Gas geochemistry of submarine hydrothermal systems at Okinawa backarc basin and Ogasawara arc

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Introduction

Since 1984, a number of active submarine hydrothermal systems have been located along the rifting axes of the middle to eastern Okinawa Trough Backarc Basin and the submarine chains of Izu-Ogasawara Arc volcanoes. The hydrothermal solutions from the mid-Okinawa Trough are extremely rich in carbon dioxide (up to 200 mM/kg), with methane and hydrogen sulphide each comprising about a few to 10 % of the former(Sakai et al., 1990). Carbon dioxide hydrate was observed to form at the sediment/seawater interface. In 1992, using 'Shinkai 2000', a Japanese submersible, we again collected hydrothermal fluids from two active fields in the mid and eastern Okinawa Trough (OKT) and one field of Suivo Seamount(SY) of the Ogasawara Arc(OGA). The water samples were analyzed for He, H₂, N₂, NH₃, Ar, CH₄ and CO₂ and the isotopic compositions of He, C of CO₂ and CH₄ and S of H₂S and sulphate. Based on the results of these new analyses, the origins of carbon and nitrogen emitted from the hydrothermal fields will be discussed here.

Methods

Hydrothermal solutions were sucked into 750 ml Plexiglas syringes, using pump-driven pistons. Onboard the mother ship, gases were separated from liquid by expansion into pre-evacuated stainless steel bottles. In a land-based laboratory the CO_2 and H_2S in the gas samples were separated from other gases using liquid nitrogen trap, and their amounts and isotopic ratios were measured. Noncondensable gases(R-gas) were then gas chromatographically analyzed. The CH_4 in the R-gases was combusted to carbon dioxide and their carbon isotopic ratios were determined. The helium isotope analysis was carried out on aliquots of samples which were stored in metal bottles.

Results and discussion

Table 1 summarizes chemical compositions of volatiles and Mg in OKT (Clam sites, Minami Ensei Knoll) and the OGA volcano (SY). Similar data of the Southern Juan de Fuca Ridge hydrothermal solutions(Evans *et al.*, 1988) are also shown for comparison. The OKT hydro-

TABLE 1. Concentration and composition of volatile components in OKT and OGA hydrothermal solutions. The data of the Southern Juan de Fuca after Evans et al. (1988)

Locality	depth meter	T ℃	Concentration (mM/kg)					R-gas composition (V%)				
			Mg	CO ₂	NH_{4}^{+}	H ₂ S	R-gas	He	H ₂	Ar	N_2	CH ₄
Clam site 1	1390	102	37	62.2	3.37	4.77	4.5	0.002	0.93	0.36	18.3	80.4
ditto	1390	112	36	89.4	2.76	6.73	7.85	0.009	0.06	0.27	14.3	85.4
Clam site 2	1390	124	27	149.8	5.59	9.66	11.8	0.002	2.38	0.14	9.6	87.9
M. ensei 1	692	278	1	65.1	4.68	1.3	3.07	0.002	0.45	0.2	14.3	85.1
M. ensei 2	700	276	0.4	66.2	4.7	1.94	3.34	0.001	0.42	0.14	8.5	90.9
M. ensei 3	692	265	0.5	96.3	4.56	1.52	7.52	0.008	0.18	0.08	7.4	92.3
Suivo Smt 1	1390	300	4.5	33.7	0	1.4	0.6	0.08	3.35	1.86	77.1	17.6
ditto	1390	300	2.1	42.1	0	1.2	1.1	0.066	3.44	1.79	77.1	17.7
Suivo Smt 2	1390	300	1.6	33.8	0	1.2	0.5	0.092	3.09	1.82	76.1	18.9
S. J. de Fuca	2300	350	0	3.92	0	3.51	0.88	n.d	39.7	1.1	49.6	9.5



FIG. 1. C-³He-N ternary plot of the OKT and OGA gases. C and N are the total carbon (CO₂ + CH₄) and the total nitrogen $(2 \times N_2 + NH_4)$, respectively. terrestrial examples are represented by White Island, N. Z. (Giggenbach, 1993).

thermal fluids are highly enriched in carbon $(CO_2 + CH_4)$ and nitrogen $(NH_4^+ + N_2)$ as compared with MOR hydrothermal systems (except for sediment-hosted ones in the Guaymas Basin), whereas SY is rich in carbon but nitrogen is essentially non-existent. The difference in carbon geochemistry between OKT and OGA is further demonstrated in the carbon isotopic ratios of CO₂ and CH₄; δ^{13} C of CO₂ and CH₄ at OKT are -4.8(Clam and M. ensei) and -32.7(Clam), -29.5 (M. ensei) permil PDB, respectively, while those at SY are -0.9 and -8.5 permil, respectively. The carbon in the latter is much heavier than the mantle carbon encountered at MOR, but the isotope fractionation between CO₂ and CH₄ is similar to MOR, suggesting their origin at deep, high temperature environments. On the contrary, methane from OKT has δ^{13} C typical of thermogenic origin and carbon dioxide has definitely higher $\delta^{13}C$ than that at MOR.

Figure 1 indicates that gases from OKT are mixtures of the mantle component, of which $C/^{3}$ He ratio was assumed to be equal to that of MORB or 2×10^9 , and the crustal components in which C/N ratios range from 12 to 20, whereas at SY the crustal component is nearly N-free as was already mentioned above. The low C/N ratios in OKT hydrothermal fluid is attributed to decomposition of organic matter in sediment-hosted hydrothermal fields. Assuming three major sources of carbon in these hydrothermal systems, that is, mantle carbon (mtlC), slab carbon(slbC) which is carbonate carbon in subducting layer 2 basalt and was formed by low temperature alteration during its journey on seafloor, and carbon in sediments which cover the active seafloor. The last category of carbon consists of marine carbonate(ccC) and organic carbon (orgC). Assuming appropriate values for essential parameters, such as $C/{}^{3}$ He, C/N and Ar/N of these components, the contribution of mtlC, orgC and slb/ccC in the hydrothermal solutions are estimated to be a few to 10, 30 to 50, and 50 to 70 %, at OKT, and 30 to 40, 1 to 3, and 58 to 69 % at OGA, respectively. Although these figures must be taken as tentative ones, they clearly indicate importance of recyclic carbon in carbon budget in the hydrothermal systems along the Western Pacific plate margins.

References

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