Fluctuation of chemical compositions of the phase-separated hydrothermal fluid from the North Fiji Basin Ridge

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ABSTRACT

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On the central ridge of the North Fiji Basin (southwest Pacific), hydrothermal activities were located in 1989. It was the second discovery of the hydrothermal fluid undergo phase separation. During a dive mission of *Shinkai* 6500 in 1991, we revisited the hydrothermal area and collected fluid samples from the same anhydrite chimney. Fluid composition of the end member shows no significant changes during this two years. Other fluid ventings 150 m apart show similar chemistry, suggesting that the phase separation and segregation processes occur throughout the hydrothermal area. Small fluctuation in chemical composition of the fluids from two sites is attributed to subseafloor fluid–rock interaction after the phase segregation.

1. Introduction

Following the discovery of the ASHES vent field in the Juan de Fuca Ridge (Massoth et al., 1989), hydrothermal activities located in the central ridge of the North Fiji Basin reconfirmed phase separation process during hydrothermal circulation. Hydrothermal fluids collected from an anhydrite chimney showed quite similar geochemistry to vapor-rich fluids from the ASHES field, which represented important evidence for subcritical boiling (Grimaud et al., 1991). These successive discoveries suggest that phase separation is a rather ubiquitous process in hydrothermal systems at relatively shallow depth.

Butterfield et al. (1990) studied fluid geochemistry from the ASHES vent field in detail and revealed characteristics of the fluid which undergoes phase separation. They proposed a vapor-liquid-seawater mixing model for fluid history at the ASHES vent field. Hydrothermal fluid rising through the ocean crust has generated vapor and liquid phases, which have become physically segregated and followed different pathways to the seafloor, cooling and mixing to varying degrees along the way with cold seawater percolating into an interstitial matrix (Butterfield et al., 1990). With respect to the mechanism of phase segregation, Butterfield et al. (1990) and Fox (1990) presented and discussed their own models. With a view to elucidate the phase separation process in submarine hydrothermal systems, it is important to accumulate clues to the constraint of models by studying the geochemistry of other cases.

In September/October 1991, two years after the previous dive mission by *Nautile*, we revisited the central ridge of the North Fiji Basin employing the Japanese submersible *Shinkai* 6500 (Auzende

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0.R.S.T.O.M. Fonds Documentaire N°: 43149Cote: 2 ex 1. et al., 1992). Seven dives were devoted to exploration of the hydrothermal area in the station 4 at $16^{\circ}58$ 'S (Fig. 1). Fluid samples were collected from the active venting, which had been a main target of the previous mission and also from other chimneys in that vicinity. In this paper, we report the chemical composition of the hydrothermal fluids and discuss their fluctuation during two years and among ventings in a 150 m wide hydrothermal area.



Fig. 1. (a) Regional map showing the central ridge of the North Fiji Basin. A star mark indicates location of the station 4. (Modified from Auzende et al., 1990.) (b) Structural and geological map of the station 4. (After Urabe et al., 1990.) (c) Detail map of hydrothermal fluid venting area. Marks indicate chimneys from which fluid samples were collected. Hatched area shows distribution of active vent community.

2. Description of sampling site

The anhydrite chimney "White Lady" was easily reconfirmed, in spite of some modification of its figure (Auzende et al., 1992; Fig. 2). The transparent hydrothermal fluid was emanated vigorously as two years ago. From our impression of observation, the fluid flux became more powerful and the living animal colonies composed of gastropods, mussels, crabs, galatheans and cirripeds seemed to extend to all around the mound and to the top. The highest fluid temperature was measured as 265°C by a platinum thermometer. It was 20°C lower than the maximum temperature recorded in 1989, although some uncertainty remains due to the use of different probes.

