Geochemical characteristics of the Sumikawa geothermal system, northeast Japan

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Chemical and isotopic data are summarized for fluids from newly drilled wells in the Sumikawa geothermal system, northern Honshu, Japan, and the geochemical characteristics of the reservoir are discussed. The fluids are mainly of the Na-Cl type with a near neutral pH, and measured temperatures in wells often exceed 300°C. The total dissolved salt concentration is low (<3000 mg/kg), less than other high temperature geothermal systems in Japan. The chemical and isotopic compositions of the fluids vary from well to well. One well (S-2) produces both acid sulfate-chloride and weakly acid pH fluids from two different feed zones, with these fluids possibly related to a local magmatic component.

The Sumikawa geothermal waters are diluted by a relatively shallow heated (~200°C) water that contains sulfate plus bicarbonate but nil chloride. The hydrogen and oxygen isotopic compositions of the total fluid indicate that the fluids are of meteoric origin, with an oxygen isotope shift of less than 2‰. The SO₄ in the meteoric-dominated neutral pH waters comes from sulfur in rocks, which is essentially a mixture of supergene and fossil marine SO₄. On the basis of the N₂, Ar and He values in the vapor fraction, the chemical and isotopic variation of the fluids appears to be due to different degrees of minor magmatic contribution of volatiles to the hydrothermal fluid. Despite the meteoric-dominant source of most water, the magmatic contribution of volatiles results in fluids that are relatively immature and out of chemical equilibrium with the host rocks and alteration minerals.

INTRODUCTION

The Sumikawa geothermal system is located on the slope of Mt. Akita-Yake in the Hachimantai volcanic region (Fig. 1). A 50 MWe geothermal power plant is under construction, while a 10 MWe power station was commissioned by Mitsubishi Materials Corporation (MMC) in 1974 at Ohnuma, 1.5 km east of Sumikawa (Fig. 2). At present, seven wells (S-4, SA-1, SA-2, SA-4, SB-1, SD-1 and SC-1) will be used for production and four wells (S-2, SB-2, SD-2 and SB-3) for reinjection of separated water (Fig. 3).

The Ohnuma-Sumikawa geothermal system has been the subject of geologic and mineralogic studies (Yora *et al.*, 1973, 1977; Kubota, 1979, Sakai *et al.*, 1986). Kubota (1985, 1988) suggested that the subsurface temperature is highest to the south due to up flow associated with the Akita-Yake volcanic activities. The convection system in this area appears to have an associated down-flow zone to the north (Fig. 2). At the Sumikawa system, a down flow is postulated by Kubota (1988).

Chemical and isotopic studies have been conducted on thermal waters and fumarolic gases in

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Fig. 1. Tectonic map of Hachimantai volcanic field (after Sakai *et al.*, 1986).

the Hachimantai area (Matsubaya *et al.*, 1975, 1983; Shigeno and Abe, 1987) and on fluids from well S-4, which was drilled prior to 1983 with other S-series wells (S-1, S-2 and S-3). Noda *et al.* (1987) suggested a mixing model, with several sources for fluids, based on the chemical character of the thermal springs and geothermal wells. They demonstrated that acid Cl-SO₄ type fluid is associated with volcanic gas that originates from beneath Mt. Akita-Yake, to the southwest, while neutral Na-Cl type fluid of low

salinity and a high B/Cl ratio discharges to the north. In the Ohnuma area, the composition of production fluids has been monitored after the start of operation of the power station. Increases in the chloride concentration and $\delta^{18}\text{O}$ are due to return of separated water injected into the reservoir (Kubota *et al.*, 1989). Based on these variations, the mass of the mixing box was estimated to be 3×10^9 kg (Kubota and Matsubaya, 1988; Matsubaya and Kubota, 1988).

The purpose of this paper is to summarize

Fig. 2. General features of the Ohnuma-Sumikawa geothermal area (after Kubota, 1988).

Fig. 3. Locality map of geothermal wells at Sumikawa. Production wells are labeled in boxes while reinjection wells have underlined labels. Closed circles: vertically drilled; open circles: observation wells; well tracks are shown as dashed lines. The contour interval is 50 m.

the geochemical data of fluids discharged mainly from the newly drilled wells at Sumikawa (SA-1, SA-2, SA-4, SB-1, SD-1 and SC-1) in the Ohnuma-Sumikawa geothermal system, and to

discuss the chemical characteristics of the reservoir. In this paper, the chemical composition of fluids collected in 1989 are not discussed in detail since a large quantity of river water was injected

into all but two production wells (SD-1 and SC-1) during this period. The composition of the reservoir fluid was affected by this river water injection, discussed elsewhere (Ueda *et al.*, 1990).

OUTLINE OF GEOLOGY

To the north of the Sumikawa system, a pronounced low gravity zone is associated with an elongated north-south graben structure (Inoue and Ueda, 1965). To the south of the Sumikawa

area (Fig. 1), an east-west trending volcanic chain including Mt. Akita-Yake (1366 mASL) to the west and Mt. Hachimantai (1614 m) to the east obscures the graben structure. Both N-S and NW-SE trending faults are assumed to occur in the Sumikawa system (MMC, unpublished).

The geology of the Sumikawa area is similar to that of the Ohnuma area (Yora *et al.*, 1973; Sakai *et al.*, 1986). It consists of Quaternary volcanic rocks, lacustrine sediments, and Tertiary formation (Fig. 4) intruded by granitic

Fig. 4. Geological section of the Sumikawa geothermal system. a) Geology, and temperature and pressure profiles (after Kubota, 1986). b) Occurrence of hydrothermal minerals (after Kubota, 1986).

rocks. The pre-Tertiary basement has not yet been reached by drilling (the deepest well at Sumikawa is 2486 m). The Tertiary formation is composed of volcanic rocks and marine sediments. The deepest formation identified so far consists of granite, granodiorite and quartz-diorite intrusives into the Tertiary formation (not shown in Fig. 4). The difference from the Ohnuma area is that in Sumikawa the thick Pleistocene lacustrine sediments are overlain by Quaternary volcanic rocks. The thickness of the Quaternary lacustrine sediments increases to the southwest (Figs. 2 and 4a). Intrusive rocks are inferred to occur at greater depths beneath the area drilled at Sumikawa.

