

## A fresh look at the mantle sources of low-degree mantle-derived melts using K and Ba isotopes

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### Introduction

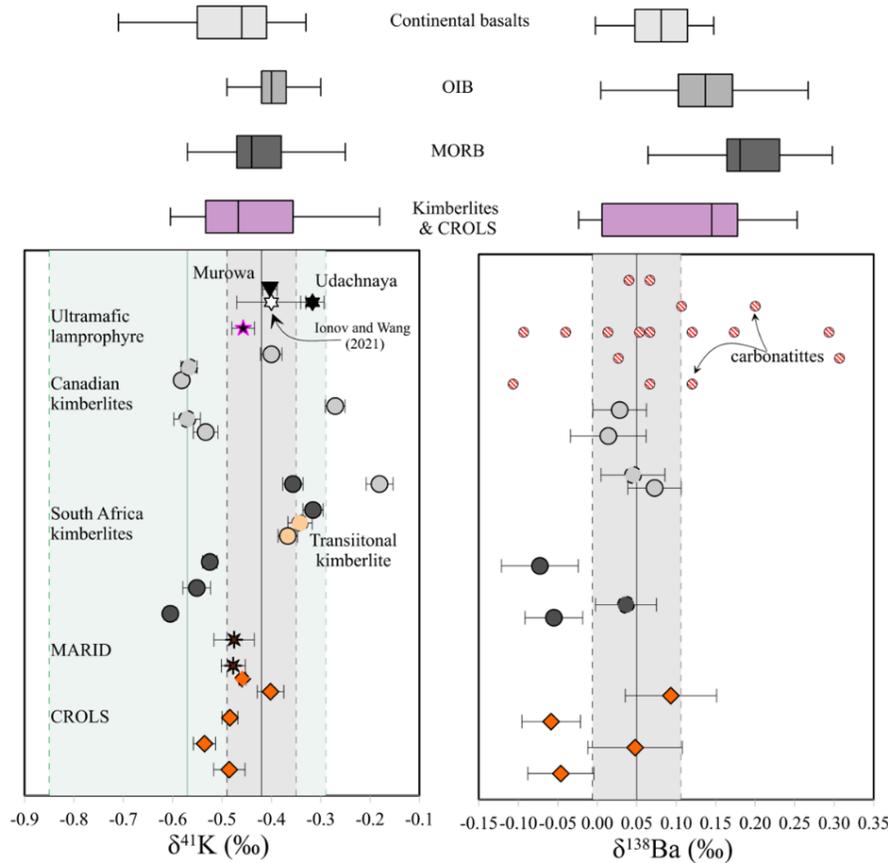
Mantle-derived, low-degree melts, such as kimberlites, carbonate-rich olivine lamproites (CROLS), and cratonic olivine lamproites, are the main carriers of diamonds. They are rare ultramafic, volatile-rich volcanic magmas, generally restricted to stable cratons, and are the deepest-sourced magmas erupted onto Earth's surface. As hybrid magmas, their formation mechanism and mantle sources remain enigmatic and highly debated, especially the nature of the processes leading to their "enriched" isotopic signatures. The often extreme isotopic compositions of Sr, Nd, Pb, and Hf suggest that the mantle sources of these magmas vary between an ancient and geochemically depleted component and various enriched components. The enriched components could include crustal material recycled into the convective mantle or metasomatized lithospheric mantle. For the latter, discriminating between assimilation by sub-lithospheric magmas during the ascent or melting of element-enriched material from within the lithospheric mantle is paramount in resolving their petrogenesis.

Radiogenic isotope tracers have the complexity of time-integrated evolution and hence can be difficult to unequivocally resolve. For instance, whether the "enriched" Sr-Nd-Hf isotope signatures in cratonic lamproites are a simple mixture of sediment in the mantle source or the result of ancient time-integrated parent-daughter isotope fractionation cannot be totally resolved using those isotopes alone. As the stable isotope compositions of K and Ba differ between surface and mantle reservoirs, they are tools well-suited to addressing the cause of different radiogenic isotopic signatures and can be used better to constrain the mantle sources of these important magmas. Here, we use collision cell multi-collector inductively-coupled-plasma mass-spectrometry (MC-ICP-MS) and traditional MC-ICP-MS to conduct the first comprehensive whole-rock K and Ba stable isotope study on a wide range of low-degree mantle-derived melts.

