether the origin of brines from oil fields in other countries is the same as that in Japan, it is an interesting fact in connection with the high radium content of Japanese brines.

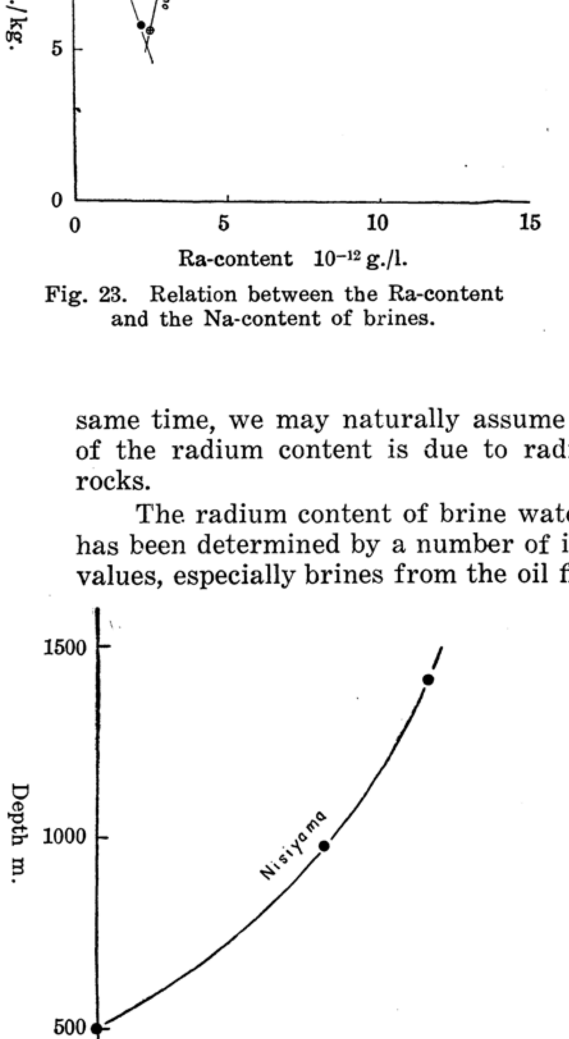

Fig. 25. Relation between the Ra-content of brine waters from the petroleum well and the depth of the well. II.

S. Gōda,(6) after studying the radium content as well as the other chemical constituents of brine waters from Tzeliutsing in Szechwan, China, reported that the radium contents of brines descreased with the increase of their sodium contents. We shall now compare the brines from a number of petroleum wells which are situated close together (V. Table 25). In Fig. 23 their radium content is plotted against their sodium content. As may be seen from this figure, there appears to be a distinct relationship between it and S. Gōda's findings. In Figs. 24 and 25 the correlation between their radium contents and the depths at which the brine samples were drawn is shown. So far as the present results are concerned, there seems to be a linear relation between them also.

Table 25.

No.*	Oil-field	Petroleum well		Radium content g./l. $\times 10^{12}$	Na g./Kg.
		No.	Depth m.		
		I. Omonogawa series (Akita Prefecture)			
1	Omonogawa	1	310	11.05	12.27
2	,,	25	226	7.14	14.83
3	Araya	1	278	10.02	13.14
4	Aburaden	1	297	7.02	14.41
		II. Nisiyama series (Niigata Prefecture)			
10	Takamati	R 36	1399	2.2	5.75
11	Iriwada	R 5	971	1.5	7.94
12	Nagamine	C 93	502	0.0	11.05
		III. Okano series** (Niigata Prefecture)			
13	Okano	R 1	78	2.47	5.63
14	,,	,,	108	2.84	6.96

* Numbers correspond to those used in Table 23.
** These two brine samples were drawn from different depths of the same well.

