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	キーワード (Ja):
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	作成者: ABIKO, Tsutomu
	メールアドレス:
	所属:
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Tsutomu Abiko

Abstract

The observed data of recent hydrothermal activity at Noboribetsu are reviewed. A hypothesis on the production of the sulfate ion and molten free sulfur is presented. Some relations between the hydrothermal activity and earthquakes which occured around Hokkaido are discussed.

Introduction

Noboribetsu has two explosion craters, Oyunuma and Jigokudani, the present activity of which is considered to originate in neutral hydrothermal solution going up to 200°C and 30 atms, and releasing annual energy at level of 10^{22} erg[1].

The chemical conponents contained in this original hydrothermal solution are distributed between steam and hot spring water phases according to their solubility when a sudden boilng phenomenon of the original hydrothermal solution takes place on the bottom of two explosion craters. (Table 1). Gaseous components

Table 1. Daily Amount of issuing Chemical Component (ton/day)

(1952)	Water	Total Salt	Cl	SO_4	HCO_3	Na	Ca	Fe	Al	Silicic Acid
Jigokudani	3×10^3	3.0	0.6	0.9	0.0	0.3	0.1	0.02	0.3	0.8
Oyunuma	1.7×10^3	0.3	0.06	0.07	0.0	0.03	0.006	0.03	0.3	0.02
Town	3×10^3	25	12	0.07	0.5	5.0	2.0	0.001	0.0001	1.0
Total	8×10^3	28	12.7	1.1	0.5	5.3	2.1	0.05	0.6	1.8

(1967)	Water	Cl	Na	Ca	Mg					
Jigokudani	3.1×10^3	0.09	0.08	0.07	0.01					
Oyunma	3.2×10^3	0.67	0 43	0.2	0.03					
Town	1.0×10^3	2.0	0.91	0.3	0.02					
Total	7.3×10^3	2.76	1.42	0.57	0.06					

are extracted in steam and common ions remain in hot spring water.

The concentration of chemical components in the original hydrothermal solution can be calculated by Equation 1.

$$\text{Original concentration} \quad C_0 = \sum V_i C_i / \sum V_i \qquad (1)$$

V_i is the amount of issuing water and C_i is the concentration of chemical component in question. Some one hundred analyses were required to get every

C_0 value. C_0 value thus calculated might be considered to reflect the characteristic property of the original hydrothermal solution. Continuous observations for a long period are considered to shown a certain relation between C_0 values and the intensity of activity if the assuption is true. The concentration of common ions such as Na and Cl thus calculated we found to be in a good accordance with observed concentration in hot spring water, secondary hydrothermal water, as shown in Table 2.

Table 2. Chemical Concentration and Composition of the Original Hydrothermal Solution, in mil equivalent.

Original Solution, C_0.

Date	Cl	Na	Ca	Na/Cl	Ca/Cl
1952	91.7	58.2	26.9	0.65	0.29
1969	70.5	—	17.5	—	0.25

Secondary Solution (No. 5), C_i.

| 1952 | 108.0 | 72.2 | 26.9 | 0.67 | 0.25 |
| 1969 | 60.4 | 49.2 | 14.0 | 0.82 | 0.23 |

Table 3. The Variation of Noboribetsu Hydrothermal Activity and the Chemical Property of a Secondary Hydrothemal Solution (No. 5) in mil equivalent

