Relation between the Contents of Radon and Radium B in Several Radioactive Mineral Springs

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(Received June 11, 1956)

Introduction

The relation between the contents of radon and its decay products was first studied by K. Kimura, K. Kuroda and Y. Yokoyama[1] at Masutomi Mineral Springs. Further studies were made by Y. Yokoyama[2] at Ikeda and Misasa Springs. Their results are summerised as follows.

(1) The contents of radium A, radium B and radium C are smaller than expected from their equilibrium states with radon.

(2) The greater the radon content, the greater is the extent to which the equilibrium states between radon and its decay products decreased.

(3) In a sealed vessel the equilibrium between radon and radium A is achieved within 20 min. after the issue, and that between radon and radium B and radium C within 3 hours.

(4) In a mineral spring, the contents (expressed in Curie) of Rn, RaA, RaB and RaC decrease in this sequence.

(5) The relationship between the contents of Rn and RaA, RaB and RaC can be accounted for when it is assumed that the latter three are produced within 2 to 45 min. before the issue from Rn free from its decay products.

S. Umemoto[3] reported, however, that RaB can be present more than the equilibrium amount to the Rn, on a continuous measurement of their contents in "Hisui-no-yu" of Misasa Hot Springs. T. Ishimori and I. Hatae[4] measured the contents of Ra, Rn, RaA, RaB, RaC, RaF, ThX, Tn, ThB and ThC in Ikeda Mineral Springs and discussed the relation between the contents of Rn and RaB, showing that RaB can be present more than the equilibrium amount to the Rn*. K. Kimura, K. Nagashima, N. Ikeda and Ka. Kimura[5] also studied the contents of RaB and Rn in the mineral springs of Naegi district, Gifu Prefecture, and found that the RaB content is merely several fractions of the equilibrium amount to the Rn. S. Umemoto[6] continuously measured the contents of Rn and RaB for a year at Misasa Hot Springs. When the content of RaB was compared with that of Rn 20 min. after the issue, the ratio RaB/Rn became greater with increase in the rate of flow, with a few exceptions. It was also made clear that the ratio RaB/Rn decreases when the rate remarkably increases. In special cases, the ratio RaB/Rn is greater than unity but decreases in autumn when water is removed from the surrounding rice field.

The present author intended to determine the content of RaB and other relating components immediately after the issue in order to disclose the relationship between the contents of Rn and RaB. The measurements were made at Hamamura (Kachimi) Hot Springs, Misasa Hot Springs and Sekigane Hot Springs, Tottori Prefecture.

Location and Methods for Experiments

These three Springs are situated in the middle of Tottori Prefecture and are found in a granite zone. Hamamura Hot Springs (I) are among sand hills along the sea side. Misasa Hot Springs (II) are found along the valley of the river Misasa. Sekigane Hot Springs (III) are found along a mountain torrent which flows into the river Ogamo (Fig. 1). Water temperature, rate of flow and the contents of chloride, sulfate, radon and radium B were measured once a month for a year at one spring in I, five springs in II and one in III. Their names are as follows:

1) K. Kimura, K. Kuroda and Y. Yokoyama, *J. Chem. Soc Japan* (*Pure Chem. Sect.*), **69**, 34 (1948).
2) Y. Yokoyama, ibid., **70**, 399 (1949).
3) S. Umemoto, This Bulletin, **26**, 160 (1953).
4) T. Ishimori and I. Hatae, Read before the symposium on geochemistry of the Chemical Society of Japan, Sapporo, July, 1952,; Read before the seventh annual meeting of the Chemical Society of Japan, Tokyo, April, 1954.

* Yokoyama's report mentioned above also shows that RaB is contained more than the equivalent amount to the Rn in Ikeda Spring No.2; (he considered that this is due to escape of Rn) hence it appears certain that the amount of RaB is more than that expected from the Rn content.

5) K. Kimura, K. Nagashima, N. Ikeda and Ka. Kimura, *Radioisotopes*, **4**, 31 (1955).
6) S. Umemoto, *J. Chem. Soc. Japan* (*Pure Chem. Sect.*), **75**, 352 (1954).