The vertical distribution of alteration minerals in the Sumikawa wells was studied by Kubota (1979, 1986), Bamba *et al.* (1987), Sato (1990) and MMC (unpublished data), as shown in Fig. 4b. Montmorillonite occurs above 500 mASL but is not observed in the Quaternary andesitic lavas. Chlorite is commonly found in the formation below the Quaternary rocks. Epidote occurs widely below 300 to 500 mASL at Sumikawa but is not seen even at 0 mASL in the lower temperature zone to the north. Amphibole exists below 200 mASL in the high temperature zone (over 250°C). Laumontite is observed to occur at random above 100 to 200 mASL and is found locally in the Quaternary rocks. In wells with many fractures at shallow depth, laumontite occurs at a shallower depth in comparison with wells with no fractures. Wairakite exists below 600 mASL but is not observed in the lower temperature zone; in well S-4, it is found in the high temperature zone (over 300°C). In the southern part of Sumikawa, wairakite is commonly observed below the calcite zone.

A thin zone of acidic alteration coincides with the feed points of acidic fluid in well S-2 (Fig. 4b), including kaolinite, pyrophyllite, anhydrite, gypsum, barite and alunite. In general, there is a good correlation between the occurrence of temperature-sensitive minerals (Browne, 1978) and the measured temperature (compare Figs. 4a and 4b), with high temperature minerals not occurring in the colder wells to

the north.

THERMAL STRUCTURE

The isotherms in this area generally have the same east-west trend as the volcanic chain from Mt. Akita-Yake to Mt. Hachimantai (Fig. 2). The subsurface temperature gradually increases southward towards the volcanic chain, while it decreases sharply to the north (Fig. 2). In the high temperature region, the temperature gradient is controlled by saturation pressure conditions of a hot water column (Fig. 5a). The measured profiles deviate from the pure water curve at greater depths. This is due to the effect of CO₂ in the reservoir fluid, though in some cases temperature inversions are due to incursion of cooler fluids. In Fig. 5a, the temperature profile for a boiling fluid with approximately 1 wt% CO₂ at 300°C is also shown. If the CO₂-bearing fluids rise and boil, CO₂ is lost and the profile will more closely match that of pure water. This model is consistent with the low gas contents in the fluids at the production depths (see below).

During drilling, temperature was measured in well S-4 in a dry condition (no circulation loss); temperature increased linearly with depth (i.e., conductive) over shallow depths (above 400 mASL, not shown in Fig. 5a; MMC, unpublished data). Based on these thermal profiles, Kubota (1986) proposed the existence of convection in the high temperature zone, while conduction dominates in the lower temperature zone. The boiling temperature profiles at shallow levels in the wells (Fig. 5a) is due to heat transfer upwards in the wells under bleed conditions, and is not the natural state.

The heat source for the Ohnuma-Sumikawa geothermal system is estimated to be located under the volcanic chain to the south. The highest temperature in the area is 317°C at an elevation of -840 mASL at the bottom of well SA-2 (Fig. 5a). A thermal inversion at -1220 mASL in the deepest well, SC-1, was measured 25 weeks after an injection test, indicating deep, relatively cold water inflow.

Kubota (1986) compiled data on filling tem-

Fig. 5a. See caption with Fig. 5b for explanation.

peratures of fluid inclusions in hydrothermal quartz and wairakite, and compared these data with temperature profiles of newly drilled wells. The present subsurface temperatures appear to have been constant at Sumikawa since inclusion formation, while they have decreased by several 10°C at Ohnuma after formation of the alteration minerals, consistent with lower temperatures (Fig. 2).

Reservoir pressure at the feedpoint of wells in three areas, Sumikawa, Ohnuma and Taninainuma in the North Hachimantai region (Fig. 2), is plotted against elevation in Fig. 4b (Kubota, 1988). Wells in the Sumikawa and

Ohnuma areas have feedpoint pressures below 400 mASL which plot near the saturated pressure profile of pure water to about -200 mASL (Fig. 5b), but above this curve (at higher pressure) at greater depth. This discrepancy may be due to a dissolved gas content of up to 1 wt% CO₂ at higher temperatures, or in some cases to the temperature of the fluid being less than that of a pure water boiling column, as proposed before. The pressure above 400 mASL is approximately constant at 3.1 MPa, indicating vapor-dominant conditions. All data for Ohnuma and Sumikawa fall on a single line (Fig. 5b), even well S-2 (issuing acidic fluid), suggesting a con-

Fig. 5. Temperature and pressure profiles of the Sumikawa wells. a) Temperature profile (after MMC, unpublished data). These data are obtained at 4000 to 15000 hours after drilling. Temperature profile starts from water level for each well except for wells SA-1 and SA-2 with no data of water level. Boiling curves of pure water and containing 1 wt% CO_2 (at 300°C) are also shown, drawn from the average water level. b) Pressure profile (after Kubota, 1988). Solid line shows the hydrostatic pressure of pure water.

Fig. 6. $\text{Cl-SO}_4\text{-HCO}_3$ equivalent composition of well discharges in the Sumikawa area.

tinuous reservoir under the Ohnuma and Sumikawa areas, in contrast to the proposal of Kubota (1988). However, the Taninainuma area, to the northeast (Fig. 2), has a distinct pressure profile, indicating the reservoir is independent from Sumikawa and Ohnuma.

The upper impermeable zone acts as a caprock to the reservoir, with a high vapor/water ratio, and also acts as a barrier to prevent infiltration of cold surface water. The distribution of the impermeable zone does not correspond with any geological formation, suggesting that the impermeable zone was formed by self-sealing (Kubota, 1988).

