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Deciphering material cycling in subduction zones through molybdenum isotopic composition of volcanic rocks from the Japan arc

（日本列島火山岩のモリブデン同位体組成から探る沈み込み帯の物質循環）

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Abstract

Since the establishment of plate tectonics, subduction zones have been recognized as the place where oceanic plates play a crucial role in the cycling of elements from the Earth's surface to the deep mantle. To elucidate the global-scale elemental cycling, a large number of geochemical studies have been conducted, analyzing major and trace elements in numerous types of rock samples, as well as isotopic compositions, including radiogenic isotopes such as Sr, Nd, Hf, and Pb, and noble gases. Among these, the isotopic composition of molybdenum ($\delta^{98/95}Mo = [(^{98}Mo/^{95}Mo_{sample})/ (^{98}Mo/^{95}Mo_{NIST\ SRM\ 3134}) - 1] \times 1000$) has received considerable attention in geochemistry as a tracer of global-scale material cycles. However, significant uncertainties remain in the understanding of geochemical variations across volcanic arcs, particularly in rear-arc volcanoes, where active phase transitions in subducted slabs and fluid migration are prevalent. The geochemical variations observed across the arc from the volcanic front (VF) to the rear arc (RA) provide valuable insights into the continuous phase transitions associated with changes in the depth of the subducting slab, thereby advancing our understanding of the processes governing subduction-zone magmatism and material cycling. In this thesis, we analyzed major and trace element abundances as well as Mo-Sr-Nd-Pb isotopic compositions of 17 volcanoes near the Japanese Islands to reassess the relationship between across-arc variations in Mo isotopic composition and the fundamental processes governing the subduction zones.

In Chapter 2, we investigated changes in the amounts of aqueous fluids and $\delta^{98/95}Mo$ values from the VF to the RA in the northern Izu (N-Izu) Islands (Izu-Oshima, Niijima, Kozushima). The N-Izu basalt shows a gradual decrease in the contribution of aqueous fluids from the subducting slab, as indicated by systematic changes in some trace

elements ratios (e.g., Ba/Th and Ce/Mo) with increasing depth in the Wadati-Benioff zone. Similarly, the $\delta^{98/95}$Mo values of the N-Izu basalts decrease with increasing depth in the slab. The simplest interpretation of the increase in $\delta^{98/95}$Mo values in the VF is that aqueous fluids derived from slabs rich in heavy Mo isotopes contributed to the mantle source of the VF volcano. In contrast, the $\delta^{98/95}$Mo values of the RA basalts are as low as the estimated depleted mantle (DM), suggesting that the slab-derived fluids in the RA region are depleted in heavy Mo isotopes as a result of continuous loss of heavy Mo isotopes due to dehydration of the VF volcano.

In Chapter 3, we analyzed the across-arc variation of $\delta^{98/95}$Mo values in eight volcanoes from the Northeast Japan (NE Japan) region, together with the isotopic compositions of Sr, Nd, and Pb. The mean $\delta^{98/95}$Mo values of the NE Japan VF samples are higher than that of the DM, while the $\delta^{98/95}$Mo variation in the NE Japan VF samples is greater than that of other oceanic island arcs. The $\delta^{98/95}$Mo values of the VF samples show no clear correlation with crustal assimilation or magma differentiation, but are highly variable. The observed variation is most likely reflects the diversity of $\delta^{98/95}$Mo values in the primary magma associated with the heterogeneity of $\delta^{98/95}$Mo values in the slab-derived fluids. On the other hand, the RA region exhibits two distinct systems based on $\delta^{98/95}$Mo values and radiogenic Pb isotopes. Chokai and Oshima-Oshima volcanoes have lower $\delta^{98/95}$Mo values, while Kampu and Sannome-gata show higher values. The former show typical across-arc geochemical variations similar to VF, while the high $\delta^{98/95}$Mo values of the latter may be due to the addition of residual crust and recycled materials due to the expansion of Japan Sea, or the variations in the input of slabs along the arc.

In Chapter 4, we analyze the $\delta^{98/95}$Mo values in volcanic rocks from three ultra-

rear arc (URA) regions, Oki-Dogo, Fukue Island, and Changbaishan, to investigate the influence of recycled slab components, deep interior dehydration, and slab melts on the generation of URA magmas. The $\delta^{98/95}$Mo values are consistent with those of ocean island basalts (OIB), continental basalts, and RA volcanic rocks from the Izu and Mariana arcs, indicating a minimal contribution of slab-derived fluid components. Oki-Dogo and Changbaishan exhibit EM-1-like $\delta^{98/95}$Mo values, while Fukue shows EM-2-like values, reflecting heterogeneity of the mantle source influenced by different recycled materials.

Finally, in Chapter 5, we integrated all the results from the analyzed volcanic rocks to explore the relationship between phase transitions in volcanic rocks within subduction zones and the subducted slab. The average $\delta^{98/95}$Mo values for VF and RA samples are determined to be -0.07 ± 0.44‰ and -0.19 ± 0.33‰, respectively, showing a general across-arc trend of decreasing $\delta^{98/95}$Mo values from VF to RA in subduction zone volcanoes. Furthermore, the variation in $\delta^{98/95}$Mo values increases with decreasing Ba/Th ratios, suggesting a possible classification into two groups according to the contribution of sediments derived from the subducting plate. We also re-evaluated the overall circulation and variation of $\delta^{98/95}$Mo values within subduction processes. In previous studies, the origin of slab-derive fluids was considered to be the hydrous phase in the subducting slab, erosion of serpentinized fore-arc mantle, and serpentinized lithospheric mantle in subducting slab. However, the presence of serpentinized fore-arc mantle is not universal, and the contribution of fluids from the serpentinized slab lithospheric mantle to the mantle wedge would be geophysically difficult to accept. Instead, we developed two alternative models for fluid origin in subduction zones with and without serpentinized fore-arc mantle and proposed a new model involving continuous phase transitions in the subducting oceanic crust.