During the 1991 dive mission, other active ventings were located about 150 m southwest of the White Lady (Fig. 1c). Fluid samples were collected from two chimneys, "Kaiyo" and "LHOS". The "Kaiyo" chimney of 2-m height stands on a steep slope and emits transparent fluid. This chimney was identified as discovered by a towing video camera survey during the 1988 Kaiyo Cruise. Samples were collected from the foot of the chimney and its exit temperature was 290°C. The "LHOS" chimney with three peaks at its top was located near а sinker of the Long-term Hydrothermalism Observation System (LHOS) deployed during the 1989 cruise. (Results obtained by LHOS will be reported in K. Mitsuzawa et al., in prep.) Transparent fluid venting from a crack of the chimney was collected. The measured temperature during the sampling was 230°C at maximum.

3. Methods

3.1. Sampling equipments

Two different types of water samplers were used for this study. One was a titanium syringe which



Fig. 2. Sketch of White Lady chimney. (a) observed by the Nautile in 1989; (b) observed by the Shinkai6500 in 1991 (after Auzende et al., 1992).

is used conventionally with the French submersible *Nautile*, also during the previous 1989 dive mission. The submersible's manipulator carries the sampler to a venting orifice and hydrothermal fluid is drawn into the syringe as a piston moves by spring force after trigger release.

The other was a pumping system which was developed for the Japanese submersibles Shinkai 2000 and Shinkai 6500 (Sakai et al., 1990). Its scheme is illustrated in Fig. 3. The sampler consists of a pump, a revolving valve, eight sample syringes made of acrylic resin and an inlet tube. As the pump expelled distilled water in one room of the syringe, sample fluid is introduced into the other room which is separated with a piston. With rotation of the revolving switch valve, the next sample syringe becomes to be ready. The inlet tube which is connected to the sample syringes by a 2-m long teflon tubing was attached on the manipulator. Close to its top, a platinum thermometer was fixed to monitor the sample fluid temperature. The inlet tube was kept on the venting orifice till filling up four sample syringes, in order to replace distilled water and/or ambient seawater which has occupied the inlet tube and the connection. Except for the first syringes, distilled water in dead volume of the sample room was less than 3% of the obtained sample fluid. Moreover, the sample syringe can keep an extra volume for gas evolution by half-way stop of the piston. In this case, evolved gas pushes up the piston to compensate its pressure increase and leakage of gas phase is avoided. We can check how gas evolves in the sample syringes before sample handling. However, in this study, significant gas evolution was not observed.

3.2. Analytical procedure

After submersible recovery, samples were transferred and processed immediately in a laboratory of the tendership *Yokosuka*. Measurement of *p*H and titration for sulfide determination were completed within half an hour. Alkalinity determination for the 0.45 μ m filtered aliquot was conducted within one hour. Concentrations of chloride and sulfate were analyzed on board within a week using ion chromatography after adequate dilution of a filtered aliquot. Aliquots for cations were brought to both Japanese and French laboratories after filtration and acidification, and analyzed using the methods listed in Table 1. Results of most species were cross checked and showed good agreement. Aliquots for gas components were



Fig. 3. A schematic diagram of the pumping sampler: (A) before sampling; (B) during sampling; (C) after sampling, fluid = sample fluid, D.W. = distilled water.