FLUID CHEMISTRY

Chemical data for fluids discharged from Sumikawa wells are presented in Table 1, and are plotted in Figs. 6 to 11.

The remarkable feature of fluids discharged from the Sumikawa wells is their low total dissolved solids (<3000 mg/l) compared to other geothermal systems in Japan (e.g. 16000 mg/l at Mori (Sato, 1988), 5000–8000 mg/l at Onikobe (Abe, 1988)). Also, the composition of the fluids is heterogeneous between wells. Most fluids discharged from Sumikawa geothermal wells are near neutral pH, Na–Cl type, except for fluid from well S-2 (Table 1).

The fluid discharged at different times from well S-2 can be distinguished into two groups; near neutral pH (though with a trend to slightly acidic conditions) and acidic chloride-sulfate types (Fig. 6). In this well, two feedpoints are recognized at 905 and 1065 m depth. During drilling, discharge tests were conducted separately at each feedpoint. Based on these tests, the acidic fluids issue from the deeper feedpoint, where acidic alteration minerals such as alunite and kaolinite are present (Sakai *et al.*, 1986). Such acidic fluids are also observed at Ohnuma at depths near sea level (Sakai *et al.*, 1986). Similar limited occurrence of deep acidic fluids within a system dominated by neutral pH, Na–Cl fluids is also reported in other geothermal systems (e.g. Hachobaru, Shimada *et al.*, 1985; several

Table 1 Chemical compositions of representative separated water samples from Sumikawa wells

Well	Date	WHP	Fstm	Fwat	Enth	pH	Na	K	Ca	Mg	Fe	HCO ₃	Cl	SO ₄	I	B	SiO ₂	T _{QZ}	T _{NKC}	T _{NK}	R _{CL}
S-2	82.07.10	0.0	3.2	0.8	2242	7.72	735	98.4	24.0	14.6	4.0	221	664	655	—	171	691	254	225	221	459
S-2(Acidic)	82.11.11	3.2	15.1	33.1	1115	2.61	458	99	45.7	104	474	0	544	1790	—	56	592	242	240	289	390
S-3	83.11.10	1.0	3.1	7.6	1060	8.48	180	27.6	4.0	0.9	0.09	134	155	91	0.034	66	547	236	223	239	113
S-4	86.7.18	11.6	28.2	13.4	1693	7.50	270	58.0	5.0	0.05	0.09	69.6	350	99	0.058	167	917	278	253	288	223
SM-2	78.11.17	—	6.9	18.7	1014	7.71	323	62.4	0.0	2.77	0.18	59.4	457	105	0.104	172	723	257	—	271	313
SA-1	88.11.25	4.4	7.9	1.4	2333	5.80	370	86.4	1.2	4.7	4.2	14.4	593	68	0.065	444	926	279	286	302	377
SB-1	89.11.16	1.9	9.2	15.2	1261	7.90	357	56.8	7.5	0.03	<0.1	128	440	134	0.10	197	676	252	234	244	306
SC-1	88.11.14	19.8	24.5	53.1	1123	7.30	312	53.2	0.6	1.0	0.0	49.9	442	101	0.092	169	850	271	266	253	302
SD-1	89.11.16	1.6	7.4	15.1	1148	8.00	288	45.1	9.7	0.04	<0.1	104	298	160	0.07	127	606	243	228	251	213

Chemical compositions are expressed in mg/kg at atmospheric pressure.

WHP: well head pressure (gauge pressure in bars).

Fstm: Fwat: flowrate (kg/sec) of vapor and water at atmospheric pressure.

Enth: measured enthalpy (kJ/kg).

T_{QZ}: quartz (silica) solubility geothermometer calculated assuming all cooling from steam loss (Fournier and Potter, 1982).

T_{NKC}: Na–K–Ca geothermometer with $\beta=1/3$ (Fournier and Truesdell, 1973).

T_{NK}: Na–K geothermometer from Truesdell, 1975.

R_{CL}: Chloride content in reservoir fluid calculated by the method outlined in Henley *et al.* (1984).

Fig. 7. Cl vs SO_4 composition (mg/kg) of separated water from the Sumikawa wells. Legend as in Fig. 6.

systems in the Philippines, Reyes, 1990).

There is a good negative correlation between Cl and SO_4 content in individual wells (Fig. 7); the SO_4 content decreases with increasing Cl content, except for well S-2, which has a much higher sulfate concentration and shows a positive correlation with chloride. The chemical compositions (except for S-2) can be explained by mixing of two end members; deep Cl-rich water with a small amount of SO_4 and a shallow water that has no chloride but a sulfate content of about 200 mg/kg plus a HCO_3 component (Fig. 7). The shallow water is heated to about 200°C, as will be discussed later (Fig. 9), by conductive heating and/or addition of steam.

Although the heating process is not yet clear, conductive heat transfer may be dominant in the Sumikawa geothermal system due to higher tem-

peratures compared with other geothermal systems (e.g., steam-heated 160°C water in Waiotapu, New Zealand; Hedenquist and Browne, 1989). The shallow heated water in Sumikawa is enriched in SO_4 and HCO_3 as mentioned above. Our experimental results of interaction between groundwater and altered andesite rocks from MB₁ layer in Sumikawa show that the solution after reaction at 200°C has 80 mg/kg SO_4 but nil Cl (HCO_3 was not analyzed) (MMC; unpublished data). This result supports the model; the shallow water was conductively heated and dissolved sulfate and carbonate from rocks during migration.

In contrast, the acidic fluid from well S-2 appears to be a mixture of a deep acid Cl- SO_4 water with a mixed fluid (deep chloride water plus shallow, conductively-heated sulfate-bicar-

bonate water, Fig. 6). The sulfate-chloride water in well S-2 has two trends (Fig. 6), with one having about three times the sulfate of the other, and a pH of about 2 in contrast to a pH of 5–7 for the lower sulfate trend.