Hot springs from oil fields. Hot springs often gush out from oil fields. According to the investigations by Y. Chitani,(60) the hot springs of Toyotomi in Hokkaidō; Tomine, Asahikawa, Yumoto in Akita Prefecture; Hanezawa, Mogami in Yamagata Prefecture; and Senami, Tukioka and Matunoyama in Niigata Prefecture, belong to this kind of hot springs. Geologically, they usually gush out from a fairly definite stratum (for example the Lower Neogene series), all of which are alkaline common salt springs, their chemical composition being nearly constant.

Of these the hot springs of Toyotomi in Hokkaidō; and Matunoyama and Senami in Niigata Prefecture, have been determined for their radium content, the results of which are given in Table 26. The chemical composition of these spring waters, analysed by the Tokyo Imperial Hygienic Laboratory,(34) are also given in this table for reference in the same way as in Table 23.

(60) Y. Chitani, *J. Geography*, **40** (1928), 626, 698; **41** (1929), 45, 141.

Table 26. Radium content and chemical composition of hot springs from oil fields.

No.	Spring	Prefecture	Radium content g./l. $\times 10^{12}$	Total solid matter g./Kg.	Composition of water								
					Na	K	Ca	Mg	Cl	SO_4	Br	I	Na g./Kg.
1	Toyotomi-Onsen	Hokkaidō	10.10	11.70	100	0.84	1.18	0.75	90.9	0.01	0.05	0.05	4.35
2	Senami-Onsen	Niigata	0.11	4.19	100	3.47	5.71	0.02	103.3	4.97			1.33
3	Matunoyama-Onsen	,,	14.30, 7.16, 6.49, 6.43 6.17, 2.62, 0.29										

Table 27. Radium content and chemical composition of mineral waters of the brine type.

No.	Spring	Prefecture	Radium content g./l. $\times 10^{12}$	Total solid matter g./Kg.	Composition of water								
					Na	K	Ca	Mg	Cl	SO_4	Br	I	Na g./Kg.
1	Kasio-Kōsen	Nagano	18.5	29.45	100	0.83	3.25	0.66	108.9	trace	0.04	trace	10.55
2	Sionoha-Onsen	Nara	5.52										

As will be seen from this table, the hot springs of Toyotomi and Matunoyama have a fairly high radium content. The radium content of the Senami hot springs is very low; according to Chitani,[60] since a large quantity of sea water is supplied continuously to this spring, the low radium concentration of this spring may be owing to mixing with sea water, which is very poor in radium.

Thus all hot springs from oil fields show comparatively high radium concentration. Although their origins are not yet clearly understood, it is an interesting fact in connection with the high radium content of brine waters from oil fields.

Mineral waters of brine type. According to T. Ishikawa and T. Baba,[38] the mineral waters of Kasio in Nagano Prefecture, belong to the mineral waters of brine type. The composition of this water is shown in Table 27 in the same way as before. Though no petroleum has yet been obtained at Kasio, it is on the famous Itoizawa-Sizuoka fault line, along which may exist an extension of the great petroleum zone of northern Japan.

The origin of the mineral waters at Sionoha in Nara Prefecture, is also believed to be connate water, the same as that of the mineral waters at Kasio, although its composition is not yet clearly known.

The radium content of these two mineral waters is shown in Table 27, from which it is also significant that they all show comparatively high radium content, the same as in the brine waters from oil fields.

XIII. Radium Content of Sinter Deposits from Mineral Springs. Some of the mineral springs, especially the earthy carbonioxated ones, produce abundant sinter deposits at the source and on the creek bed on which they flow, as well as in the conduits and at the baths.

On account of the lowering in pressure and temperature of spring water after gushing out from the ground, a part of the calcium bicarbonate that is held in solution loses its carbonic acid and forms a neutral carbonate which settles as a deposit. At the same time other mineral constituents that are dissolved in the spring waters also deposit as the result of physical and chemical reactions.

