

Halogen analysis of mantle xenoliths by extending Ar-Ar and I-Xe dating methods

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An extension of the Ar-Ar and I-Xe dating methods enables us to determine trace amount of halogens (Cl, Br, and I), K, Ca, Ba, and U. These elements are converted to corresponding isotopes of Ar, Kr, and Xe by neutron irradiation in a nuclear reactor. Noble gases in the irradiated samples are analyzed by noble gas mass spectrometry and the conversion factors from parent elements to noble gas isotopes are determined by analyses of standard samples [1]. The detection limits of this technique for Cl, Br, and I are 10^{-10} to 10^{-11} , 10^{-13} to 10^{-14} , and 10^{-14} to 10^{-15} mol, respectively, which are several orders of magnitude lower than those of conventional methods. Using this method, we are investigating halogen geochemistry in the mantle.

Halogens, except for F, are incompatible elements that are strongly partitioned into aqueous fluids. They are concentrated at the Earth's surface in distinct compositions and scarce in the mantle. These features enable us to trace water subducted into the mantle using halogens. Halogens are also thought to play an important role in transporting LIL (large-ion lithophile) elements. Enrichment of LIL elements in arc magma is explained by addition of slab-derived aqueous fluids. These elements are up to 10 times more strongly partitioned into aqueous fluids when the fluids contain halogens [2]. Because of these geochemical characteristics, halogens are among the most important elements in slab-derived fluids. The fluid inclusions of mantle xenoliths provide the most primary information on slab-derived fluids supplied to the mantle wedge. However, the halogen concentrations in such materials are too low to analyze by conventional methods and thus little research has been conducted on them.

We determined the halogen concentrations in mantle xenoliths from various geological settings: Western-Pacific subduction zones (Kamchatka, the Philippines, and N. Japan), Europe (Eifel), and North America (San Carlos and Kilbourne Hole). The halogen compositions of fluid inclusions were obtained by crushing extraction of noble gases. Melting extraction was also conducted to obtain those of whole samples.

The H₂O-rich fluid inclusions of the mantle xenoliths from volcanic fronts of subduction zones show high I/Cl ratios with relatively constant Br/Cl ratios, which are similar to those of marine sedimentary pore fluids (seawater trapped in pores of deep-sea sediments) [3]. Halogen compositions obtained from melting of the same samples show a small addition of Cl; however, they are still similar to sedimentary pore fluids. Similar signatures were found in mantle peridotites from the Sanbagawa metamorphic belt, suggesting subduction of sedimentary pore-fluid-derived halogens thorough incorporation into hydrous minerals such as serpentine in the subducting oceanic lithosphere [4]. Although the halogen concentrations are lower, some mantle xenoliths from rear-arc regions of subduction zones also show sedimentary pore fluid-like signatures. The I/Cl and Br/Cl ratios of mantle xenoliths from intraplate settings (Europe and North America) show common correlation, suggesting halogen fractionation from MORB source-like composition.

The halogen signatures of mantle xenoliths from subduction zones suggest that subduction of sedimentary pore-fluid-derived halogens [4] significantly extend into the mantle beneath volcanic front. These subducted halogens also extend to rear-arc regions, although their influence is small. The correlated I/Cl-Br/Cl ratios of mantle xenoliths from intraplate settings may have resulted from a metasomatic event, which could generally take place in subcontinental lithospheric mantle.

[1] Turner (1965) JGR 70, 5433. [2] Kawamoto et al. (2014) EPS 66, 1. [3] Kobayashi et al. (2014) JpGU Meeting 2014, SCG62-04. [4] Sumino et al. (2010) EPSL 294, 163.

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