The discharge tests after drilling were usually conducted for more than 100 days, though only 10 days for wells S-1 and S-2. In well SM-2, the Cl content increased from 410 to 490 mg/kg, whereas SO_4 increased from 95 to 110 mg/kg, 150 days from the start of discharge. After this period, both Cl and SO_4 contents were almost constant (MMC, unpublished). In contrast, in well S-2, Cl and SO_4 contents both increased with time after the start of discharge. In this well, an amount equal to only one third of the drilling fluid lost to the formation (14500 tons) was discharged (MMC, unpublished). Therefore, fluids discharged from well S-2 may have been influenced by the drilling water.

In 1989, a large volume of river water was injected into the Sumikawa production wells, and the fluids subsequently discharged have been geochemically monitored. From these results, approximately 70% of the injected water was estimated to have been discharged from well SB-1 within 50 days (Ueda *et al.*, 1990). These results indicate that the injected surface water is not always completely discharged during later discharge tests. In this well, the Cl content increased with decreasing SO_4 content for 5 days after the start of the post-injection discharge. After this period, both Cl and SO_4 increased. This enrichment of SO_4 shortly after the start of discharge is explained by the addition of SO_4 derived from oxidation of H_2S , based on isotopic evidence (Ueda *et al.*, 1990). Fluids from well S-4 have been monitored for 7 years, with an inverse correlation between Cl and SO_4 also observed (Fig. 7). This result strongly supports the model presented above; mixing of a deep Cl-rich water and a shallow heated SO_4 - HCO_3 rich water with nil Cl.

The quartz and alkali temperatures of the fluids studied (Fig. 8) agree within an uncertainty of about 20°C. They vary about from 220°C to 320°C within all wells sampled, and also agree

well with the measured temperature at each feedpoint (Fig. 4a). In contrast, the calculated temperature from the measured enthalpy is higher than the quartz temperature, especially in well S-4 (Fig. 8b). This indicates that the fluid discharged has excess enthalpy, i.e., there are two phases at the feedpoint or vapor addition from another feedpoint (Hedenquist, 1990; Seki, 1990). The latter case is unlikely at Sumikawa because the vapor-dominant zone exists above 400ASL, where the well is sealed by casing. The fluids from well S-4 have been monitored for 6 years and the quartz and enthalpy temperatures have a tendency to increase with time.

The enthalpy and Cl concentration in reservoir fluid (Fig. 9) are calculated by the method outlined by Henley *et al.* (1984), with the enthalpy based on the quartz geothermometer temperature. Based on an intersection of the boiling (vapor gain or vapor loss) and dilution trends which envelop all data (except for well S-2 and the excess enthalpy discharges of well S-4), the parent Sumikawa geothermal fluid has a minimum temperature of about 290°C (slightly below the maximum measured temperature), with a Cl content of only 300 mg/kg. This fluid either boils, or is diluted with heated water of nil chloride and a temperature of about 200°C. This nil chloride endmember is enriched in SO_4 and HCO_3 (Fig. 6). The deep parent fluid is estimated to be similar to fluid from well SC-1, which has the highest flowrate of all wells (57 kg/sec (200 tons/hrs) vapor and 86 kg/sec (300 tons/hrs) water). Fluid from well S-4 has a higher enthalpy than that from well SC-1 (Figs. 8 and 9). However, since well S-4 has discharged geothermal fluids with an excess enthalpy (which has not been corrected for), the observed trend for this well (Fig. 9) can be explained by vapor gain into deep fluids similar to those from well SC-1, and by dilution with water of nil chloride.

In this model, the representative deep fluid at Sumikawa has ~300 mg/kg Cl (Fig. 9). The fluid composition from all wells (except well S-2) can be explained by mixing of the deep fluid or a boiled deep fluid with shallow heated water of nil chloride and some sulfate and bicarbonate. The

Fig. 8. Geochemical temperature of fluid from the Sumikawa wells. Legend as in Fig. 6. a) Quartz and Na-K-Ca temperatures. T_{QZ} : quartz geothermometer assuming maximum steam loss (Fournier and Potter, 1982). T_{NKC} : Na-K-Ca geothermometer with $\beta = 1/3$ (Fournier and Truesdell, 1973). b) Quartz and enthalpy temperature of fluid from the Sumikawa wells. The enthalpy temperature is from the measured enthalpy at well site.

waters from both feedpoints in well S-2 have the highest chloride, though a relatively low temperature. The observed trend for well S-2 (Fig. 9) cannot be explained by a simple boiling from the

deep fluid. If it is directly related to the neutral pH Sumikawa fluid, the difference may be due to multi-step boiling of the deep fluid or dilute water. Alternatively, the high Cl (and SO_4) of S-

Fig. 9. Enthalpy and Cl diagram of reservoir fluid at Sumikawa. Legend as in Fig. 6. Enthalpy is calculated from the quartz solubility temperature, with Cl in reservoir fluid determined by the method outlined by Henley *et al.* (1984). The effect of excess enthalpy (e.g., S-4 discharges) has not been corrected for.

2 fluids might be due to HCl (and SO₂) contribution from magmatic vapors. The similarity between the acid and neutral waters from this one well suggests that these are closely related, with the acidity neutralized by water-rock interaction.

Well SC-1 discharges the highest flowrate among the Sumikawa wells. This well is characterized by a reverse temperature profile with depth below -1000 mASL (Fig. 4a). This indicates that relatively cool water inflows at this depth (Kubota, 1988). Such cold water inflow at depth in geothermal systems is common on the margin (e.g. Youngman, 1989; Hedenquist, 1990). Reyes (1990) indicated such temperature reversals in the center of Philippine hydrothermal systems, and demonstrated down flow along faults.