Since the sinter deposits from mineral springs consist mainly of calcium carbonate, besides calcium sulphate, ferric oxide, manganese dioxide, etc., if the waters contain radium, it should be absorbed by the calcium carbonate, calcium sulphate, ferric oxide and manganese dioxide into the sinter deposits.[61]

The sinter deposits from mineral springs rich in radium, such as the Masutomi Mineral Springs in Yamanasi Prefecture, and the Ikeda Mineral Springs and the Yugakai Hot Springs in Simane Prefecture, were tested for radium. The sample from the Ikeda Mineral Springs, which was obtained at the source of Spring No. 1, consists mainly of calcium carbonate, colored brown by ferric hydroxide. That from the Yugakai Hot Springs is a mixture of sinter deposits obtained at the sources of Springs Nos. 1–3. It is also colored brown. These two samples were obtained in August, 1939. The samples from the Masutomi Mineral

(61) Refer to next chapter.

Springs are called "radium-fossil", which were deposited from mineral waters by the same mechanism as just mentioned, increasing gradually in size and compactness during a long interval of time. They also consist mainly of calcium carbonate, being colored a light yellow by ferric hydroxide. The samples were drawn in October, 1936.

The results of these determinations are given in Table 28. As might be expected, they all show an extraordinary high radium content, the sinter deposits obtained at the source of the springs being especially rich in radium.

Table 28. Radium content of sinter deposits from mineral springs.

Spring	Location	Radium content of water g./l. $\times 10^{12}$	Radium content of deposit g./g. $\times 10^{12}$
Radium Kōsen No. 1	Ikeda	36.43	872
Yugakai Onsen No. 1	Yugakai	35.06	202
,, No. 2		30.57	
,, No. 3		13.45	
Tuganerō-simo-no-yu	Masutomi	25 05	175
Higasiobi-no-izumi	,,	35.65	19.2
Siokawara	,,		18.4
Ginsentō-huru-yu	,,	9.39	9.6
Kuridaira No. 3 (Kozizimeki)	,,	6.56	8.8

XIV. Extraction of Radium from Mineral Waters. The extraction of radium from mineral waters containing radium was studied for the following purposes: first, to investigate a technique for separating radium from natural waters containing large amounts of it, without the necessity of using a large quantity of reagents, and, secondly, from the geochemical point of view, in order to make clear the process of migration and concentration of radium by natural waters.

For the purpose of this experiment the following method was adopted:

First, as "a carrier of radium", solutions of various metallic salts were added to the water sample. Then by adding suitable precipitants into this solution, these metals were precipitated from them, when the radium in the solution is separated from them, being precipitated with or adsorbed by these precipitates, and concentrated in them.

As "a carrier of radium" for adding to the water sample, such metals as manganese, iron, barium, calcium and strontium were used for the following reasons: first, the radium content of the sediments from the spring waters or that of the ocean-bottom sediments are closely related to the amount of manganese or iron in them; second, since metallic radium belongs to the group of alkali-earths, its salts closely resemble those of barium. Besides these metals, aluminium, chromium, copper, zinc and bismuth were also used in this experiment.

Methods. As water samples for these experiments, the mineral waters from the spring of Arima Hon-Onsen, containing 71.29×10^{-12} g. Ra/l., that of Wadamatuba-Kōsen at Masutomi, containing 82.66×10^{-12} g. Ra/l. and that of Tuganerō-no-yu No. 1 at Masutomi, containing 64.53×10^{-12} g. Ra/l. were used. We also used waters of much higher radium content made by concentrating these spring waters.

Since the sediments in mineral waters contain fairly large quantities of radium, these sediments were dissolved by adding a few c.c. of hydrochloric acid to the water. As the sediments consist mainly of calcium carbonate, they are dissolved almost completely with hydrochloric acid. The insoluble residue is then filtered off and the filtrate thus obtained used as standard solution.

