



Tracking continental-scale modification of the Earth's mantle using zircon megacrysts

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Introduction

Metasomatism is an important process generating regions of mantle enriched in volatile and incompatible elements that may subsequently melt, giving rise to a range of magma types. The spatial extent of metasomatic processes is poorly understood because geographically extensive studies of relevant metasomatic minerals with known ages are rare. Zircon megacrysts, an uncommon, large (cm-sized) and somewhat unusual mineral occurrence, recovered during the processing of kimberlites to extract diamonds, may fill this gap. Their trace element patterns (e.g., Belousova *et al.*, 2002) and low $\delta^{18}\text{O}$ (Page *et al.*, 2007) indicate that they are not of crustal origin, but crystallised within the mantle and experienced only minimal chemical interaction with the host magmas that transported them to the surface. While details of their petrogenesis (and the origin of megacryst suites more broadly) remain a subject of active research, there is agreement that zircon megacrysts are produced by metasomatic melts in some way related to kimberlite magmas (e.g., Kinny *et al.*, 1989; Nowell *et al.*, 2004; Page *et al.*, 2004). They record precise U-Pb ages and initial $^{176}\text{Hf}/^{177}\text{Hf}$ isotope ratios providing important constraints on the age and nature of the metasomatic events occurring in their mantle sources. We present the first geographically-extensive survey of Hf-isotope and U-Pb age distributions for zircon megacrysts in southern African kimberlites, representing widely spaced intrusions spanning both cratonic (Kaalvaal, Zimbabwe) and non-cratonic settings.

Results and Discussion

Zircons have very low Lu/Hf ratios and thus preserve the initial $^{176}\text{Hf}/^{177}\text{Hf}$ of their source metasomatic melts. Our results reveal an entirely unexpected first order observation; that is, remarkable large-scale isotopic homogeneity among southern African zircon megacrysts across lithospheric domains with widely differing ages. Although a restricted isotopic range in Hf-isotopes has been noted previously in a much smaller dataset of kimberlite megacrysts from this area (Griffin *et al.*, 2000), our analyses show near identical isotopic compositions in samples derived from numerous intrusions distributed across a region of >1 million km².

The data form two homogeneous yet distinct compositional groups, which we term A and B (Fig. 1a); a distinction also mirrored in Nd-isotope data. Some kimberlite pipes contain both zircon groups (e.g., Wesselton, Koffiefontein), as previously reported for the Orapa and Jwaneng kimberlites (Kinny *et al.*, 1989, Griffin *et al.*, 2000). Remarkably, the subtle variations in $^{176}\text{Hf}/^{177}\text{Hf}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ in zircons of the larger Group A correlate with age and may reflect radiogenic ingrowth in the source of the metasomatic zircon parent melts (Fig 1b,c). Although the $^{176}\text{Hf}/^{177}\text{Hf}$ – age correlation is largely defined by the off-craton samples that show the greatest range of ages, it remains true that the cluster of on-craton samples also lies along this array. All results from this study plot below the Nd-Hf isotope mantle array.

Group A zircons yield precise and concordant U-Pb ages which generally approximate the (usually less precise) age estimates of their kimberlite hosts. In contrast, U-Pb systematics for Group B zircons are disturbed, precluding accurate dating, and suggesting a more protracted history.

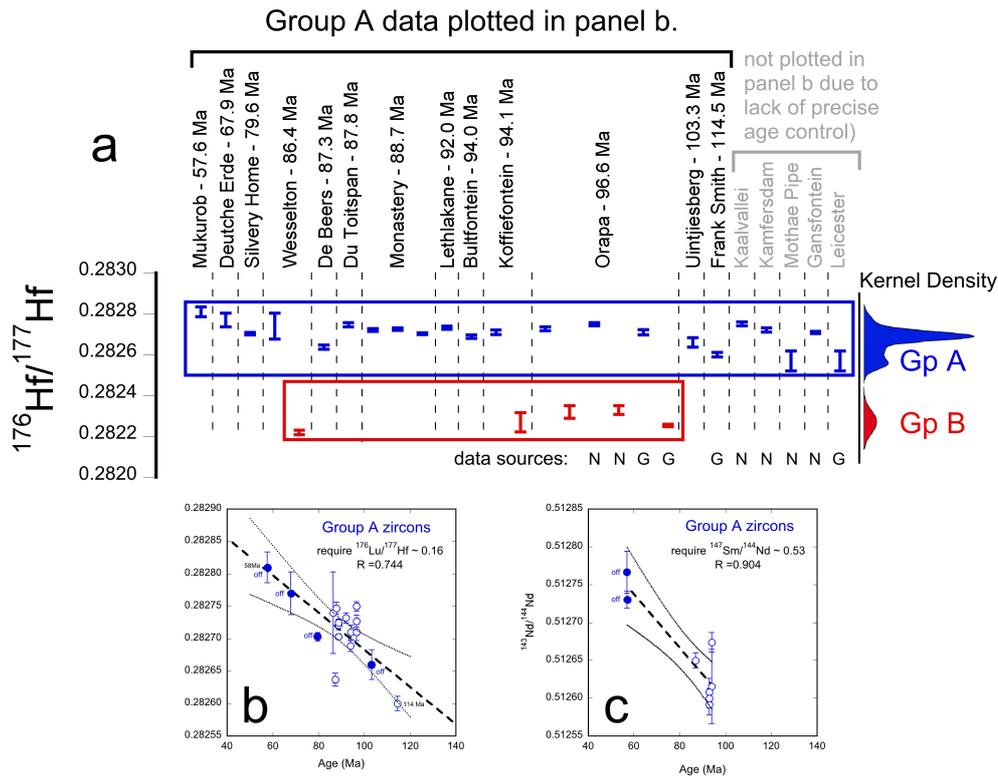


Figure 1 Age and isotopic composition data for zircon megacrysts.