Date		HCO$_3$	Cl	SO$_4$	Na	Ca	Na/Cl	Ca/Cl
July,	1949	2.31	108	0.8	72.2	26.9	0.69	0.25
July,	1951	2.2	109	0.8	74.3	26.6	0.68	0.24
Nov.,	1951		9.07		68.1	23.8	0.75	0.26
Feb.,	1952	3.0	93.5	0.8	64.3	22.5	0.68	0.24
Mar.,	1952	3.0	85.8		59.8	20.8	0.69	0.24
June,	1952	3.8	70.7		50.5	16.0	0.72	0.23
July,	1952		62.7		47.2		0.75	
Oct.,	1952	3.5	53.7		42.2	12.0	0.79	0.22
Mar.,	1953	3.9	36.0		30.7	6.5	0.85	0.18
May,	1953	3.9	31.5	1.3	27.4	6.7	0.87	0.21
Aug.,	1953	3.4	29.6	1.5	26.1	5.1	0.88	0.17
Sept.,	1953		31.5		26.3	5.1	0.83	0.16
Oct.,	1953		33.1		27.7	6.0	0.84	0.18
Dec.,	1953		33.5		27.9	6.5	0.83	0.19
Mar.,	1954		37.7		30.5	8.1	0.81	0.22
May,	1954		43.4		34.7	9.9	0.80	0.23
July	1954		47.6		37.0	10.6	0.78	0.22

Table 3 (continued)

Date		HCO$_3$	Cl	SO$_4$	Na	Ca	Na/Cl	Ca/Cl
Sept.,	1954		50.2		38.2	11.3	0.76	0.23
Aug.,	1962		83.9		57.6	21.3	0.67	0.25
Jan.,	1963		82.9		57.6	20.1	0.70	0.24
Mar.,	1963		79.8		54.6	19.3	0.68	2.24
May,	1963		78.2		55.0	18.9	0.70	0.24
June,	1963		74.9		50.7	18.0	0.68	0.24
Dec.,	1963		25.7		55.0	18.0	0.73	0.24
Feb.,	1964		72.7		50.5	17.5	0.69	0.24
April,	1964		72.7		50.7	17.5	0.70	0.24
June,	1964		73.6		54.1	17.0	0.74	0.24
Nov.,	1964		77.5		56.4	19.0	0.73	0.25
Mar.,	1965		84.5		60.8	21.4	0.72	0.25
June,	1965		85.8		61.4	21.9	0.71	0.26
Aug.,	1965		87.5		62.9	22.6	0.72	0.26
Oct.,	1965		88.1		62.8	22.1	0.71	0.25
Dec.,	1965		83.4		60.0	20.7	0.72	0.25
Mar.,	1966		81.8		58.7	20.3	0.72	0.25
Aug.,	1966		79.2		56.8	19.2	0.72	0.24
Jan.,	1967		76.6		54.8	18.3	0.72	0.24
Feb.,	1967		74.0	0.7	54.0	17.5	0.73	0.24
Mar.,	1967		72.7	0.7	52.7	17.1	0.73	0.23
May,	1967		75.3	0.7	55.2	18.5	0.73	0.24
June,	1967		73.3		54.1	17.7	0.74	0.24
July,	1967		71.9	0.6	53.2	17.3	0.74	0.24
Aug.,	1967		73.4	0.5	54.0	17.7	0.74	0.24
Sept.,	1967		72.2		54.2	17.3	0.75	0.24
Oct.,	1967		71.4	0.8	51.8	17.1	0.73	0.24
Nov.,	1967		71.4	0.8	52.4	17.1	0.73	0.24
Jan.,	1968		71.9	0.8	52.6	17.2	0.73	0.24
April,	1968		67.3	0.8	50.5	16.0	0.75	0.24
May,	1968		66.1	0.8	49.4	15.5	0.75	0.23
June,	1968		59.9	1.0	48.9	13.6	0.82	0.23
July,	1968		57.2	0.8	45.2	13.3	0.79	0.23
Aug.,	1968		57.6	0.9	42.0	13.7	0.73	0.24
Sept.,	1968		58.2	0.8	43.5	14.0	0.75	0.24
Oct.,	1968		59.7	0.8	45.7	14.3	0.76	0.24