To 500 c.c. of this solution is added a solution of salts of such metals[62] as manganese, iron, barium, calcium, strontium, aluminium, chromium, copper, zinc and bismuth, and these metalic salts are precipitated from the water by the methods shown below. The precipitates thus obtained are filtered off and dissolved by the methods shown below.

Methods of experiment. 1) MnO_2: A solution of manganese chloride is added to the standard solution. By treating it with sodium hydroxide and hydrogen peroxide, manganese is precipitated as manganous acid. The precipitate is filtered off and dissolved in hydrochloric acid.

2) MnS: A solution of manganese chloride is added to the standard solution. After adding ammonium chloride and ammonia, the solution is boiled, after which hydrogen sulphide is passed through it. The precipitate of manganous sulphide is filtered off and dissolved in hydrochloric acid.

3) $Fe(OH)_3$: A solution of ferric oxide is added to the standard solution. By adding amomnium chloride and ammonia, iron is precipitated as its hydroxide. The precipitate is filtered off and dissolved in hydrochloric acid.

4) $Al(OH)_3$, 5) $Cr(OH)_3$, 6) $Bi(OH)_3$: Solutions of aluminium sulphate, chromium sulphate and bismuth nitrate are in turn added to the standard solution. Aluminium, chromium and bismuth are precipitated as their hydroxides. The precipitates are filtered off and dissolved in hydrochloric acid.

7) $BaSO_4$, 8) $SrSO_4$: After adding a few c.c. of sulphuric acid, solutions of barium chloride and strontium chloride are added in turn to the standard solution. The precipitates of barium sulphate and chromium sulphate are filtered off, ignited and fused with a carbonate mixture. (a) The fused mass is leached with distilled water to dissolve the sodium sulphate. (b) The insoluble residues of barium carbonate and strontium carbonate are dissolved in hydrochloric acid.

9) $BaCO_3$: Barium chloride solution is added to the standard solution, and the latter heated to boiling. After rendering it alkaline with ammonia, a solution of ammonium carbonate is added slowly. The precipitate of barium carbonate is filtered off and dissolved in hydrochloric acid.

The mineral waters of Tuganerō-no-yu, however, hold a fairly large concentration of sulphate ions, so that when it is used as standard solution, barium sulphate is precipitated by addition of barium chloride. Ammonium carbonate is added to this solution, when the barium ion remaining in solution is precipitated as barium carbonate. The precipitate is filtered and (a) barium carbonate is dissolved in hydrochloric acid, (b) the insoluble residue of barium sulphate being dissolved by the same treatment as in (7).

10) $CaCO_3$, 11) $SrCO_3$: Solutions of calcium chloride and strontium chloride are added to the standard solution and calcium carbonate and strontium carbonate precipitated by the same treatment as in (9), and then filtered off and dissolved in hydrochloric acid.

12) CuS: A solution of cupric chloride is added to the standard solution.

(62) These reagents were ascertained by blank tests to contain no radium.

After making the solution acidic with hydrochloric acid, hydrogen sulphide is passed through it and cupric sulphide is precipitated, which is filtered off and dissolved in aqua regia.

13) ZnS: A solution of zinc chloride is added to the standard solution. After rendering the solution alkaline with ammonia, hydrogen sulphide is passed through it and zinc sulphide is precipitated, which is filtered off and dissolved in hydrochloric acid.

14) $Fe(OH)_3$, etc. in standard solution: By adding ammonium chloride and ammonia to the standard solution (the mineral waters of Arima Hon-Onsen), the iron, aluminium, etc. present in them are precipitated as mixtures of their hydroxides, which are filtered off and dissolved in hydrochloric acid.

The resulting solution, containing all the radium adsorbed by the precipitate, is put into a curie bottle and sealed. The radium content of this solution is determined by the method described in the previous section. By comparing this result with that of the standard solution, it is possible to calculate the proportion of radium in the standard solution that has been separated from it with the precipitate.