TABLE 1

Chemical composition of the samples collected during the 1992 dive program

Component -	Method ^a	W.L. ^b 77-Ti ^c	W.L. 77-6	W.L. 78-Ti	Kaiyo 80-Ti	Kaiyo 80-3	Kaiyo 80-4	LHOS 93-2	LHOS 93-3	LHOS 93-4
Li (µmol/kg)	FAAS	20	19	150	25	82	255	28	160	107
Na (mmol/kg)	I.C.	461	439	303	459	351	276	451	348	386
K (mmol/kg)	I.C.	10.2	9.7	10.2	10.7	10.3	13.9	9.8	12.5	11.8
Rb (µmol/kg)	FAAS	1.7	1.8	7.8	1.9	7.8	n.a.	2.0	10.0	7.3
Mg (mmol/kg)	I.C.	52.4	49.8	18.5	52.0	31.5	8.5	51.0	24.4	33.5
Ca (mmol/kg)	I.C.	9.98	9.51	7.61	9.86	8.86	8.98	9.93	10.03	9.73
Sr (µmol/kg)	FAAS	85	88	49	75	60	47	77	63	64
Ba (µmol/kg)	ICP	0.57	0.15	3.32	0.70	3.05	4.40	0.13	2.88	1.85
Mn (µmol/kg)	ICP	0.12	0.13	6.56	0.51	8.04	21.3	0.42	15.2	10.0
Fe (µmol/kg)	FAAS	0.6	0.5	3.7	0.6	4.0	8.2	0.5	3.7	2.4
B (mmol/kg)	ICP	0.447	0.422	0.462	n.a.	0.412	0.463	0.417	0.449	0.452
Al (µmol/kg)	ICP	< 0.1	0.7	3.5	< 0.1	2.3	5.1	0.4	3.4	2.2
Si (mmol/kg)	ICP	0.14	0.14	8.45	0.27	4.33	11.3	0.25	7.37	4.81
Cl (mmol/kg)	I.C.	564	539	357	556	420	318	538	401	452
Br (µmol/kg)	HPLC	848	767	547	838	600	492	809	613	667
SO ₄ (mmol/kg)	I.C.	28.9	27.6	10.0	27.8	16.7	4.5	26.7	13.3	18.1
pH (25°C)		7.85	7.02	4.98	7.77	5.40	4.80	7.18	5.30	5.30
alk. (mmol/kg)	titr.	2.40	2.28	0.50	2.45	0.65	0.02	2.12	0.28	0.79
H ₂ S (mmol/kg)	titr.	< 1.12	< 1.84	3.32	n.a.	n.a.	n.a.	n.a.		
CO ₂ (mmol/kg)	coul.	n.a.	2.23	9.65	2.27	5.02	10.2	2.34	6.39	5.36

^aAnalytical method (precision): coul.=coulometer (CO₂: 3%); FAAS=flameless atomic absorption spectrometry (Li: 2%, Sr: 2%, Rb: 2% and Fe: 5%); HPLC=high pressure liquid chromatography (Br: 5%); I.C.=ion chromatography (Na: 3%, K: 3%, Mg: 3%, Ca: 3%, Cl: 3% and SO₄: 3%); ICP=inductively coupled plasma atomic emission spectrometry (Ba5%, Mn5%, B3%, Al5%, Si3%); titr.=potentiometric titration (alk.: 0.5% and HS: 1%). ^bW.L.=White Lady.

^cSample name of "-Ti" indicates the sample collected by the titanium syringe and others were sampled by the pumping system. ^dn.a.=failed in analysis.

transferred into glass vials capped with rubber septum without head space. They were poisoned by mercury chloride and stored in a refrigerator until analysis.

4. Results and discussion

4.1. Chemical composition of hydrothermal end member

Analytical results of nine samples collected during the 1991 dive mission are given in Table 1. Figure 4 illustrates the relationship with magnesium concentration, in order to estimate end member values according to the conventional method (Edmond et al., 1982). In these magnesium diagrams, data of samples collected in 1989 from the White Lady are also plotted (data from Grimaud et al., 1991).

Samples collected in 1991 from the White Lady are plotted close to a regression line formed by the 1989 samples (shown as a solid line). Although only one sample is available for the estimation, it indicates the same end member value with respect to all the species. This result insists that the White Lady fluid has not changed its chemical composition during this two-years period.

On the other hand, samples from the southwestern site, the Kaiyo and the LHOS chimneys, seem to have a different composition from the White Lady fluid. Magnesium diagrams (Fig. 4c-f) show another linear relationships (shown as a broken line), while the difference is not so clear with respect to the Cl-Mg diagram (Fig. 4b).