So, daily values of C_i as partially lised in Table 3 have been used for C_0 with much ease to reflect the intensity of hydrothermal activity (Fig. 1). It has been found that C_i values of common ions such as Na, K, Mg, Ca and Cl, and
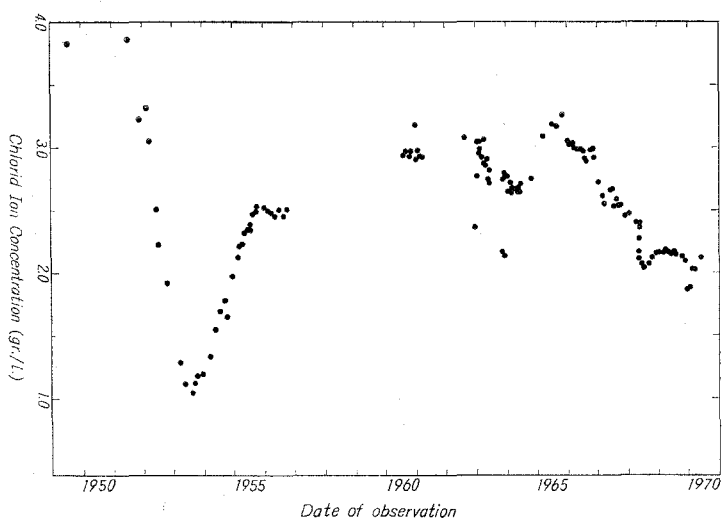
Fig. 1. The Variation of the Chloride Ion Concentration in a Secondary Hydrothermal Solution (No. 5)

also of protium become concentrated when the hydrothermal activity increases its intensity and vice versa. The concentration factor of each cationic component, however, has a different value for chloride ion. For example, Na/Cl becomes smaller when the intensity increases, but gets larger when the activity goes down to weaker side (Fig. 2). On the contrary, Ca/Cl becomes larger during the violent
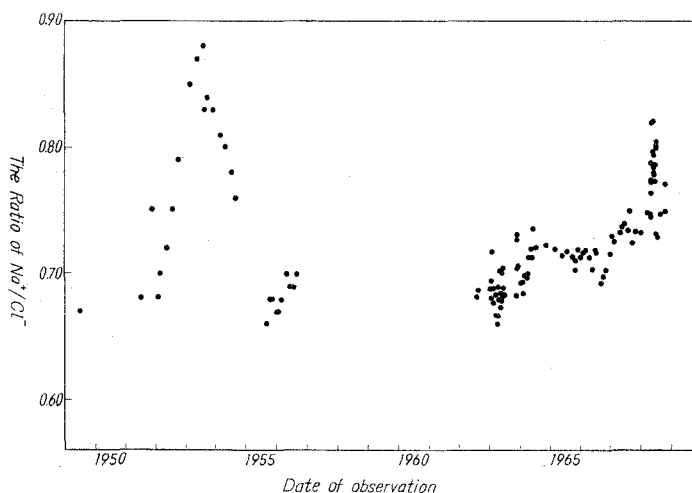
Fig. 2. The Variation of the Ratio of Na/Cl in a Secondary Hydrothermal Solution (No. 5)

activity period but goes down when the activity declines.   Ionic exchange phenomenon between Na and Ca seems to proceed in pararell with the variation of activity, and finally at the weakest intensity the chemical composition of the original hydrothermal soluttion shows a similarity to that of sea water, deutrium being concentrated (Table 4).

Table 4. Isotopic Fractionation of Hydrogen in
a Secondary Hydrothermal Solution

Date		Mean Deviation from Tokyo Tap Water*	Chloride Ion Concentration	Volcanic Activity
Feb.,	1952	− 1.33%	4.50 gr/ℓ	Increasing
Oct.,	1952	− 1.22	3.89	
Mar.,	1953	+ 1.73	3.45	
May,	1953	− 0.52	3.37	Decreasing
Aug.,	1953	− 0.25	3.39	
Oct.,	1953	+ 0.70	3.24	
Dec.,	1953	+ 1.29	3.12	
Mar.,	1954	+ 1.40	2.83	
May,	1954	+ 1.29	2.84	
July,	1954	+ 0.91	2.06	
Sep.,	1954	+ 0.54	3.12	

*   Analyzed by M. Kobayakawa, Tokyo Metropolitan University

According to daily observation of these, C_i values, Murozumi could predict the sudden variation of the activity 6–10 months before the apparent change took place.

