

Estimation of sulfur and halogen fluxes at mid-ocean ridges deconvolving contributions from vesicle and solid components of MORB glasses

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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) have been estimated based on compositions of glassy rims of MORBs (MOR Basalts) which retain those of magmas derived from the upper mantle. However, they may be overestimated, 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 and estimated MOR fluxes of S and halogens to constrain the evolutionary history of the Earth's surface. We analyzed glasses of MORBs collected at 2 sites on the East Pacific Rise, 2 sites on the Mid-Atlantic Ridge and 2 sites on the Central Indian Ridge. After volatiles in vesicles were extracted using the frozen crushing method, concentrations of helium-3 (³He) 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 ³He (S/³He, F/³He and Cl/³He) and estimated MOR fluxes of S and halogens calibrating against the known ³He flux (527 mol/yr). Using molar ratios, it is not necessary to discuss variations of volatile concentrations due to differences in porosities of each sample. ³He concentrations in vesicles were (1.8-6.3)×10⁻¹⁵ mol/g. Global averages of S/³He, F/³He and Cl/³He were calculated. For vesicle components, they are (4.2±1.6)×10⁷, (1.4±0.7)×10⁶ and (2.6±1.0)×10⁷, respectively. For bulk compositions, they are (0.3-1.2)×10¹⁰, (1.6-6.5)×10⁹ and (0.7-3.0)×10⁹, respectively. Using these ratios and the known ³He flux at MORs, MOR fluxes were estimated to be (2.2×10¹⁰-6.6×10¹²) mol/yr for S, (7.1×10⁸-3.4×10¹²) mol/yr for F and (1.4×10¹⁰-1.6×10¹²) 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 the most part of surface S and halogens were degassed in the early Earth.