At first, 1 g. of each metallic salt (weight of cation) was added to the standard solution as the radium carrier, while for such metals as manganese, iron, barium, calcium and strontium, which adsorb especially large quantities of radium, the amounts of these metalic salts to be added to the solution were varied from 0.01 g. to 1 g. (weight of cation).

Results. The results of the experiments when 1 g. (weight of cation) of each metalic salt has been added in the standard solution as "radium carrier" are shown in Table 29. When iron, aluminium, chromium, bismuth or manganese are precipitated from the standard solution as their hydroxides, a small quantity of iron, alumium, etc. that are held in the standard solution are also precipitated with them, whence it is presumed that the results include the amount of radium precipitated along with the pricipitates of iron, aluminium, etc. from the standard solution.

For such metals as manganese, iron, calcium, strontium and barium, which show results of 100% or nearly so in Table 29, the quantities of these metallic salts to be added to the standard solution were varied from 0.01 g. to 1 g. (weight of cation). The results are given in Table 30.

Finally it would be interesting to know whether or not the result when the precipitate of iron is added to that of manganese or calcium is the sum of the two results. The results of this experiment are shown in Table 31.

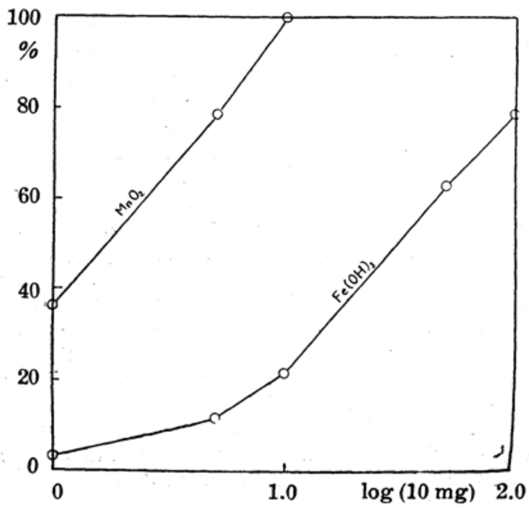

Fig. 26. Relation between the quantity of the radium carrier and the percentage of radium adsorbed from the water.

Discussions. 1) As will be seen from Table 30, the quantity of radium that is extracted from the solutions seems to increase with increase in the quantity of the precipitate caused by the carrier. If, in order to ascertain more exactly the relation between these two quantities, the points are plotted on a diagram with the logarithm of the quantity of the radium carrier to be added as abscissae and the percentage of radium adsorbed from the water as ordinates, we obtain Fig. 26, in which the points lie fairly closs to that of a straight line.

2) As seen in Table 29 and Table 30, the best results were obtained when sulphates and carbonates of barium, calcium and strontium were used as carrier.

It is generally considered that radioactive cations are adsorbed by precipitates containing anions that form insoluble compounds with the former, and that when both these compounds containing the same anion

Table 29.

Carrier	Standard solution	Quantity of radium in standard solution 10^{-12} g.	Quantity of radium in precipitate 10^{-12} g.	Per cent of radium adsorbed from the water %
1) MnO_2	B_2	76.2	76.2	100.0
2) MnS	B_2	76.2	0.0	0.0
3) $Fe(OH)_3$	B_1	44.7	35.2	78.7
4) $Al(OH)_3$	B_2	76.2	7.2	9.5
5) $Cr(OH)_3$	B_2	76.2	52.5	68.9
6) $Bi(OH)_3$	B_2	76.2	3.4	4.5
7) $BaSO_4$				93.7
(a) Na_2SO_4	A	35.0	0.0	{ 0.0
(b) $BaCO_3$	A	35.0	32.8	{ 93.7
8) $SrSO_4$				92.0
(a) Na_2SO_4	A	35.0	0.0	{ 0.0
(b) $SrCO_3$	A	35.0	32.2	{ 92.0
9) $BaCO_3$	B_2	76.2	76.2	100.0
10) $CaCO_3$	B_2	76.2	76.2	100.0
11) $SrCO_3$	B_2	76.2	76.2	100.0
12) CuS	A	35.0	4.9	14.0
13) ZnS	A	35.0	23.8	68.0
14) $Fe(OH)_3$ etc. in standard solution (Arima Hon-Onsen)	A	35.0	1.8	5.1