TABLE I

OBSERVATIONAL RESULTS IN I

Time of sampling	Weather	Temperature °C	Rate of flow l./min.	Water temperature °C	Cl^- mg./l.	Rn Mache	Rn 10^{-10} c./l.	RaB	RaB/Rn	SO_4^{2-} mg./l.
1954, 26/V a. m. 11.40	Cloudy Fine later	20.5	18.38	69.7	290.6	20.0	72.8	14.6	0.200	392.1
25/VI a. m. 11.40	Cloudy	20.2	17.21	69.7	289.6	19.5	70.9	9.08	0.128	396.1
6/VII a. m. 11.15	Fine	30.5	19.10	69.7	289.6	20.8	75.7	18.2	0.240	437.6
26/VIII a. m. 11.17	Cloudy	32.8	22.17	69.6	290.6	21.9	79.7	16.6	0.208	465.1
27/IX a. m. 11.40	〃	22.0	21.49	69.7	285.1	21.4	77.9	19.5	0.250	426.1
13/X a. m. 11.07	〃	13.8	17.12	69.7	287.6	20.1	73.2	27.5	0.376	409.1
17/XI a. m. 11.11	Rainy	9.3	17.73	70.0	281.6	19.8	72.1	15.3	0.212	416.4
16/XII a. m. 11.22	Fine	4.4	19.32	70.0	286.6	23.2	84.4	25.8	0.306	460.2
1955, 14/I a. m. 11.14	Cloudy	5.0	20.06	70.0	288.1	19.7	71.7	20.8	0.290	452.6
15/II a. m. 11.09	Fine	5.4	16.24	70.0	297.0	20.7	75.3	28.1	0.373	456.2
14/III a. m. 10.54	Cloudy	6.0	17.45	70.0	294.0	19.3	70.2	12.5	0.178	449.8
27/IV a. m. 11.17	Fine	14.0	21.72	69.9	293.5	21.5	78.3	26.8	0.342	436.5

TABLE II

OBSERVATIONAL RESULTS IN II-1

Time of sampling	Weather	Temperature °C	Rate of flow l./min.	Water temperature °C	Cl^- mg./l.	Rn Mache	Rn 10^{-10} c./l.	RaB	RaB/Rn
1953, 1/VI a. m. 11.00	Cloudy	24.0	2.860	63.2	414.7	33.3	121	14.9	0.123
23/VI a. m. 9.10	Cloudy Rainy later	25.8	3.973	66.4	411.7	22.0	80.1	15.7	0.196
6/VII a. m. 10.18	Cloudy	28.6	4.610	66.6	390.2	26.6	96.8	26.4	0.273
11/VIII a. m. 10.20	〃	30.4	2.538	69.6	605.4	21.0	76.4	23.6	0.309
1/IX a. m. 10.05	Rainy Cloudy later	27.0	2.403	69.8	401.7	20.1	73.1	18.1	0.248
22/IX a. m. 10.10	Fine Cloudy later	22.1	2.750	69.3	400.2	18.5	67.3	18.2	0.271
13/X a. m. 10.07	Cloudy Fine later	19.0	1.710	68.2	396.2	10.7	38.9	14.5	0.373
9/XI a. m. 10.16	Fine	16.6	1.893	67.6	395.2	14.8	53.9	18.0	0.334
14/XII a. m. 10.10	〃	7.2	2.476	68.0	405.7	11.1	40.4	12.9	0.319
1954, 11/I a. m. 10.35	Rainy	7.8	2.645	67.0	400.7	14.4	52.4	13.5	0.258
15/II a. m. 10.40	Cloudy	6.0	2.091	66.2	400.2	13.2	48.0	15.1	0.314
15/III a. m. 10.22	〃	5.0	1.993	66.5	402.7	14.5	52.8	16.6	0.314
20/IV a. m. 10.32	〃	13.0	1.585	65.8	377.2	10.7	38.9	7.5	0.192
20/V a. m. 10.15	〃	21.6	1.673	67.3	399.2	13.8	50.2	12.1	0.240