Fluid equilibria

Figure 10 shows a Na-K-Mg diagram (Giggenbach, 1988) to examine the degree of chemical equilibration between the fluids and their reservoir rocks and minerals. The fluid com-

positions lie on a trend from a Mg-rich component towards the full mineral-fluid equilibrium line. Most fluids are not in equilibrium with reservoir alteration minerals (e.g. feldspars, clays) that comprise the buffer curve which is plotted. In particular, the acidic Cl-SO₄ fluid from well S-2 is enriched in Mg and is the most immature liquid at Sumikawa. Its composition is similar to that expected (Mg-rich) for andesite dissolution by acid (Fig. 10). Both the neutral to weakly acid fluid and the highly acid fluid from well S-2 plot on the same trend, indicating that the neutral to weakly acid fluid formed by neutralization of the acidic SO₄-Cl fluid through simple interaction with reservoir rocks (Fig. 10) and mixing with neutral pH fluids (Fig. 6). This is independently supported by the similar chloride-enthalpy relationship for these two fluids (Fig. 9).

Fluids from SB-1 and SC-1 plot near the full equilibrium curve (Fig. 10). These fluids represent the most mature fluid in the Sumikawa geothermal system in terms of mineral-fluid equilibrium, consistent with the model that these

Fig. 10. Na-K-Mg diagram of separated water from the Sumikawa wells. Legend as in Fig. 6. The upper broken line is for full equilibrium conditions with feldspars and clays (Giggenbach, 1988). The region of immature water corresponds to the composition of waters related to rock dissolution without any control by mineral equilibria (Giggenbach, 1988).

Fig. 11. B and Cl content in separated water from the Sumikawa wells. The B/Cl mole ratio is contoured. Legend as in Fig. 6.

fluids are representative of the deep fluid at Sumikawa (Fig. 9), and supported by their isotopic compositions, described later. Therefore, the trend in Fig. 10 may be explained by dilution of the deep fluid with shallow heated water of nil chloride, in which the latter has not attained chemical equilibrium with the alteration minerals. Fluids discharged from well S-4 have been monitored over a long period. Their composition has become Mg-poor with time, shifting towards the full equilibrium line. However, the fluids collected recently are relatively enriched in Mg. These facts indicate that the composition of reservoir fluid of well S-4 is variable and susceptible to changes with time.

One of the distinct chemical characteristics of fluids at Sumikawa is the high B/Cl mole ratio (Fig. 11), compared with other geothermal systems in Japan (compiled in Fig. 2 of Chiba, 1991). The B/Cl ratio of fluid from well S-2 is relatively lower than that of fluids from other wells. Especially, the ratio of the acid fluid from well S-2 is the lowest of the Sumikawa wells and is almost the same as that of altered andesite from MB₁ layer and Yakeyama volcanic rocks in this area (MMC, unpublished), suggesting rock dissolution (Fig. 10) may be affecting the B/Cl ratio. On the basis of the experimental results of Ellis and Mahon (1964, 1967), Shigeno and Abe

(1983) demonstrated that the B/Cl ratios of fluids reflect those of the reservoir rocks. The high B/Cl ratio of most fluids at Sumikawa can be explained by a model where the fluid circulated through pre-Tertiary marine sedimentary rocks (Shigeno and Abe, 1987). In the Sumikawa geothermal system, such pre-Tertiary rocks have not been observed even in the deepest well (2486 m).

GAS CHEMISTRY

Gas compositions from the Sumikawa wells are presented in Table 2 and are plotted in Figs. 12 to 17.

The gas content varies from 0.001 mole% in the reservoir fluid for a liquid feed well (such as S-4) up to 1 mole% of total discharge in wells with high vapor/water ratios (such as SA-2). The total discharge composition of the excess enthalpy wells, however, will greatly over estimate the actual gas content of the liquid which is feeding the well (Seki, 1990). Figure 12 shows the CO₂-H₂S-Rgas (residual gas; not absorbed in alkaline solution) composition of fluid from Sumikawa and other geothermal systems in Japan. The data from the Japanese systems are taken from Table 1 in Chiba (1991), and Yoshida (1984). In plotting the gas data, the H₂S

Table 2. Chemical compositions of representative vapor fractions from Sumikawa wells

Well	Date	WHP	Fstm	Fwat	Ysep	G/S	CO ₂	H ₂ S	H ₂	CH ₄	N ₂	O ₂	Ar	He	Ne
S-2	82.07.09	0	0.97	0.17	0.851	0.0007	74.7	9.9	2.16	0.49	9.9	1.8	0.15	—	—
S-2	82.11.11	3.2	4.31	10.7	0.288	0.0012	12.8	7.08	46.5	0.32	29	0.48	0.64	—	—
S-3	83.11.10	1	0.86	2.11	0.29	0.0008	49.7	25.9	—	—	19	5.2	0.22	—	—
S-4	88.11.24	5.58	21.2	26.8	0.363	0.00031	21.7	55.1	—	—	—	—	—	—	—
SA-1	88.11.04	5.08	—	—	—	0.00131	14.3	70.8	2.54	0.07	12	0.061	0.15	0.00024	0.00073
SA-2	88.11.18	2.27	—	—	1	0.00719	78.7	18.8	1.25	0.058	1.2	0.037	0.0046	0.00046	—
SA-4	88.11.13	2.48	—	—	1	0.0136	69.9	22.2	3.29	0.25	4.2	0.077	0.066	0.0006	—
SM-2	78.11.17	1.8	2.19	5.39	0.289	0.0003	48.4	49.9	—	—	—	—	—	—	—
SB-1	89.10.12	2.0	0.95	1.66	0.393	0.00074	21.7	16.8	0.355	0.37	60	0.59	0.76	0.00073	0.0011
SC-1	88.11.09	19.2	29.8	55.9	0.348	0.00074	22	16.3	0.447	0.23	59	0.22	0.73	0.00062	0.0011
SD-1	89.11.16	1.6	7.36	15.1	0.328	0.00089	34.6	40.6	1.9	0.27	22	0.42	0.3	0.00052	0.00032

WHP: well head pressure (gauge pressure in bars).