Standard solution:

A 35.0×10^{-12} g. Ra/500 c.c. Arima Hon-Onsen

B_1 44.7×10^{-12} g. Ra/500 c.c. Wadamatuba-Kōsen (concentrated)

B_2 76.2×10^{-12} g. Ra/500 c.c. ,, (concentrated)

C 42.2×10^{-12} g. Ra/500 c.c. Tuganerō-no-yu No. 1 (concentrated)

Table 30.

Quantity of radium carrier (weight of cation)	Standard solution	Quantity of radium in standard solution 10^{-12} g.	Quantity of radium in precipitate 10^{-12} g.	Per cent of radium adsorbed from the water %
I. MnO_2				
1) 0.01 g.	B_2	76.2	27.6	36.2
2) 0.05 g.	B_2	76.2	59.9	78.6
3) 0.10 g.	B_2	76.2	76.0	99.7≒100
4) 0.50 g.	B_2	76.2	76.2	100.0
5) 1.00 g.	B_2	76.2	76.2	100.0
II. $Fe(OH)_3$				
1) 0.01 g.	B_1	44.7	1.5	3.4
2) 0.05 g.	B_1	44.7	5.1	11.4
3) 0.10 g.	B_1	44.7	9.5	21.3
4) 0.50 g.	B_1	44.7	28.0	62.6
5) 1.00 g.	B_1	44.7	35.2	78.7
III. $CaCO_3$				
1) 0.01 g.	C	42.2	37.6	89.1
2) 0.05 g.	C	42.2	38.6	91.5
3) 0.10 g.	C	42.2	39.4	93.4
4) 0.50 g.	C	42.2	42.2	100.0
5) 1.00 g.	C	42.2	42.2	100.0
IV. $SrCO_3$				
1) 0.01 g.	C	42.2	37.6	89.1
2) 0.05 g.	C	42.2	41.2	97.6
3) 0.10 g.	C	42.2	41.2	97.6
4) 0.50 g.	C	42.2	42.2	100.0
5) 1.00 g.	C	42.2	42.2	100.0
V. $BaCO_3$				
1) 0.01 g.				91.5
(a) CO_3''	C	42.2	38.6	{ 91.5
(b) SO_4''				none
2) 0.05 g.				96.2
(a) CO_3''	C	42.2	4.6	{ 10.9
(b) SO_4''	C	42.2	36.0	85.3
3) 0.10 g.				100.5≒100
(a) CO_3''	C	42.2	1.6	{ 3.8
(b) SO_4''	C	42.2	40.8	96.7
4) 0.50 g.				100.9≒100
(a) CO_3''	C	42.2	3.6	{ 8.5
(b) SO_4''	C	42.2	39.0	92.4
5) 1.00 g.				99.5≒100
(a) CO_3''	C	42.2	18.4	{ 43.6
(b) SO_4''	C	42.2	23.6	55.9

Table 31.

Quantity of radium carrier (weight of cation)	Standard solution	Quantity of radium in standard solution 10^{-12} g.	Quantity of radium in precipitate 10^{-12} g.	Per cent of radium adsorbed from the water	
				Observed %	Calculated %
I. $MnO_2+Fe(OH)_3$					
1. Mn 0.01 g.+Fe 0.1 g.	B_1	44.7	36.1	80.8	36.2+21.3=57.5
2. Mn 0.01 g.+Fe 0.05 g.	C	42.2	28.2	66.8	36.2+11.4=47.6
II. $CaCO_3+Fe(OH)_3$					
Ca 0.01 g.+Fe 0.01 g.	C	42.2	42.2	100.0	89.1+ 3.4=92.5

* Calculated from the results in Table 30.

are isomorphous, their coprecipitating reactions are promoted. The present results support well this conclusion.

