

## 論文の内容の要旨

### Ultrasensitive analysis of halogens in mantle xenoliths: Implications for volatile transportation into the mantle

(マントル捕獲岩の超高感度ハロゲン分析：揮発性物質の沈み込み過程の解明)

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Volatiles are transported from the Earth's surface into the mantle carried by subducting slabs. However, the detailed mechanism of volatile transportation into the mantle has been still enigmatic. Halogens are potentially powerful tracers of volatile transportation from the Earth's surface into the mantle because of (i) their high incompatibilities in mineral phases, (ii) the large contrast in concentrations between the surface and interior of the Earth, (iii) their distinct compositions in each reservoir. In order to gain a better understanding of volatile transportation mechanism into the mantle and its impact on the volatile compositions in the mantle, I analyzed halogens in mantle xenoliths, which are expected to provide direct evidence of volatile signatures in the mantle.

Studied mantle xenoliths are from various geological settings: volcanic fronts, rear-arc regions, and intraplate settings. Because mantle xenoliths contain only trace amounts of halogens, I applied an ultrasensitive analytical technique using neutron irradiation in a nuclear reactor and noble gas mass spectrometry. Most of the neutron irradiated samples were analyzed using an analytical system that I had improved. I also analyzed compositions of major and trace elements, and fluid inclusions.

Because of their geochemical characteristics, halogen/ $^{40}\text{Ar}^*$  ratios ( $^{40}\text{Ar}^*$  is excess  $^{40}\text{Ar}$  from air) are expected to reflect the extent of subduction influence. I found that the halogen/ $^{40}\text{Ar}^*$  ratios of the mantle xenoliths decrease toward the depleted mantle (DM) values in the order of volcanic fronts >

rear-arc regions > intraplate settings, indicating that the extent of subduction influence decreases in this order.

Halogen and noble gas elemental ratios obtained from the volcanic front samples are similar to those of sedimentary pore fluids and serpentinites. The consistency between halogen and noble gas signatures is strong evidence that the halogen and noble gas signatures derived from sedimentary pore fluid have been preserved during the subduction processes without significant fractionation. This indicates that subducted volatiles significantly extend into the mantle beneath volcanic front. On the other hand, Cl/H<sub>2</sub>O and <sup>36</sup>Ar/H<sub>2</sub>O ratios of the studied samples are higher than those in sedimentary pore fluids and serpentine. Based on the halogen-noble gas-H<sub>2</sub>O systematics, I propose subduction mechanism of sedimentary pore fluid-derived volatiles: (i) water derived from sedimentary pore fluid is incorporated into serpentine in a closed system formed along fracture zones developed at the outer rise, where oceanic plates bend prior to entering subduction zones, (ii) dehydration-hydration process within the oceanic lithospheric mantle maintains the closed system until the final stage of serpentine dehydration, which is a stepwise process accompanied with elemental fractionation of halogens, (iii) a portion of the fluids released at the final stage of serpentine dehydration hydrates the overlying slab whereas sedimentary pore fluid-like halogen and noble gas signatures are preserved in the remainder fluids due to highly channelized fluid flow.

The samples from rear-arc regions showed a wide range of Br/Cl and I/Cl ratios. All data can be explained by mixing among three components: sedimentary pore fluid-like high I/Cl halogens, high Br/Cl halogens, and DM-like halogens. Trace element patterns and Sr-Nd isotopic ratios are different among samples with different halogen signatures, indicating that these components derived from different sources. Whereas the high I/Cl and Br/Cl halogens are slab-derived, the DM-like halogens are expected to be those inherent in the mantle. This indicates that subducted halogens are not overwhelming in rear-arc regions, whereas they are overwhelming in the mantle beneath volcanic fronts

I/Cl and Br/Cl ratios obtained from the intraplate samples show two correlation trends extending from the DM-like values. Because arc-type signatures are not observed in trace element signatures, which are coupled with those of halogens, the influence of subducted halogens is considered not to extend into the SCLM (subcontinental lithospheric mantle) beneath the studied localities. Halogen and trace element signatures suggest that one trend would result from elemental fractionation during partial

melting and the other would be formed by mixing between DM-like halogens and melt-derived halogens. The geochemical signatures obtained from the intraplate samples suggest that the SCLM in the studied localities, which has been isolated from the mantle convection since about 1.6 Ga, had similar halogen signatures to those of the present-day DM before fractionation and mixing processes. This indicates that halogen signatures in the convecting mantle has been constant at least for 1.6 Gyrs.

I applied a box model to estimate the halogen subduction fluxes required to have maintained the I/Cl ratio in the mantle in a global scale over the past 1.6 Gyr. The box model calculation indicates (i) that the I/Cl ratio in the mantle is sensitive to their subduction fluxes and mechanism, (ii) halogen subduction fluxes estimated from chemical compositions of subducting materials are too high to have maintained the I/Cl ratio in the mantle over the past 1.6 Gyr, (iii) halogens are subducted into greater depth than noble gases, which have been thought to be expelled from subducting slabs at upper mantle depths due to their high volatilities, and not incorporated into the mantle convection.