Fig. 4. Plot of elemental concentrations versus magnesium for fluid samples. Symbols used in the figure: $\bigcirc =$ White Lady in 1989 (from Grimaud et al., 1991); $\bigcirc =$ White Lady in 1991; $\triangle =$ Kaiyo; $\square =$ LHOS. Solid line and broken line show regression of the White Lady 1989 samples and the Kaiyo-LHOS samples, respectively. (a) Sulfate; (b) chloride; (c) potassium; (d) lithium; (e) silica; and (f) carbon dioxide.

TABLE 2

Chemical composition of the end member fluids of the North Fiji Basin and comparison with the ASHES hydrothermal system

Component	North Fiji	Basin	ASHES °		Seawater ^b
	White ^a Lady	Kaiyo ^b + LHOS	Virgin Mound	Inferno	
T (°C)	285	291	299	328	<u> </u>
Li (µmol/kg)	200	282	184	636	24
Na (mmol/kg)	210	239	148	499	457
K (mmol/kg)	10.5	14.5	6.98	26.8	10.2
Rb (µmol/kg)	8.8	17.1	31	28	
Mg (mmol/kg)	0	0	0	0	51.8
Ca (mmol/kg)	6.5	9.0	10.2	46.8	9.9
Sr (µmol/kg)	30	43	46	192	0.432
Ba (µmol/kg)	5.9	5.3	6	26	
Mn (µmol/kg)	12	26	142	1150	
Fe (µmol/kg)	13	8.8	12	1065	
B (mmol/kg)	0.465	0.475	0.45	0.59	0.432
Al (µmol/kg)	6.0	6.0	5.3	4	
Si (mmol/kg)	14.0	13.3	13.5	15.1	0.14
Cl (mmol/kg)	255	267	176	624	552
Br ^d (µmol/kg)	306	407	250	956	832
SO ₄ (mmol/kg)	0	0	0	0	27.8
pH (25°C)	4.7	4.7	4.4	3.5	7.6
Alk. (mmol/kg)	0.12	-0.87	0.66	-0.48	2.32
H ₂ S (mmol/kg)	2.0	4.0	18	7.1	
CO ₂ (mmol/kg)	14.4	11.1	285	50	2.3

^aGrimaud et al. (1991). ^bThis study. ^cButterfield et al. (1990). ^dCampbell and Edmond (1989).

Composition of the common end member of the southwestern site is calculated by extrapolation to an assumed value of zero magnesium, and listed in Table 2 together with that of the White Lady fluid.

4.2. Phase separation process in submarine hydrothermal system

Bischoff and Rosenbauer (1984, 1988) investigated the phase relations of seawater experimentally and indicated that the two-phase boundary of seawater coincides exactly with that of 3.2 wt%of NaCl in H₂O system (Fig. 5). When a fluid encounters the phase boundary at temperatures below the critical point, subcritical boiling forms nearly pure water vapor phase from saline liquid phase (brine). Until the boiling temperature approaches to within $10^{\circ}-15^{\circ}$ C of the critical point, the vapor phase contains much less salt than the liquid phase. On the other hand, when a fluid crosses the boundary at temperatures higher than the critical point, the result of supercritical phase separation is condensation of a small amount of highly saline brine from a vapor-like fluid. In either case, phase separation process causes a drastic chlorinity change of hydrothermal fluid.

Since the discovery of high-temperature fluid venting, it has been an argument on phase separation process in submarine hydrothermal system. A variety of evidence, especially wide chlorinity variation is attributed as supporting that venting fluid has experienced boiling during hydrothermal circulation. Von Damm and Bischoff (1987) demonstrated high chlorinity and other cation concentrations of fluids from the Southern Juan



Fig. 5. Two-phase boundary curve for 3.2 wt% NaCl in H_2O system (Data from Bischoff and Rosenbauer, 1988). Each mark indicates the exit temperature of fluid venting at the seafloor: •=Kaiyo site in the North Fiji Basin; = Virgin Mound in the ASHES vent field; \Box =Inferno in the ASHES vent field (from Butterfield et al., 1990).

de Fuca Ridge, and proposed a model of mixing between hydrothermally evolved seawater, and the vapor and brine phases produced during phase separation. Contrary to this, Campbell and Edmond (1989) insisted that a single phase, nonfractionating process accounts for controlling chlorinity variability of most of submarine hydrothermal fluids including the Southern Juan de Fuca Ridge fluid, based on their similar Br/Cl ratio to seawater.