It is also well known that the method of coprecipitating radium by barium sulphate has been used from olden times when radium was extracted from uranium minerals, such as pitchblende and carnotite.

3) From the geochemical point of view, it is noteworthy that the quantity of radium precipitated along with (perhaps adsorbed by) the precipitate of manganese and iron is very large.

I. D. Kurbatov,(2) after studying the sediments deposited from spring waters at Pyatigorsk, Russia, reported that the quantity of radium in a deposit depends not upon the sulphate ions contained in the sediment, but on the quantity of ferric and manganese oxides in it. These sediments, which consist mainly of calcium carbonate, are colored a light yellow to brown by ferric hydroxide according to the amount of ferric hydroxide present. The quantity of radium in the deposit depends upon the amount of ferric oxide, as shown by the following data (Table 32) given in terms of gram per 100 g. of sediment.

Table 32.

	Deposit 1.	Deposit 2.	Deposit 3.	Deposit 4.
Ra	3×10^{-10} g.	10×10^{-10} g.	59×10^{-10} g.	111×10^{-10} g.
Fe_2O_3	0.05	0.18	1.07	1.83

The present results given in Tables 29–31, agree well with these facts. The results in Table 31 show that when the precipitate of iron is added to that of calcium carbonate and manganese dioxide, the quantity of radium adsorbed by these precipitates increases remarkably.

As to the relation between radium and manganese or iron in the ocean-bottom sediments, C. S. Piggot(63) reported that the oceanic manganese concretions show an extraordinary high radium content. L. M.

Kurbatov[64] also found two manganese concretions from the Pacific Ocean containing a marked high radium content of 1.47 and 0.48×10^{-10} g. per g. More recently, H. Hamaguti[65] upon determining the radium, manganese and iron contents of deep-sea deposits from the Pacific Ocean, reported that the radium content increases with increasing manganese or iron content, which facts may be explained by the mechanism just mentioned.

4) From the present experiments, it has become possible to separate radium from natural waters without the necessity of using a large quantity of reagents. If, therefore, a mineral spring containing a sufficiently large quantity of radium is found in a country poor in radium ores as in Japan, it may be possible to extract it profitably. Much to our regret, however, so far as the present results are concerned, the natural waters in Japan are also poor in radium, so that for the present, we shall have to leave this problem merely as a suggestion, and wait until a natural water with sufficient radium concentration can be found.

In conclusion, the writer wishes to express his hearty thanks to Professor K. Kimura of the Tokyo Imperial University for kind advices and suggestions received during the course of this work, and also to Dr. F. Kodera, Director of the Tokyo Imperial Industrial Research Institute and Mr. T. Nishimura, Chief of the first department of the Institute, for their kind guidance and for permission to publish this report. He is also indebted to Mr. K. Yamasaki and Mr. K. Kuroda of the Tokyo Imperial University, Mr. K. Hosino and Mr. Y. Nemoto of the Tokyo Imperial Industrial Research Institute, and to the Hot Spring Society of Japan for their wholehearted assistance in collecting the water samples used in this study. A part of the cost of this investigation was defrayed from grants given to Prof. K. Kimura by the Nippon Gakuzyutu Sinkō-kwai (the Japan Society for the Promotion of Scientific Research) and by the Hattori Hōkōkwai Foundation to which also the writer wishes to record his thanks.

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(63) C. S. Piggot, *Am. J. Sci.*, **25** (1933), 229.
(64) L. M. Kurbatov, *Am. J. Sci.*, **33** (1937), 147.
(65) H. Hamaguti, *J. Chem. Soc. Japan*, **59** (1938), 171, 675; **61** (1940), 67.