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# Estimation of fluxes at mid-ocean ridges and geochemical cycles on the Earth's surface of sulfur and halogens

Takanori Kagoshima<sup>1\*</sup>, Yuji Sano<sup>1</sup>, Naoto Takahata<sup>1</sup>, Bernard Marty<sup>2</sup>

<sup>1</sup>Atmosphere and Ocean Research Institute, the University of Tokyo, <sup>2</sup>Centre national de la recherche scientifique

## [Introduction]

Volatile elements on the Earth's surface have been accumulated by degassing from the solid Earth. Noble gases have been used as tracers to constrain the degassing history. Such studies suggested that the most part of the atmospheric argon had been accumulated in the early Earth [1], and estimated carbon and nitrogen cycles by comparisons with well determined helium-3 (<sup>3</sup>He) fluxes from the inside to the surface of the Earth [2, 3]. They constrain the evolutionary history of the Earth's surface. On the other hand, though sulfur (S) and halogens (fluorine (F) and chlorine (Cl)) form various compounds on the Earth's surface and significantly affect the environment, their geochemical cycles are not well understood. Fluxes of S and halogens at MORs (Mid-Ocean Ridges), where material migrations are dominant because of intensive volcanic activites, have been estimated based on compositions of quenched glassy rims of MORBs (Mid-Ocean Ridge Basalts) which retain those of magmas derived from the upper mantle. However, they may be overestimated [4], and plausible estimations based on compositions of both silicate melts and fractions released as hydrothermal fluids are needed. We determined compositions of hydrothermal fluid and melt components retained respectively in vesicles and solids of MORB glasses (Fig. 1) and estimated MOR fluxes of S and halogens to constrain the evolutionary history of the Earth's surface.

### [Analyses]

We analyzed glasses of MORBs collected at 2 sites on the East Pacific Rise, 2 sites on the Mid-Atlanric Ridge and 2 sites on the Central Indian Ridge. After volatiles in vesicles were extracted using the frozen crushing method [5], concentrations of <sup>3</sup>He were measured using a noble gas mass spectrometer (VG-5400) and those of S and halogens were measured using an ion chromatography (ICS-2100). Concentrations of S and halogens in solids were measured using a secondary ion mass spectrometer (NanoSIMS 50). We calculated relative molar ratios of S and halogens to <sup>3</sup>He (S/<sup>3</sup>He, F/<sup>3</sup>He and Cl/<sup>3</sup>He) and estimated MOR fluxes of S and halogens calibrating against the known <sup>3</sup>He flux (527 mol/yr [6]) [2, 3]. Using molar ratios, it is not necessary to discuss variations of volatile concentrations due to differences in porosities of each sample.

#### [Results and Discussion]

<sup>3</sup>He concentrations in vesicles were  $(1.8-6.3)x10^{-15}$  mol/g. Global averages of S/<sup>3</sup>He, F/<sup>3</sup>He and Cl/<sup>3</sup>He were calculated. For vesicle components, they are  $(4.2+-1.6)x10^7$ ,  $(1.4+-0.7)x10^6$  and  $(2.6+-1.0)x10^7$ , respectively. For bulk compositions, they are  $(0.3-1.2)x10^{10}$ ,  $(1.6-6.5)x10^9$  and  $(0.7-3.0)x10^9$ , respectively. Using these ratios and the known <sup>3</sup>He flux at MORs, MOR fluxes were estimated to be  $(2.2x10^{10}-6.6x10^{12})$  mol/yr for S,  $(7.1x10^8-3.4x10^{12})$  mol/yr for F and  $(1.4x10^{10}-1.6x10^{12})$  mol/yr for Cl. Mass balances were discussed comparing MOR fluxes in this study with arc fluxes, influxes at subduction zones and rates of accretion to the continental crust. When we assume the continuous degassing with the low MOR fluxes estimated using vesicle (hydrothermal fluid) compositions, the amount of S accumulated on the Earth's surface are calculated to be 50 times lower than its surface inventory, and halogens are calculated not to be accumulated on the Earth's surface because of dominant influxes. This implies a possibility that significant amounts of chemically reactive volatile elements such as S and halogens were accumulated on the Earth's surface in the early Earth.

#### [References]

[1] Hamano Y. and Ozima M. (1978) Terrestrial Rare Gases, 155-171, Japan Scientific Societies Press. [2] Marty B. and Jambon A. (1987) Earth Planet. Sci. Lett., 83, 16-26. [3] Sano Y. et al. (2001) Chem. Geol., 171, 263-271. [4] Tajika E. (1998) Geophys. Res. Lett., 25, 3991-3994. [5] Kagoshima T. et al. (2012) Geochem. J., 46, e21-e26. [6] Bianchi D. et al. (2010) Earth Planet. Sci. Lett., 297, 379-386.

Keywords: sulfur, halogen, helium, mid-ocean ridge basalt, flux, geochemical cycle

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(1) Bubbles connect each other and make paths in ascending magmas. Components fractionated into gaseous phases are released to the ocean.  $\Rightarrow MOR fluxes dominated by hydrothermal fluid$ (volcanic gas) components can be estimated based oncompositions of vesicles in MORB glasses.

(2) Erupted silicate melts are altered and bulk (c) Empire suite metris are interest of a metric of market components are released to the ocean. ⇒ MOR fluxes provided by intensive alteration processes of MORBs can be estimated based on bulk compositions of MORB glasses.