The first unequivocal identification of phaseseparated effluents from a seafloor venting system was reported by Massoth et al. (1989). They found a variety of fluid chlorinity which ranges from 35% to 115% of seawater, within a 60-m diameter of the ASHES vent field in the Juan de Fuca Ridge. Butterfield et al. (1990) studied the fluid chemistry in detail and pointed out their unique characteristics, as (1) wide range of chlorinity and covariation of most major elements concentration, (2) inverse correlation of gas content and chlorinity and (3) depletions in iron of vapor-rich fluid probably due to iron-sulfide precipitation during phase separation. They demonstrated that the entire range of fluid compositions are best explained by subcritical phase separation while fluid rising through the oceanic crust, followed by partial segregation of two phases.

4.3. Chemical characteristics of vapor-rich fluid from the North Fiji Basin hydrothermal site

Table 2 presents data of end member fluid compositions of the North Fiji Basin hydrothermal site and the ASHES vent field for comparison. The Virgin Mound and the Inferno in the ASHES field are representative of the vapor-rich fluid and the brine-rich fluid, respectively.

Both the White Lady fluid and the Kaiyo-LHOS fluid show similar characteristics to the vapor-rich fluid in the ASHES vent field. Their chlorinity is 47-49% of seawater and sodium concentration is proportional to the chlorinity dilution. The CO_2 enrichment is not so considerable as the ASHES fluids, but it is notable. The iron concentration is significantly low. The depletion of metals probably accounts for the formation of the anhydrite chimney of "White Lady", as in the case of the Virgin Mound in the ASHES field. The fluid chemistry of the North Fiji Basin hydrothermal site strongly supports that it has experienced phase separation and segregation during hydrothermal circulation, although counterpart brine-rich fluids have not been observed in this area.

The North Fiji Basin fluids do not show a high CO_2 concentration, in comparison with the Virgin Mound fluid where significant gas enrichment due to gas partition according to Henry's law has been observed (Butterfield et al., 1990). Escape of gas components during sample recovery is likely denied from the good linear relationship of most samples in the CO₂-Mg diagram (Fig. 3f). One possible process for a decrease in the CO₂ concentration of vapor-rich fluid is boiling in an open system. In such a case, gas components would distribute into the first fraction of vapor phase and be removed to elsewhere. As the process progresses, the final fraction of vapor phase would become deplete in gas components. An alternative more plausible explanation is that the ASHES hydrothermal fluids are extremely enriched in gas components even if they do not undergo phase separation. Butterfield et al. (1990) proposed that magmatic exsolution of a CO₂-rich fluid and injection into the circulating hydrothermal fluids may be responsible for the gas enrichment of all of the ASHES fluids. They estimated that the Virgin Mound fluid of 285 mmol/kg CO_2 is induced by 15-20% boiling in a closed system from single phase fluid of 100 mmol/kg CO_2 . Assuming the same boiling ratio, the original single-phase fluid in the North Fiji Basin hydrothermal site would contain CO_2 in a concentration of about 10 mmol/kg. This range is comparable to other mid-oceanic ridge hydrothermal systems. The CO₂ concentration of the North Fiji Basin system would be in a proper range for vapor-rich fluid.

Also with respect to physical properties, the North Fiji Basin fluids show similar tendency to the ASHES fluids. Exit temperatures of venting fluids are $230^{\circ}-290^{\circ}$ C, which are much lower than the boiling temperature at the seafloor condition (Fig. 5). As discussed in Butterfield et al. (1990), this observation strongly suggests subseafloor cooling by admixture of cold ground seawater while fluid ascending in the upflow zone after phase separation and segregation. Vapor-rich fluid would be subject to such subseafloor processes.