TABLE III

OBSERVATIONAL RESULTS IN II-2

Time of sampling	Weather	Temperature C°	Rate of flow l./min.	Water temperature °C	Cl⁻ mg./l.	Rn Mache	Rn 10⁻¹⁰ c./l.	RaB 10⁻¹⁰ c./l.	RaB/Rn
1953, 16/X a. m. 10.06	Cloudy	16.6	0.731	58.0	613.9	13.5	49.1	39.0	0.794
13/XI a. m. 10.06	〃	11.0	1.022	59.0	629.8	13.8	50.2	29.4	0.586
17/XII a. m. 10.04	〃	12.0	0.696	57.3	655.6	14.1	51.3	40.8	0.795
1954, 16/I a. m. 10.14	〃	7.2	0.745	58.3	663.3	16.5	60.1	41.0	0.682
25/II a. m. 10.30	Fine Cloudy later	7.0	1.060	58.0	657.8	15.7	57.1	36.0	0.630
18/III a. m. 10.10	Cloudy	8.5	1.063	59.3	650.8	19.0	69.2	42.1	0.608
23/IV a. m. 10.29	Fine	16.7	1.053	59.4	655.3	15.0	54.6	38.9	0.713
19/V a. m. 10.12	Cloudy	21.6	1.030	59.5	660.3	15.7	57.1	36.9	0.646
22/VI a. m. 9.54	〃	19.0	1.500	61.0	661.8	16.7	60.8	13.8	0.227
23/VII a. m. 10.01	Fine	28.5	1.260	60.4	620.2	17.1	62.2	32.8	0.527
23/VIII a. m. 10.06	〃	31.2	1.000	61.0	591.2	19.9	72.4	22.9	0.316
21/IX a. m. 9.50	〃	25.0	1.250	59.0	611.7	17.5	63.7	17.0	0.268

TABLE IV

OBSERVATIONAL RESULTS IN II-3

Time of sampling	Weather	Temperature C°	Rate of flow l./min.	Water temperature °C	Cl⁻ mg./l.	Rn Mache	Rn 10⁻¹⁰ c./l.	RaB 10⁻¹⁰ c./l.	RaB/Rn
1953, 4/VI a. m. 10.10	Cloudy	22.8	5.06	41.6	505.6	268	976	248	0.254
24/VI a. m. 9.50	Fine Cloudy later	27.1	11.28	44.4	354.2	357	1300	884	0.680
8/VII a. m. 10.15	Cloudy Rainy later	28.0	11.59	44.0	355.2	372	1350	1060	0.788
12/VIII a. m. 10.23	Fine	31.3	9.82	41.3	188.1	473	1720	2100	1.22
2/IX a. m. 10.40	Fine Cloudy later	29.0	8.78	40.0	182.1	471	1710	1120	0.655
28/IX a. m. 10.13	Fine	21.2	7.42	39.2	216.6	449	1630	311	0.191
14/X a. m. 10.14	〃	17.2	2.87	39.4	375.1	414	1510	1450	0.960
10/XI a. m. 10.20	Cloudy Rainy later	14.7	2.68	40.0	505.2	483	1760	2650	1.503
15/XII a. m. 10.15	Cloudy	8.0	3.60	41.7	460.7	440	1600	1700	1.064
1954, 13/I a. m. 10.41	Rainy	5.0	4.91	42.0	495.2	311	1132	1260	1.114
24/II a. m. 10.15	Fine	6.3	6.95	43.0	392.2	619	2250	1660	0.739
16/III a. m. 10.15	Cloudy	7.2	5.90	42.4	340.1	610	2220	2850	1.285
21/IV a. m. 10.10	Fine	13.5	3.89	42.2	471.7	496	1805	2096	1.161
18/V a. m. 9.29	〃	20.9	3.88	42.6	492.2	393	1430	1270	0.886
24/VI a. m. 10.00	Cloudy	19.6	8.35	44.0	369.1	471	1714	864	0.504
25/VII a. m. 9.55	Fine	29.0	7.73	42.7	240.1	420	1530	1580	1.035
24/VIII a. m. 10.20	〃	30.0	6.50	42.4	283.6	568	2070	2047	0.989
22/IX a. m. 9.50	〃	25.0	4.06	41.3	272.6	379	1380	903	0.654

TABLE V

OBSERVATIONAL RESULTS IN II-4

Time of sampling	Weather	Temperature C°	Rate of flow l./min.	Water temperature °C	Cl⁻ mg./l.	Rn Mache	Rn 10^{-10} c./l.	RaB 10^{-10} c./l.	RaB/Rn
1954, 24/V a. m. 10.09	Cloudy Rainy later	20.5	2.927	46.7	580.6	244	888	674	0.759
23/VI a. m. 10.00	Rainy	19.7	4.010	46.3	505.7	266	968	1066	1.101
26/VII a. m. 9.49	Cloudy	29.0	3.000	50.0	635.5	237	863	803	0.930
25/VIII a. m. 10.08	Fine	30.8	3.150	50.0	603.8	240	873	947	1.085
24/IX a. m. 10.07	Cloudy	25.4	2.992	49.0	496.7	228	828	801	0.967
14/X a. m. 11.01	Fine Cloudy later	14.0	3.339	47.0	560.2	236	859	729	0.849
16/XI a. m. 10.30	Cloudy Fine later	13.0	3.140	46.0	522.7	227	826	686	0.830
14/XII a. m. 10.08	Rainy	5.2	2.964	43.0	522.7	245	892	901	1.01
1955, 24/I a. m. 10.33	Fine	5.0	3.258	39.2	545.2	231	841	717	0.852
14/II a. m. 10.48	Fine Cloudy later	3.6	2.719	40.9	615.2	213	775	721	0.930
11/III a. m. 10.30	Fine	8.3	0.810	43.2	634.2	218	793	495	0.624
29/IV a. m. 10.06	Cloudy	14.0	1.230	45.3	609.1	243	885	717	0.810