In this paper, present author trys to discuss the oxygenation of H_2S gases to molten free sulfur and sulfate ion through the chemical composition of the hot spring water at Oyunuma.   According to daily observation of C_i values during last two decades, it is discussed that there should be some relations between the intensity change of the hydrothermal activity of Noboribetsu and last two Off-Tokachi earthquakes which occured in 1952 and 1968.

Results and discussion

(1) Production of molten free sulfur and sulfate ion

Noboribetsu hydrothermal activity, originating in the end energy of Kuttara volcanism, has spouted the high amount of acidic and neutral hot spring water (Table 1).

Major gaseous components contained in hot spring water and fumarolic gases are steam (H_2O), CO_2 and H_2S. SO_2 is minor.   Therefore, it is considered that sulfate ion and molten sulfur are not existence in the original hydrothermal solution, and these sulfate ions and molten free sulfur are produced by the oxyge-
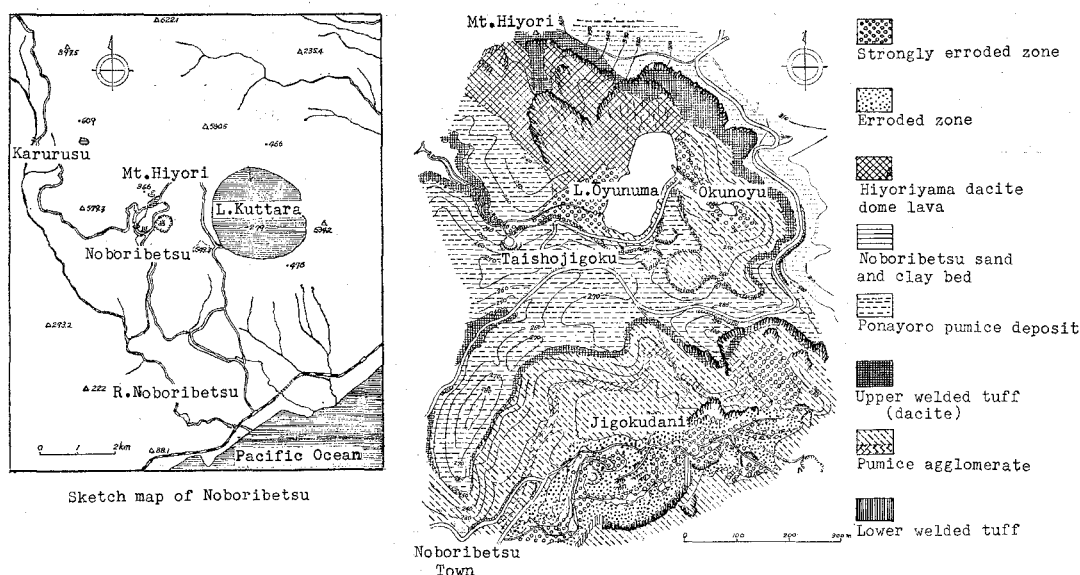
Sketch map of Noboribetsu

Noboribetsu
Town

Fig. 3. Geological map of Noboribetsu J. Suzuki et al. (1958)

nation of H_2S gases near the surface. The oxygenation is considered to be undergoing with the following reaction,

$$2H_2S + O_2 = 2S\downarrow + 2H_2O \qquad (1)$$

$$2S\downarrow + 3O_2 + H_2O = 2H_2SO_4 \qquad (2)$$

$$4FeS_2 + 15O_2 + 2H_2O = 2Fe_2(SO_4)_3 + 2H_2SO_4 \qquad (3)$$

The oxidant used in these reaction is oxygen, the source of which is considered to be both of atmospheric and dissolved oxygen. Most acidic hot spring water (6200m^3/day) is supplied from underground water in which the daily amount of dissolved oxygen is 2×10^3 mole with high estimation. While the daily amount of sulfate ion thus produced is 2.16 ton. The amount of oxygen 9.2×10^4 mole is daily required to oxygenate H_2S to sulfate ion of 2.16 ton. But the amount of dissolved oxygen is not enough to required oxygen. Therefore atmospheric oxygen is used to these oxygenation reactions in the fumaroles and hot springs and the solution thus oxygenated is considered to permeate repeatedly through the underground. And these reactions are undergoing near the surface of the earth.