Fstm·Fwat: flowrate (kg/sec) of vapor and water at atmospheric pressure.

Ysep: vapor fraction for gas sample.

G/S: Gas/steam volume ratio.

Gas composition is expressed in volume %.

Fig. 12. $R_{\text{gas}}\text{-H}_2\text{S-CO}_2$ in vapor fraction from Sumikawa (large triangle) and other areas (inset). Symbols from the Sumikawa wells are the same as in Fig. 6 and circle with a dot means gas from the summit of Mt. Akita-Yake (MMC, unpublished data). Symbols in the inset are; square: Matsukawa, double square: Okuaizu, open circle: Kirishima, triangle: Takigami, closed star: Onikobe. These data are taken from Table I in Chiba (1991), Kiyosu (1986) and Yoshida (1984).

and R_{gas} contents are relatively negligible in the water fraction. However, since the amount of HCO_3^- in the liquid phase may make a significant contribution to total CO_2 , the CO_2 as plotted is less than the total CO_2 to some extent, more so in low gas wells.

The Sumikawa fluids vary in gas components from well to well (Fig. 12), and are divided into four types; H_2S -poor (S-2, S-3), R_{gas} -rich (SC-1), H_2S -rich (SA-1, SA-2, SA-4) and intermediate compositions (S-4, SB-1, SD-1). Wells S-2 and S-3 were drilled before 1985 and their feedpoints are shallower (136 and 294 mASL, respectively) than other wells (below -150 mASL) except for SD-1 (215 mASL). In contrast, the feedpoint of SC-1 is the deepest (-1228 mASL) at Sumikawa, though the temperature is lower than at shallower levels (Fig.

4a). This well was drilled in 1988 by an aerated mud drilling method. During the discharge, the R_{gas} component decreased with time. The $\text{N}_2\text{-He-Ar}$ composition is similar to that of air minus oxygen (see below). Although 4×10^5 tons of fluid has so far been discharged from well SC-1, it is not clear whether such a high R_{gas} fraction is due to air dissolved in drilling fluids or due to dissolved air in the cool downflow, as mentioned before.

H_2S -rich gas is characteristic of the fluids with high vapor/water ratios (Fig. 12). The Matsukawa geothermal system, 15 km southeast of Sumikawa, discharges vapor-dominant fluid, though the gas is enriched in CO_2 (Fig. 12). H_2S -rich gas is only observed in the Kirishima and Onikobe geothermal systems (Kodama and Nakajima, 1988; Kiyosu, 1986) of Japan,

Fig. 13. CO_2 (mm/100 m)– $\text{CO}_2/\text{H}_2\text{S}$ ratio for total discharge compositions. Legend as in Fig. 6. Calculated liquid and vapor model curves are shown. This situation has been modelled assuming a 270°C liquid of composition similar to SC-1, with the parent 300°C liquid composition calculated for single step separation, as shown by Hedenquist (1990).

Fig. 14. Plot of temperature (of quartz (symbols) and Na–K–Ca (small dot) geothermometers) versus logarithm of the equilibrium constant for reaction (1), illustrating the effect of steam loss (boiling) and steam gain on CO_2 – H_2 – CH_4 gas equilibria (Giggenbach, 1980). Legend as in Fig. 6.

Fig. 15. Plot of temperature (of quartz (symbols) and Na–K–Ca (small dot) geothermometers) versus $\log \text{H}_2/\text{H}_2\text{S}$ for pyrite–Fe silicate and pyrite–pyrrhotite equilibrium (Giggenbach, 1980). Legend as in Fig. 6.

Fig. 16. N_2 -He-Ar content of gases from the Sumikawa wells. Legend as in Fig. 6 except for closed circles indicating air and air in water. N_2/Ar ratios from S-2 are also shown by arrows. The composition of magmatic gas is defined by volcanic gases of Mt. Usu, Hokkaido, Japan (Matsuo et al., 1982).

possibly related in both cases to nearby volcanic activity. The gas from an active fumarole on the summit of Mt. Akita-Yake is also enriched in H_2S (Fig. 12).

The CO_2 content in the total discharge from the Sumikawa wells is higher in fluids with high vapor/water ratios due to the preferential fractionation of gas to the vapor (Fig. 13). However, most Sumikawa gas data can be accounted for by boiling and/or a variable degree of excess enthalpy, as modelled in Fig. 13. Exceptions are wells S-2 and S-3, which are relatively depleted in H_2S (i.e., high CO_2/H_2S ratio), compared with fluids from other wells. This may be accounted for by relatively oxidizing conditions in these wells, with H_2S converted to sulfate (and perhaps some sulfide deposition). These two wells have the highest SO_4/Cl ratio of all wells in Sumikawa (Fig. 6), supporting this suggestion. Clearly, their gas compositions are distinct from the other wells.

The equilibrium constant for reaction (1) is plotted versus temperature in Fig. 14, following the method of Giggenbach (1980), with the effects of vapor gain and loss also calculated:

The analytical quotients for gases from Sumikawa are plotted versus their quartz and Na-K-Ca geothermometer temperatures. Most data lie in the region of vapor gain, despite the fact that only wells SA-1, S-4 and SB-1 in Fig. 14 are affected by excess enthalpy conditions. This indicates that equilibrium between gases in the system $H_2O-H_2-CH_4-CO_2$, according to reaction (1), has not been fully attained at Sumikawa. A similar diagram (Fig. 15) has been constructed to examine the mineral buffer control on H_2 and H_2S (Giggenbach, 1980). The data have a large scatter, particularly for individual wells, with much of the scatter possibly due to variable (often high) H_2 . This would also ac-

Fig. 17. CO_2 content in total fluid from the Sumikawa wells and the He/Ar ratio of the gas. The composition of groundwater is from our unpublished data. The He/Ar ratio of magmatic gas is taken to be 3.25 after Kiyosu (1985), and the CO_2 composition is estimated by extrapolating the linear relation between the He/Ar and CO_2 content in the Sumikawa fluids. Legend as in Fig. 6.

count for the lack of apparent equilibrium of reaction (1). In the Sumikawa system, the fluids appear to be very immature (Fig. 10), consistent with the presence of a deep magmatic component which is only slowly being neutralized by reaction with the host rock. Such reaction would tend to produce H_2 gas (including reaction of acid fluids with well casing), possibly accounting for the lack of apparent equilibrium among the gases and minerals. Such equilibrium is often noted for more "mature" geothermal systems elsewhere (Giggenbach, 1980, 1988).