TABLE VI

OBSERVATIONAL RESULTS IN II-5

Time of sampling	Weather	Temperature C°	Rate of flow l./min.	Water temperature °C	Cl⁻ mg./l.	Rn Mache	Rn 10^{-10} c./l.	RaB 10^{-10} c./l.	RaB/Rn
1953, 13/VI a. m. 10.11	Fine	22.3	2.600	57.3	268.7	21.5	78.3	18.1	0.231
25/VI a. m. 9.50	Cloudy	28.0	2.892	57.7	286.2	22.9	83.4	22.2	0.266
9/VII a. m. 10.20	〃	25.5	3.130	58.0	273.5	22.0	80.0	21.9	0.274
13/VIII a. m. 10.15	Fine Cloudy later	31.0	2.644	57.6	280.2	21.4	77.9	21.4	0.275
3/IX a. m. 10.15	Cloudy	26.3	2.756	57.7	279.2	21.9	79.7	38.1	0.478
29/IX a. m. 10.15	Rainy	20.7	2.898	57.8	270.6	21.4	77.9	24.2	0.311
15/X a. m. 10.07	Fine	17.0	2.345	57.2	260.1	19.7	71.7	28.5	0.398
12/XI a. m. 11.25	Cloudy	10.3	3.759	57.6	275.9	20.2	73.5	38.7	0.527
16/XII a. m. 10.18	Fine	6.2	2.505	57.3	255.6	20.7	75.3	30.8	0.409
1954, 14/I a. m. 10.35	Rainy	7.0	2.904	57.6	257.6	21.8	79.4	29.5	0.371
16/II a. m. 10.31	〃	5.0	3.017	57.9	256.1	21.2	77.2	31.9	0.413
17/III a. m. 10.22	Cloudy Fine later	8.4	3.307	58.0	251.6	22.0	80.1	32.1	0.401
22/IV a. m. 10.13	Cloudy	13.5	3.149	58.0	232.6	20.7	75.3	35.1	0.466
17/V a. m. 10.05	Cloudy Fine later	17.0	3.178	58.0	214.1	19.7	71.7	14.7	0.201

TABLE VII

OBSERVATIONAL RESULTS IN III

Time of sampling	Weather	Temperature °C	Rate of flow l./min.	Water temperature °C	Cl^- mg./l.	Rn Mache	Rn 10^{-10} c./l.	RaB 10^{-10} c./l.	RaB/Rn	SO_4^{2-} mg./l.
1954, 27/VIII a. m. 10.10	Fine	31.5	11.62	46.0	101.5	36.2	132	65.1	0.493	84.7
28/IX a. m. 10.05	Rainy	—	11.44	46.2	104.0	37.5	137	70.0	0.511	81.5
12/X a. m. 10.28	Cloudy	12.5	11.48	46.1	102.0	32.1	117	64.4	0.550	80.5
18/XI a. m. 10.29	Fine	12.0	11.44	46.5	108.5	33.2	121	57.2	0.473	83.6
15/XII a. m. 10.05	〃	4.5	10.61	46.4	108.5	31.9	116	64.5	0.556	87.2
1955, 12/I a. m. 10.43	Fine Cloudy later	2.0	11.76	46.0	106.0	31.6	115	65.3	0.568	95.4
17/II a. m. 10.04	Cloudy	—	11.49	46.0	110.5	32.6	119	67.2	0.565	94.8
15/III a. m. 10.02	Fine	7.3	11.91	46.0	109.7	32.1	117	36.0	0.308	95.6
26/IV a. m. 10.20	〃	13.5	11.91	45.7	110.5	26.9	97.9	36.1	0.369	91.9
19/V a. m. 10.25	〃	22.3	11.35	46.0	106.7	36.7	133.5	70.2	0.526	91.9
21/VI a. m. 10.18	〃	27.5	11.45	47.0	106.2	35.0	127.4	72.2	0.567	90.9
21/VII a. m. 10.07	〃	31.0	11.00	47.0	107.5	38.2	139	83.4	0.600	92.0

