Potassium isotope fractionation during silicate-carbonatite melt immiscibility and phlogopite fractional crystallization

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Abstract
Potassium (K) isotopes have been used as a tracer of K recycling in the Earth, but K isotope fractionation during magma evolution is poorly constrained. Here, we present K isotope data for a magmatic suite of alkaline silicate-carbonatite affinity. The suite was formed from liquid-liquid immiscibility and subsequent phlogopite fractionation. The K isotopic signatures of different rock types are in the following order: alkaline silicate lavas (Δ41K = –0.424 to 0.090‰) > carbonatitic silicate lavas (Δ41K = –0.640 to –0.035‰) > carbonatites (Δ41K = –0.858 and –0.258‰). Phlogopite phenocrysts in the silicate lavas are isotopically lighter (Δ41K = –0.628 to –0.534‰) than the lavas in which they occur (Δ41Kphlogopite-whole rock = –0.502 to –0.109‰). Correlations between Δ41K values and chemical proxies of melt immiscibility and phlogopite fractionation indicate that K isotopes are significantly fractionated by both processes at a ~0.6‰ magnitude. Such K isotope variation overlaps the range of Δ41K in arc lavas. Compilations of literature data further confirm the critical roles of melt immiscibility and phlogopite fractionation in K isotope variations of high-K lavas (K2O >1 wt%) from post-collision orogenic and intra-continental settings. In comparison, basaltic arc lavas are depleted in K2O (mostly <1 wt%) and lack evidence of significant phlogopite fractionation. The K isotope variations of arc lavas are mainly controlled by their mantle sources, which were metasomatized by melt or fluid releases from the subducting slab. Therefore, K recycling and K isotope variation are controlled by distinct mechanisms in different tectonic settings.

Keywords: K isotopes, silicate rock, carbonatite, melt immiscibility, phlogopite, fractional crystallization

Introduction
Potassium (K) displays a large contrast between Earth’s crust and mantle in concentration due to its high incompatibility during crust-mantle differentiation (Mittlefehldt 1998; Lyubetskaya and Korenaga 2007; Palme and O’Neill 2007) and is thus a powerful tool to trace crustal recycling and crust-mantle interaction. It is also a fluid-mobile element, and its heavy isotopes are, in most cases, preferentially partitioned into fluids (Mittlefehldt 1998; Li et al. 2017; Tuller-Ross et al. 2019; Hu et al. 2020; Wang et al. 2021a). Dehydration of the subducting slab releases isotopically heavy melts and/or fluids to elevate Δ41K, as shown by arc lavas (Liu et al. 2020; Hu et al. 2021a; Parendo et al. 2022). It has been proposed that further melting of dehydrated slabs is taken to generate magmas showing light K isotopic compositions in tectonic settings far away from the subduction trench and arc (Parendo et al. 2022). However, recent studies have revealed that mantle-derived, potassic-ultrapotassic rocks in post-collisional and intra-continental environments mostly do not show light K isotope compositions compared to arc lavas (Sun et al. 2020; Wang et al. 2021b; Hu et al. 2021a; Liu et al. 2021; Parendo et al. 2022). While heavy K isotopic compositions are generally ascribed to hydrous fluids from subducting slabs, the probable isotope fractionation during magma ascent is widely assumed to be negligible (e.g., Tuller-Ross et al. 2019; Parendo et al. 2022). However, mantle-derived potassic-ultrapotassic rocks commonly undergo fractional crystallization of K-rich minerals such as phlogopite and leucite, which could result in compositionally different isotopic trends compared to other systems dominated by K-poor minerals. Thus, clarifying K isotope fractionation during magma differentiation is essential in using K and its isotopes as a robust geochemical tracer.

Primary carbonatite melts are thought to be alkaline in composition (Chen et al. 2013), but the partitioning and evolution of alkali elements in those melts might be complex due to the combined effects of melt immiscibility and fractional crystallization. Mantle-derived alkaline silicate lavas are also commonly rich in K and, in some cases, associated with carbonatites (Stoppa and Schiazza 2013). Although many alkaline silicate rock and carbonatite occurrences on the Earth formed...