

Geodynamic cycles of sulfur, carbon and nitrogen

KAGOSHIMA, Takanori^{1*}; SANO, Yuji¹

¹Atmosphere and Ocean Research Institute, University of Tokyo

Sulfur has important roles not only in biological activities but in industry and medicine. However, its geochemical cycle between the mantle and the surface environments has not been well quantified. ³He, one of the primordial noble gas isotopes, is useful to investigate to evaluate S cycles. ³He fluxes from the mantle to atmosphere and ocean are well constrained, which enables us to use this isotope to estimate other volatile fluxes. The recent study reported S and C fluxes from the mantle to the surface environments based on He, S, and C isotopic compositions in MORB, hydrothermal fluids, and volcanic gases [1]. The fluxes were estimated with the recently reported ³He flux at ridges, 530 mol/y [2]. The S/³He ratio at MOR was calculated to be 1.9x10⁸ as an average of S/³He ratios in six MORB vesicles (13N, 17S on EPR; 15N, 37N on MAR; 24S-25S on CIR) and 10 high temperature (>200 °C) hydrothermal fluids (11N-47N, 17S-19S on EPR; 23N-38N on MAR). Multiplying this ratio and the ³He flux together, S flux at ridges was estimated to be 100 Gmol/y. An average S/³He ratio of 15 high temperature (>200 °C) volcanic gases collected from circum-Pacific regions was calculated to be 6.5x10⁹, providing S flux of 720 Gmol/y from arc volcanoes calibrating against ³He flux of 110 mol/y determined by its MOR flux. This flux is higher than that from ridges. However, S in volcanic gases does not originate only from the mantle. S/³He ratios and δ³⁴S values in volcanic gases can be explained by mixing of three components: the upper mantle; subducted sedimentary pyrite; and subducted sulphate. The S contribution from the upper mantle was calculated to be 2.9% in volcanic gases, providing 21 Gmol/y from the mantle at arcs, which is lower than the S flux at ridges. Carbon flux at ridges was calculated to be 1200 Gmol/y using the CO₂/³He ratio of 2.2x10⁹ [3] based on compositions of MORB and hydrothermal fluids. An average CO₂/³He ratio of 24 high temperature (>200 °C) volcanic gases collected from circum-Pacific regions was calculated to be 2.0x10¹⁰, providing C flux of 2200 Gmol/y from arc volcanoes. CO₂/³He ratios and δ¹³C values in volcanic gases can be explained by mixing of three components: the upper mantle; subducted organic sediments; and subducted limestone with slab [4]. The C contribution from the upper mantle was calculated to be 11% in volcanic gases, providing 240 Gmol/y from the mantle at arcs. The S and C fluxes from the mantle to atmosphere and ocean are 121 Gmol/y and 1440 Gmol/y, respectively. The C/S flux ratio was calculated to be 12, which is comparable to the surface inventory ratio of 13 [5]. This suggests that the main source of surface S and C is the upper mantle. Assuming steady-state surface environments, subducted amounts of S and C become 820 Gmol/y and 3400 Gmol/y, respectively. Then 15% of subducted S and 42% of subducted C do not return to the surface environments and recycle back into the depth. Nitrogen cycles will also be quantified and discussed in the current study.

Reference: [1] Kagoshima et al. (2015) *Sci. Rep.* **5**, 8330. [2] Bianchi et al. (2010) *EPSL* **297**, 379-386. [3] Marty & Tolstikhin (1998) *Chem. Geol.* **145**, 233-248. [4] Sano & Marty (1995) *Chem. Geol.* **119**, 265-274. [5] Hilton et al. (2002) *RiMG* **47**, 319-370.

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