### Results

A set of 31 low-degree mantle-derived melts and xenoliths (14 archetypal kimberlites, one transitional kimberlite, one UML, 4 CROLS, 13 cratonic lamproites, and two MARID xenoliths) from a wide range of geographic locations, eruption times and geochemical characteristics were selected for this study. Overall, kimberlites show limited  $\delta^{41}\text{K}$  and  $\delta^{138}\text{Ba}$  variability, with a median  $\delta^{41}\text{K}$  of  $-0.40 \pm 0.06\%$  (2SD) and  $\delta^{138}\text{Ba}$  of  $0.00 \pm 0.07\%$  (2SD), similar to MORB and OIBs and within error relative to an estimated bulk silicate Earth [(BSE:  $\delta^{41}\text{K} = -0.42 \pm 0.07\%$  (2SD) and  $\delta^{138}\text{Ba} = 0.03 \pm 0.04\%$  (2SD)], suggesting

significant convecting mantle input (Fig. 1). Given that the K isotopes show no apparent systematic change relative to the BSE and lithospheric reservoirs throughout the last ~550 Ma, no clear sign of recycled crustal material is observed. A primitive mantle reservoir for kimberlites is consistent with recent  $\delta^{44}\text{Ca}$  measurements and modeling conducted on a range of archetypal kimberlites from the South African Cretaceous Kimberley cluster (Antonelli et al. 2023).

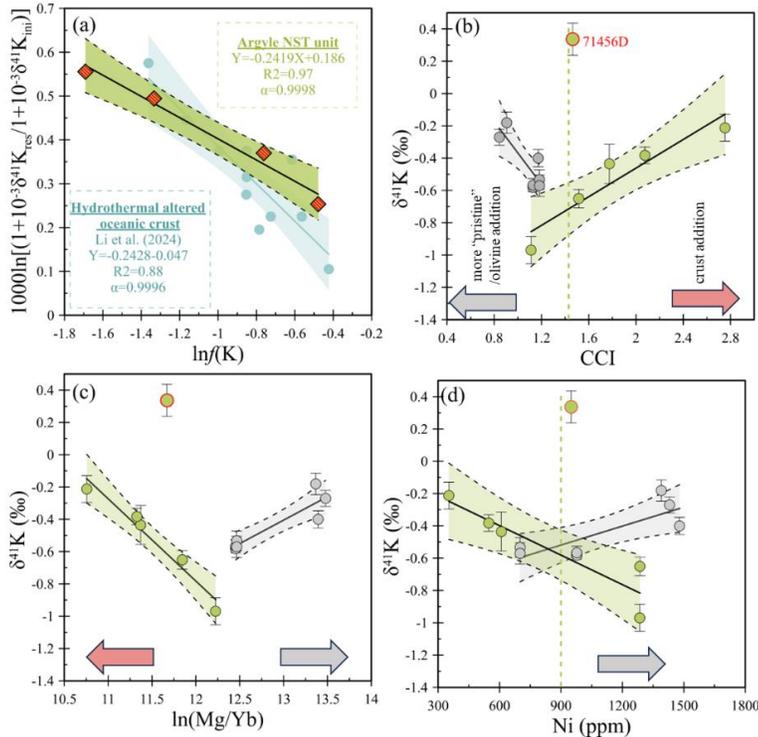


**Figure 1.** K and Ba isotopic compositions of kimberlites and CROLS ultramafic lamprophyre and other mantle-derived melts. Also shown are 'box and whiskers' diagrams of  $\delta^{41}\text{K}$  and  $\delta^{138}\text{Ba}$  for this study and the literature. MORB (mid-ocean ridge basalt), OIB (ocean island basalt), and continental basalts are also shown. The 'whiskers' are the  $1.5 \times$  interquartile range, while the boxes represent the median, 25<sup>th</sup>, and 75<sup>th</sup> percentile. Data sources for K isotopes are from the compilation of Wang et al. (2021) and references therein. For  $\delta^{138}\text{Ba}$  data, MORB (Nielsen et al. 2018; Wu et al. 2023), OIB (Bai et al. 2022), continental basalts (Zhao et al. 2021), and carbonatites are from (Li et al. 2020). Grey-shaded areas for the  $\delta^{41}\text{K}$  and  $\delta^{138}\text{Ba}$  are BSE estimations from basalts. The green shaded area is the fertile and lightly metasomatized peridotites from Ionov and Wang (2021).