Gas sources

The N_2 , He and Ar in volcanic gases in Japan are a mixture of magmatic gas and air dissolved in water (Kiyosu, 1985); the N_2/Ar and He/Ar ratios increase with increasing contribution of magmatic gas. Fig. 16 shows the N_2 - He - Ar components from Sumikawa wells, following the presentation method of Giggenbach (1986). The results indicate that these gases are a mixture of magmatic gas and air or air dissolved in water.

Figure 16 also suggests that the magmatic gas component is larger in wells with high vapor/water ratios compared to wells with lower ratios at Sumikawa. In particular, fluid from well SA-2 has a large component of magmatic gas.

The CO_2 content in the total fluid increases with increasing He/Ar ratio of the gas (Fig. 17). This indicates that the CO_2 component in the fluid is also a mixture of local meteoric water (the CO_2 content is less than 50 mg/kg) and magmatic CO_2 . The He/Ar ratio of deep magmatic gas in Japan is estimated to be 3.25 (Kiyosu, 1985). Assuming this is true for Sumikawa, the CO_2 content in magmatic gas feeding Sumikawa can be roughly estimated by extrapolation to be about 1 mole% (Fig. 17).

ISOTOPE COMPOSITIONS

The δD and $\delta^{18}\text{O}$ values of fluids from the Sumikawa geothermal system are shown in Table 3 and Fig. 18. The isotope values of total fluids are calculated from the analyzed values and the vapor/water proportions, based on the mass flow rate ratios of vapor and separated

Fig. 18. δD and $\delta^{18}\text{O}$ compositions of fluids from Sumikawa wells, calculated to total discharge values. Legend as in Fig. 6 for geothermal fluids; open circle with cross is well S-1. Small dots mean meteoric waters in the north Hachimantai region (MMC, unpublished). The straight line is the meteoric water line in this region ($\delta\text{D} = 8\delta^{18}\text{O} + 17$; Matsubaya *et al.*, 1983).

Table 3. Isotopic composition of fluids from the Sumikawa geothermal system

WELL	Sampling Date	WATER		VAPOR		TOTAL FLUID		³ H (TR)	SO ₄ $\delta^{34}\text{S}$ (‰)
		δD (‰)	$\delta^{18}\text{O}$ (‰)	δD (‰)	$\delta^{18}\text{O}$ (‰)	δD (‰)	$\delta^{18}\text{O}$ (‰)		
S-1	82. 4.28	—	—	-72.3	-10.3	-72.3	-10.3	—	—
S-2	82. 7. 9	-48.3	-5.8	-78.4	-12.0	-73.4	-11.0	—	+16.4
S-2(Acidic)	82.11. 3	-61.8	-9.5	-87.5	-14.6	-69.1	-10.9	—	+16.4
S-3	83. 7.28	-65.6	-8.8	-89.7	-13.8	-72.5	-10.2	—	+10.2
S-4	88.11.24	-65.9	-8.7	-76.1	-11.2	-71.6	-10.1	—	+15.1
SA-1	88.11.25	-60.7	-6.6	-75.0	-9.9	-72.9	-9.4	—	+15.0
SA-2	89.10.27	—	—	-68.6	-9.6	-68.6	-9.6	—	—
SA-4	89.11.15	—	—	-76.4	-11.5	-76.4	-11.5	—	—
SB-1	89.11.16	-58.7	-6.9	-83.7	-12.6	-68.2	-9.0	0.49±0.08	+8.8
SC-1	88.11. 9	-66.5	-7.6	-82.2	-11.5	-72.0	-9.0	0.39±0.08	+14.0
SD-1	89.11.16	-62.0	-7.3	-87.7	-12.9	-70.4	-9.2	0.59±0.08	+14.9

Isotopic composition of total fluid is calculated by a mass balance of vapor and water and their flow rates.

Tritium content is expressed in Tritium ratio and the error is $\pm 1\sigma$.

$\delta^{34}\text{S}$ of sulfate in separated water is also shown.

water. Although the data scatter due to the uncertainty of the mass flow rates, it is clear that the $\delta^{18}\text{O}$ value of the total fluids is up to 2‰ enriched in ^{18}O compared to local meteoric waters, similar to that observed in other geothermal systems in Japan (e.g., Otake; Mizutani, 1972). The fluids from well SC-1 and SB-1 are most enriched in ^{18}O , and the isotopic compositions from other wells can be explained by dilution of the fluids from wells SC-1 with meteoric waters (Fig. 18) according to the relationship between the $\delta^{18}\text{O}$ and Cl contents (Tables 2 and 3). These results are consistent with the observed features in Figs. 9 and 10. The isotopic composition of fluids collected at depth of two feedpoints (-1900 and -2300 m) from well SC-1 are similar, averaging -72.2 and -8.6‰ in δD and $\delta^{18}\text{O}$, respectively. These values are nearly the same as the calculated value for total discharge from well SC-1 (Fig. 18). The enrichment of ^{18}O in the deep fluid at Sumikawa is mainly due to the oxygen isotope shift during water-rock interaction, as demonstrated for other geothermal systems in the world (e.g., Craig, 1963). Despite evidence for other components having a magmatic source, the water appears to be dominantly meteoric in origin.