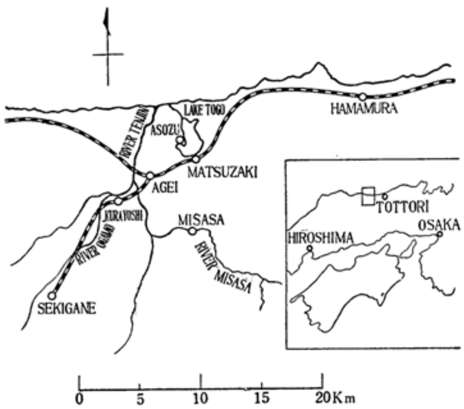

Fig. 1. Locality map

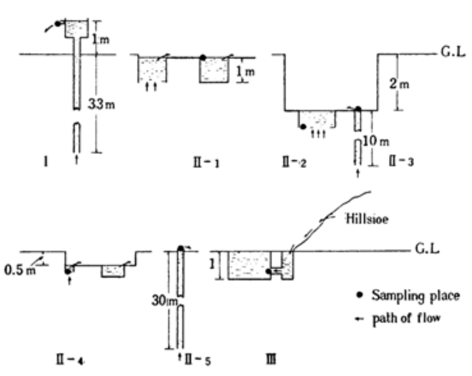

Fig. 2. Schematically states of springs

I	Hamamura (Kachimi)	The Institute for Live Stock.
II-1	Misasa	Bun-aburaya.
II-2	Misasa	Hisui-no-yu (orifice).
II-3	Misasa	Hisui-no-yu (bathtub).
II-4	Misasa	Gunze-shinsenryo.
II-5	Misasa	Tanaka-no-yu.
III	Sekigane	Torikai-ryokan.

The present states of these spings are schematically shown in Fig. 2. Chloride was determined by Mohr's method, sulfate by the barium chromate method and Rn with I. M. fontactoscope. RaB was measured by the dithyzone extraction method[7] immediately after the sampling. The same water was equally treated after three hours and the result was taken as standard*. Measurements were made by Lauritzen electroscope. Results are shown in Table I to VII.

Discussion

Concerning the daily change in the content of various elements, some springs (I, II-2, II-5 and III) show very little fluctuation with time. Others show marked

7) S. Umemoto, *Japan Analyst*, **2**, 201 (1953).

* It is possible that RaB is adsorbed on the glass surface while kept for 3 hours. When the surface is washed with nitric acid (1:9) or acetic acid (1:9) for 3 min. after the mineral water is decanted off after 3 hours' storage, only 1 to 5 percent. of the initial activity of the original RaB is found in the extract. This value can not be regarded to be significant for the following discussion.

change in the content to different extents. (II–3 very marked; II–1, II–4 less so) This fact depends on the following factors:

(1) the depth of the well (i. e. the depth of sampling)

When water is collected at a depth of 10 m. or more, the fluctuation in the content is generally small.

(2) the location of the well.

The fluctuation is more marked when the spring is among other springs comprising water of different salt content. When the salt content of other springs in the neiborhood is similar, only a very slight fluctuation is observed with time. Springs which show marked fluctuation in the contents are about 3 m. deep on the northern beach of the river Misasa, where springs of various salt contents are found. The latter shows a marked difference between the content of the component of one compared with another, but the ratio of the components is rather similar. It is, therefore, predicted that the difference is due to the different extent to which ordinary ground water is mixed with an identical hot water which originates in a same source[8]. This also accounts for the observation that flactuation is greater for a shallower spring.

Nothing conclusive can be stated from the present data about the annual change in the content.

On the examination of the diagrams temp.—Cl, Rn—Cl, RaB—Cl, Rn—RaB and rate of flow—ratio RaB/Rn, only a few correlations are found. The correlation coefficients for rate of flow—ratio RaB/Rn in II–4, II–2 and III are respectively 0.661, −0.789 and −0.627. (Fig. 3–5) (All these coefficients are significant stochastically)