Table 5. Daily Amount of issuing Sulfur Component (ton/day)

	Water	SO$_4$	S	H$_2$S[d]
Oyunuma	3200[a]	0.67	30[c]	0.006
Jigokudani	3000[a]	1.44	0	0.2
Town	1100[b]	0.04	0	10^{-4}
Total	7300	2.16	—	0.2

a) determined in July 1969
b) determined in Feb. 1969
c) monthly average of taken sulfur from 1940 to 1956
d) from reference paper, 1)

Table 6. Chemical Properties of Oyunuma hot lake water (determined in May 1964)

Depth m	Temp. °C	pH	Cl^- mg/ℓ	SO_4^{2-} mg/ℓ	Na^+ mg/ℓ	K^+ mg/ℓ	$\sum Fe$ mg/ℓ
0	48.5	3.02	123	268	85	12	5.4
1	48.2						
2	48.2	3.02	121	330	85	12	3.9
3	48.2						
4	48.1	3.02	119	300	85	12	3.4
5	48.0						
6	48.1	3.07	118	317	85	12	5.0
7	43.2						
8	48.2	3.03	119	291	85	12	3.7
9	48.1						
10	48.3	3.02	121	290	85	12	5.1
11	48.3						
12	48.2	2.98	121	328	85	12	6.7
13	77.0						
14	99.5	3.18	316	289	180	16	8.7
15	107.0						
16	112.5	3.27	359	283	190	16	22.1
17	113.7						
18	114.7	3.31	370	280	187	15	34.9
19	114.5						
20	114.5	3.24	355	282	180	15	26.4
21	120.5						
22	120.7						
23	121.2						
24	122.0	3.30	361	319	183	16	37.4
25	122.3						

Hot-lake Oyunuma, the maximum depth of 26 m, is formed of two layers. Table 6 shows its chemical components. Upper layer is homogeneous and lower layer is heterogeneous in chemical and thermometric. Later layer is sluggish with molten sulfur, pyrite and weathering rock particles.

Free sulfur is considered to be produced by the reaction (1) in the upper layer because of no dissolved oxygen to oxygenate H_2S gases in the lower. Free sulfur thus produced precipitates and melts by high temperature fumarolic gases on the bottom of Oyunuma.

Pyrite particles mix in molten sulfur. This pyrite is considered to be produced near the surface, because iron is dissolved in the underground water and H_2S is dissolved in original hydrothermal solution.

(2) Some relations between the intensity change of the Hydrothermal activity of Noboribetsu and earthquakes

It is known that Noboribetsu has repeatedly shown remarkable changes of activity since 1880. No such changes of activity on a large scale have been observed in any other hot spring region in Japan. The phenomena are rather similar to volcanic activity.

It was discussed that daily values of C_i have been used for C_0 with much ease to reflect the intensity of hydrothermal activity (Fig. 1).

The variation of C_i has been observed three times during last two decades. Before and after the periods of these variation, large scale earthquakes had happened around Hokkaido (Table 7). The accumulated energy of the earth shell

Table 7. Large scale Earthquakes around Hokkaido since 1940

Date	Name	Magn.	Location	
			long.	lat.
1952. 3. 4	Off-Tokachi	8.2	144.1	41.8
1958. 11. 7	South Off-Iturup	8.0	148.5	44.3
1963. 10. 13	Off-Iturup	8.3	150.0	44.0
1968. 5. 16	Off-Tokachi	7.9	143.35	40.44

is considered to be effect to volcanic and earthquake energy, therefore these facts should show some relations between the intensity change of the hydrothermal activity of Noboribatsu and earthquakes which occured around Hokkaido.

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