Like kimberlites, South African CROLS also show limited K isotope variability with a median  $\delta^{41}\text{K}$  of  $-0.48 \pm 0.02\text{‰}$  (2SD). Their compositions are non-resolvable from two mica-amphibole-rutile-ilmenite-diopside (MARID) xenoliths, consistent with their well-known relationship. The  $\delta^{138}\text{Ba}$  for CROLS also shows limited variation, with a median  $\delta^{138}\text{Ba}$  of  $0.00 \pm 0.07\text{‰}$  (2SD). Similar to the case recorded in  $\epsilon^{205}\text{Tl}$  (Fitzpayne et al. 2020) and  $\delta^7\text{Li}$  (Tappe et al. 2023), the BSE like  $\delta^{41}\text{K}$  and  $\delta^{138}\text{Ba}$  of CROLS and MARID diverge from sediment-derived melts and subduction zone magmas and xenoliths, which share 'enriched' Sr-Nd-Hf-Pb isotope signatures. This suggests a major contribution from the convecting mantle with minimal sediment recycling in the source, consistent with Nd-Hf-Sr isotope modeling (Tappe et al., 2023).

Compared to the other low-degree mantle-derived melts, cratonic olivine/leucite-bearing lamproites from the Argyle and west Kimberly fields in Western Australia show a wide range in  $\delta^{41}\text{K}$  ( $-0.97\text{‰}$  to  $+0.34\text{‰}$ ) and  $\delta^{138}\text{Ba}$  ( $-0.30\text{‰}$  to  $+0.27\text{‰}$ ) values. The Argyle lamproites define robust correlations between potassium and barium elemental abundances, and their diverse stable isotopes call for significant hydrothermal fluid-assisted leaching and isotopic fractionation after emplacement. Assuming constant equilibrium fractionation between the leaching fluid and lamproite, we calculated the fractionation factor  $\alpha$  [defined as  $\alpha = \frac{(^{41}\text{K}/^{39}\text{K})_{\text{fluid}}}{(^{41}\text{K}/^{39}\text{K})_{\text{lamproite}}}$  and  $\frac{(^{138}\text{Ba}/^{136}\text{Ba})_{\text{fluid}}}{(^{138}\text{Ba}/^{136}\text{Ba})_{\text{lamproite}}}$ ] to be  $\alpha = 0.9998$  for K (Fig. 2a) and  $0.9997$  for Ba (not shown) isotope systems. West Kimberly lamproites do not form linear

trends against K and Ba but are characterized by direct correlations between and  $\delta^{41}\text{K}$  crustal/mantle indexes, possibly from the interaction between sublithospheric melt and wall rock SCLM (Fig. 2b-d).



**Figure 2:** K isotope systematics in cratonic lamproites. (a) Effects of hydrothermal leaching in Argyle. Relationship between  $\ln f(\text{K})$  vs.  $1000\ln[(1 + 10^{-3}\delta^{41}\text{K}_{\text{res}})/(1 + 10^{-3}\delta^{41}\text{K}_{\text{ini}})]$  for the Argyle samples. For comparison, trends for hydrothermal leaching in altered oceanic crust (AOC) from the east Pacific rise (blue circles; Li et al., 2024). The slope in the data relates to  $1000(1 - \alpha K)$ , where  $\alpha K$  is the kinetic isotope fractionation between K in the leaching fluid and the residual K in the lamproite. (b-d) linear correlations between  $\delta^{41}\text{K}$  and crustal and SCLM contamination indexes. Regression lines with 95% confidence intervals (shaded areas) are shown. Green circles are lamproites from the west Kimberley field. Sample 71456D is considered an outlier. Grey symbols are for SCLM and crustal-contaminated kimberlites from Canada. Arrows show the proposed general effects of crustal and SCLM contamination. The Green dashed line is the estimation of primary olivine lamproite are from (Jaques and Foley 2018)

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