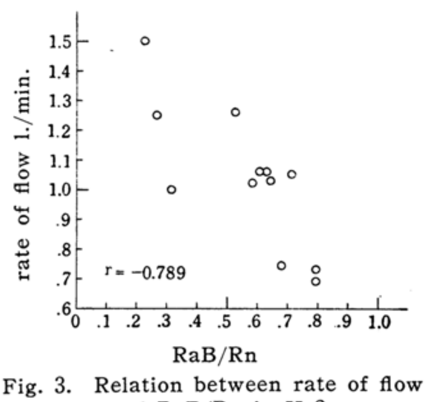

Fig. 3. Relation between rate of flow and RaB/Rn in II–2

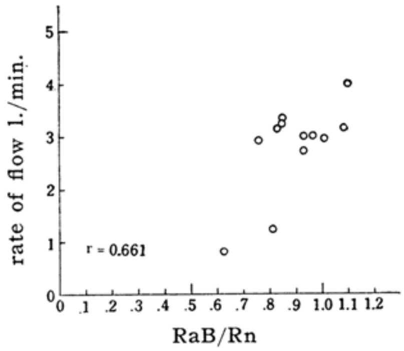

Fig. 4. Relation between rate of flow and RaB/Rn in II–4

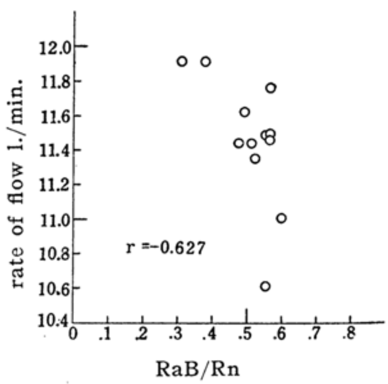

Fig. 5. Relation between rate of flow and RaB/Rn in III

The fact that the latter two have negative values of correlation coefficient provides support to the view of Kimura, Kuroda and Yokoyama about the relationship of the content of Rn and that of its decay products. They claim that radium B in the mineral spring is produced only from Rn which was supplied free from its decay products, the chemical behavior of both species being similar until they reach the surface. Since the contents of chemical constituents in II–2 and III remain almost unchanged with time, it could be reasonably assumed that a radioactive substance is supplied and springs out in a definite manner.

The correlation coefficient between the rate of flow and the ratio of RaB to Rn is positive for II–4; an inversed relationship to the above mentioned case can be predicted. Their assumption appears not to be valid in this case. Since the ratio RaB to Rn is sometimes greater than unity, there must be cases when the RaB is supplied not by the Rn only or when a part of the Rn escapes. The depth at which the mineral water of both II–4 and II–3 (sometimes both having the ratio

8) S. Umemoto, *J. Chem Soc. Japan* (*Pure Chem. Sect.*), **74**, 94 (1954).

greater than unity) is taken, is less than 3 m. and both show very strong overall activity. Strongly radioactive springs are also found in their neighborhood as well as radioactive ground water[9]. They all come from shallow wells less than 3 m. in depth. It is thus possible that Rn and RaB are supplied to the mineral water near the surface or Rn escapes preferentially near the surface, resulting in unusual contents of both species as mentioned above. Another example of a mineral spring that has a greater value than unity for the ratio RaB to Rn, is provided in Ikeda Mineral Springs in Shimane Prefecture. The correlation between the rate of flow and the ratio RaB/Rn in II–4 appears to arise from a still uncertain regularity including the state of supply of both species.

No significant correlation between the rate of flow and the ratio RaB/Rn was observed in other springs. Considering the variation of the main constituents, the ratio in II–3 seems to be liable to fluctuation of influence near the surface, and also those in II–1; nothing regular can be expected for either of them. For I and II–5, the fluctuation in main constituents is less, but it is considered that the contents of minor constituents are not sufficient to fulfill the necessary assumption for Kimura et al's consideration. From the fact that more than the half of the examples are not in accord with their comments, not many mineral springs appear to satisfy the conditions suggested by them. It does not imply, however, that their examples have been rather exceptional. Their second statement appears not to be true for the present example; i. e. the extent to which the RaB content is decreased from the equilibrium amount to the Rn does not increase with increasing Rn content.

9) T. Sugihara, *Repts. Balneol. Lab. Okayama Univ.*, 9, 37 (1953).

Conclusion

The above observed mineral springs are classified into three groups regarding the correlation between the rate of flow and the ratio RaB/Rn: (1) negatively correlating, (2) positively correlating and (3) without significant correlation. In the first group Rn appears to be free from its decay products at the point of its supply and it is the source of all other radioactive species in the mineral water, all of them showing a chemical behavior similar to one another. For the second group no appropriate explanation has been available as yet. For the third group, it could be assumed that the source, path of supply and the chemical behavior after the supply of Rn are too complicated or that these are disturbed by secondary phenomena near the surface. It appears as if the third group comprises most of mineral springs.

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