The $\delta^{34}\text{S}$ values of SO_4 in fluids from the Sumikawa geothermal system are $+14 \pm 1\%$, ex-

cept for well SB-1 (Table 3). Takahashi and Noda (1987) showed that the $\delta^{34}\text{S}$ value of SO_4 increases from -2 to +25‰ with increasing SO_4 concentration in thermal waters from the north Hachimantai volcanic area. This tendency is also reported in the Okiura, Ikarigaseki and Yunosawa geothermal systems, 80 km north of Hachimantai (Muraoka and Ueda, 1991). Sakai and Matsubaya (1974) studied $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values of SO_4 in thermal waters from the Green Tuff region in Japan, and found that the SO_4 is a mixture of supergene SO_4 ($\delta^{34}\text{S} = \sim -2\%$, $\delta^{18}\text{O} = \sim 0\%$), derived from oxidation of H_2S , with fossil marine SO_4 in the formation ($\delta^{34}\text{S} = +22 \pm 2\%$, $\delta^{18}\text{O} = +10 \pm 2\%$). Similar results are also observed in other geothermal systems in Japan (e.g., Hakone; Matsuo *et al.*, 1985).

In the Hachimantai area, the $\delta^{34}\text{S}$ values of SO_4 in neutral Na-Cl type thermal waters increase with increasing $\delta^{18}\text{O}$ values, while the $\delta^{34}\text{S}$ of SO_4 in the acid sulfate thermal waters is almost constant despite a variable $\delta^{18}\text{O}$ (M. Takahashi, H. Shigeno and T. Noda, unpublished). This indicates that the SO_4 in the Na-Cl type thermal waters here is also a mixture of supergene and fossil marine SO_4 . In contrast, the SO_4 in the acid SO_4 type thermal waters is formed by oxidation of H_2S , with the $\delta^{18}\text{O}$ value

later modified by an isotopic exchange reaction with water, assisted by the fact that the rate of isotopic exchange increases with decreasing pH (Chiba and Sakai, 1985).

In the Sumikawa geothermal wells, the $\delta^{34}\text{S}$ values of SO_4 in separated fluids are almost constant ($+14 \pm 1\%$), independent of variation in the SO_4 content of fluids (MMC, unpublished). This demonstrates that the SO_4 came from leaching of sulfate in the reservoir rocks, which is essentially a mixture of supergene and marine SO_4 as discussed above, independent of the deep-Cl fluid and the shallow heated SO_4 - HCO_3 -rich fluids.

The $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values of SO_4 from the Ohnuma wells and well S-4 deviate from the tendency of simple leaching of a mixture of supergene and marine SO_4 ; the $\delta^{18}\text{O}$ value is relatively depleted in ^{18}O ($\delta^{34}\text{S} = +11 \sim +16\%$, $\delta^{18}\text{O} = -4 \sim -2\%$; M. Takahashi, H. Shigeno and T. Noda, unpublished). If isotopic exchange occurs completely between SO_4 and fluid in the reservoir ($\delta^{18}\text{O} = -9 \pm 1\%$; Fig. 18) at 200 to 300°C, the $\delta^{18}\text{O}$ value of the SO_4 should be -4 to 0% , based on the isotope fractionation factor (9 to 5‰; Lloyd, 1968; Mizutani and Rafter, 1969), respectively. This result is in good agreement with the observed $\delta^{18}\text{O}$ value. According to Chiba and Sakai (1985), the half-life of the oxygen isotope exchange reaction between SO_4 and water is 40 days to 100 years in neutral pH fluid at 300°C and 200°C, respectively. This suggests that the residence time of the SO_4 in the reservoir at Ohnuma and Sumikawa should be more than several tens of years. These results indicate that, in contrast to the acid sulfate waters, the SO_4 in the neutral pH fluids from the Ohnuma-Sumikawa geothermal system is leached from a mixture of supergene and fossil marine SO_4 in volcanic rocks, with the $\delta^{18}\text{O}$ values then equilibrating with fluids in the reservoir. Therefore, the SO_4 in the neutral pH fluids is related to circulation of the meteoric system, and is not simply derived from a magmatic component. The magmatic contribution to the system is local, confined mainly to well S-2.

The tritium ratios of the fluids from the

Ohnuma-Sumikawa wells are lower than the analytical detection limit (<0.3 TR), except for fluids collected earlier during discharge, whereas those of meteoric waters in this region are approximately 10 TR (MMC, unpublished data). As mentioned before, the fluids from the Ohnuma-Sumikawa geothermal wells are of meteoric origin. Therefore, the residence time is estimated to be more than 100 years on the assumption that the system is closed. These results support the view that the $\delta^{18}\text{O}$ values of SO_4 in fluids from the Ohnuma-Sumikawa wells are in equilibrium with the fluids in the reservoir.

CONCLUSIONS

The geochemical characteristics of the Sumikawa geothermal system include the notably low salinity compared to other Japanese geothermal systems, and the chemical variation from well to well. The neutral pH fluids are derived from a deep fluid (~ 300 mg/kg Cl) or a boiled deep fluid diluted with relatively shallow heated water that contains sulfate but nil chloride. Most fluids from Sumikawa are not in equilibrium with reservoir alteration minerals. The waters are essentially of meteoric origin, though there appears to be a magmatic contribution to the volatiles, and maybe to the deep acid Cl- SO_4 fluids. The fluids from Sumikawa are immature based on all indicators of chemical equilibria, probably due to the youth of system and the magmatic flux of volatiles. In particular, the magmatic component to the system is localized, with most of the SO_4 in the neutral pH fluids derived from leaching of a mixture of supergene and fossil marine SO_4 in the meteoric-dominated system. Thus, the magmatic components are superimposed on a steady-state meteoric convection cell.

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