Since the North Fiji Basin hydrothermal system is located in tectonic context of a marginal basin, it would be interested in comparison of their fluid composition with that of the Lau Basin, which is a back-arc basin in the Southwest Pacific. Table 3 summarizes the chemical comparison of hydrothermal end member composition including data of the mid-oceanic ridge hydrothermal system. Although it is difficult to compare them directly due to dilution by the phase separation process,

TABLE 3

Comparison of vent fluid composition with other hydrothermal systems

Component	North Fiji Basin	Lau Basin ^a	EPR21N ^b	SIFR°	MAR ^d
<i>T</i> (°C)	285-291	334	273/354	300/350	335/350
Li (µmol/kg)	200-282	580-745	891-1322	1110-1810	843-849
Na (mmol/kg)	210-239	520-615	432-510	660-800	510
K (mmol/kg)	10.5-14.5	55-80	23.2-25.8	37.3-51.6	24
Rb (µmol/kg)	8.8-17	60-75	27-33	28-37	11
Ca (mmol/kg)	6.5-9.0	28-41	11.7-20.8	77.3-96.4	10
Sr (µmol/kg)	30-43	105-135	65-97	230-312	50
Ba (µmol/kg)	5.3-5.9	20-60	~ 10	5-35	
Mn (µmol/kg)	12-26	5800-7100	700-1000	2610-4480	490
Fe (µmol/kg)	9–13	1200-2900	750-2400	10,300-18,700	1830-2180
Fe/Mn	0.3-1.1	0.2-0.4	0.9-2.9	4.0-5.2	0.4
B (mmol/kg)	0.46-0.47	0.77-0.87	0.50-0.55°	0.49	0.52-0.53
Si (mmol/kg)	13.3-14.0	14.0	15.6-19.5	22.7-23.3	18.2
Cl (mmol/kg)	255-267	650-800	489-579	896-1090	559
Br ^f (µmol/kg)	306407		802-929	1422-1832	847
$Br/Cl \times 1000$	1.20-1.52		1,.60-1.69	1.59-1.68	1.52
pH (25°C)	4.7	2	3.3-3.8	3.2	3.7-3.9
alk. (mmol/kg)	-0.87 - +0.12		-0.50 - 0.19	~0	-0.24 - 0.06
H ₂ S (mmol/kg)	2.0-4.0		6.57-8.37	3.0-4.4	5.9
CO ₂ (mmol/kg)	11.1-14.4		5.7 ^g		

^aLau Basin, Valu Fa Ridge; Fouquet et al. (1991).

^bEast Pacific Rise 21°N; Von Damm et al. (1985).

Southern Juan de Fuca Ridge; Von Damm and Bischoff (1987).

^dMid-Atlantic Ridge, MARK site; Campbell et al. (1988b).

Spivack and Edmond (1987).

^fCampbell and Edmond (1989).

^sCraig et al. (1980).

the North Fiji Basin fluids seem to show a close chemistry to that of the mid-oceanic ridge systems according to comparison of ratio among major elements. Common chemical characteristics among hydrothermal systems in back-arc tectonic setting is unlikely to be confirmed, because the Lau Basin fluid show a unique chemistry due to its strong acidity (pH=2) and high chloride concentration. Such signature would appear in chemical abundance of trace elements and/or isotopic composition of gas species.

4.4. Fluctuation of chemical composition during two years

As described in the section 4.1, the White Lady fluid shows no significant changes of chemical composition from 1989 to 1991. Also in the case of the ASHES vent field, Butterfield et al. (1990) reported no composition change of the vapor-rich fluids during two years. Hydrothermal fluid which undergoes phase separation would retain its chemical composition steady in such time scale.