(a) Zircon Hf-isotope data showing a natural compositional subdivision into two distinct groups, further illustrated by Kernel Density estimates. Note the remarkable isotopic homogeneity within each group despite large variations in geographic location and age. All data from this study unless marked: N = Nowell *et al.* (2004), G = Griffin *et al.* (2000).

(b) Inset showing a statistically significant correlation between zircon $^{176}\text{Hf}/^{177}\text{Hf}$ composition and age. Only zircons for which precise U-Pb Concordia ages are available are used to construct this plot. Literature data with less precise age determinations (greyed out in 1a) are excluded.

(c) An equivalent plot to 1b for $^{143}\text{Nd}/^{144}\text{Nd}$ isotope variations.

Our results provide a consistent picture of megacryst parental melts which tapped an isotopically homogeneous source extending over hundreds of kilometres, and encompassing a time interval of nearly 70 Myr, the range of U-Pb ages (114-56 Ma) recorded by the zircons. The apparent $^{176}\text{Hf}/^{177}\text{Hf}$ – age relationship defined by the Group A zircons places important constraints on the nature and evolution of their mantle source(s). To produce such a correlation these source rocks must have been relatively homogeneous initially and subsequently evolved rapidly with a strongly super-chondritic $^{176}\text{Lu}/^{177}\text{Hf}$ ratio (~ 0.16 , Fig. 1b). The initial $^{143}\text{Nd}/^{144}\text{Nd}$ values for Group A zircon megacrysts also correlate with age (Fig. 1c), consistent with a source that evolved with a moderate-high $^{147}\text{Sm}/^{144}\text{Nd}$ ratio of ~ 0.53 (although both parent-daughter ratios are poorly defined, based on the paucity of the data). Importantly, prior to rapid radiogenic ingrowth, the initial source rock composition must have been located off the mantle-array, displaced to lower $^{176}\text{Hf}/^{177}\text{Hf}$ for a given $^{143}\text{Nd}/^{144}\text{Nd}$. This also provides important insights into both the nature of the original mantle source rocks and the metasomatic fluid that modified them.

We postulate that the mantle source rocks originally had a protracted history of unusually low Lu/Hf and Sm/Nd and developed initial $^{176}\text{Hf}/^{177}\text{Hf}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ that are low relative to MORB mantle (i.e., ‘enriched mantle’). Subsequent metasomatism of these source rocks not only drastically raised Lu/Hf to drive rapid ^{176}Hf ingrowth for at least ~ 70 Ma (Fig. 1b) but must also have had a) low Hf contents to preserve the original unradiogenic $^{176}\text{Hf}/^{177}\text{Hf}$ signature of the protolith but b) sufficient Nd

to modify the $^{143}\text{Nd}/^{144}\text{Nd}$ to values more typical of OIB. Metasomatism therefore decoupled Hf from Nd (and presumably Sr) isotope compositions to generate source rocks, and ultimately zircon megacrysts, with compositions to the right of the Nd-Hf mantle array.

The occurrence of near-homogeneous $^{176}\text{Hf}/^{177}\text{Hf}$ in megacryst zircons across two cratons (Kaapvaal and Zimbabwe) and the surrounding Proterozoic requires the inferred metasomatic processes to postdate final tectonic assembly of these crustal domains. This suggests the source of Group A zircons postdates the ~1300 Ma amalgamation of the Kaapvaal craton and the Namaqua-Natal belt (Eglington, 2006), the youngest terrane with Cretaceous kimberlites; a younger limit is provided by the age of the oldest host kimberlite, the 114 Ma Frank Smith pipe. Importantly, the rapid isotopic evolution of the modified mantle source required by the zircon data, make it unlikely that the metasomatic event occurred more than a few hundred million years ago.

Concluding remarks

Our new Hf-isotope data provide clear evidence for a discrete metasomatic event in the southern Africa mantle operating at a continent-wide scale between 114 Ma and several hundred million years ago, and subsequently sampled by separate kimberlite eruptions over a period of at least 70 Ma. The possibility of a link between such large-scale mantle metasomatism and formation of the Karoo large igneous province has previously been suggested (Konzett *et al.*, 1998; Ernst and Bell 2010), and would be consistent with the very large thermal and magmatic perturbation resulting from Karoo activity. New geochronological data for metasomatised mantle xenoliths from the Kimberley kimberlites also suggest a direct association of these events (Giuliani *et al.*, 2014). A link between widespread Karoo magmatism, modification of the southern African continental mantle, initiation of kimberlite magmatism, and megacryst formation therefore appears an intriguing possibility worthy of further study. A more disturbed and less sampled suite of zircon megacrysts supports the occurrence of a similar but older event.

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