Campbell et al. (1988a) reported a 6-year time series study of fluid compositions obtained from the EPR 21°N field and the Guaymas Basin. Most of the fluids showed stable composition on a decadal time scale. They considered the result as consistent with the rock buffering model where equilibrium between fluid and a green schist facies mineral assemblage control the major elements composition of the fluid.

A hydrothermal system where phase separation and segregation occurred, would have opportunities to change the fluid composition drastically. A change of segregation ratio between the vapor and brine phase and/or a change of the mixing ratio between the vapor phase and circulated seawater at its recondensation induce a substantial change of chlorinity and concentration of major elements. However, our results indicate that these parameters are rather stable. The physical structure of hydrothermal circulation, for instance represented by porosity would be maintained in such a time scale. This problem is of interest, although its mechanism is not unknown at present.

4.5. Fluctuation of chemical composition between two sites

The hydrothermal fluids in the ASHES vent field show a diverse range of chlorinity, although the fluid ventings distribute within a small area of 60 m in diameter. Butterfield et al. (1990) revealed a good correlation between end member cation concentration and end-member chloride concentration for the individual vents, which suggests a simple mixing of a brine with a vapor of near-zero salinity (Fig. 6). The entire range of the ASHES fluid composition was attributed to phase segregation of varying degrees.

In the case of the North Fiji Basin fluids, phase separation and segregation process control fluids chemistry in the 150 m wide hydrothermal field of station 4. However, fluctuation of chemical composition between two sites, the White Lady and the southwestern ventings (Kaiyo–LHOS), seems not to be so simple. In spite of the clear difference of some cations such as potassium and lithium, both chloride and sodium concentrations do not show quite a difference between the two sites. Moreover, the relationship between the two North Fiji Basin end members does not agree with the mixing line which contains a zero salinity component (Fig. 6).

As discussed in Butterfield et al. (1990), subseafloor fluid-rock interaction after phase segregation would modify the fluid composition considerably. While fluid ascending in the upflow zone, vapor-rich fluid would experience hydrolysis of silicate minerals and/or admixture of cold seawater. Lower exit temperature of the vapor-rich fluids is in accordance with such subseafloor processes. The SiO₂ concentration of both the White Lady and the Kaiyo-LHOS fluid were in a range of quartz saturation at seafloor depth and fluid exit temperature. This result expresses clear evidence for interaction occurs subsequent to phase separation probably in the upflow zone. However, it is difficult to discriminate other effects, such as an increase of alkalinity and concentrations of extractable elements which were referred as additional evidence for the subseafloor interaction in the ASHES vent field (Butterfield et al., 1990). Although it is difficult to confirm from data of



Fig. 6. Plot of end member elemental concentrations versus end member chloride concentration for individual vents. Circles and crosses indicate data of the North Fiji Basin fluid and the ASHES vent field fluid (from Butterfield et al., 1990), respectively. Value of seawater is plotted as a small circle. (a) Sodium; (b) potassium; and (c) bromide.

only two venting fluids, that subseafloor fluid-rock interaction is plausibly responsible for the fluctuation of the chemical composition in the hydrothermal site.

5. Conclusion

We revisited hydrothermal sites on the North Fiji Basin Spreading Ridge and collected fluid samples two years after the previous study. Fluid chemistry is characterized by low chlorinity, enrichment in gas species and depletion in iron. The result insists evidence for phase separation and phase segregation process during hydrothermal circulation, although only vapor-rich fluids were confirmed in this site.

The phase separation and segregation seems to control the composition of the fluids venting from over the 150 m wide hydrothermal site, based on their common fluid chemistry. The small fluctuation of the chemical composition among them was attributed to consequence of subseafloor fluidrock interaction. No significant change of fluid composition from the same venting during two years implies a steady progress of the phase separation and segregation process in such time scale, although their mechanism is not well known.

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Fluctuation of chemical compositions of the phase-separated hydrothermal fluid from the North Fiji Basin Ridge

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