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Sources and ages of diamond-forming fluids in the lithospheric mantle

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Introduction

High-density fluid (HDF) microinclusions in diamonds carry chemical fingerprints that provide the opportunity to directly examine the nature of carbon- and water-rich media in the deep Earth. These HDFs vary in composition between four end-member types globally (Weiss et al., 2022): hydrous-silicic, rich in Si, Al, K and H_2O; low-Mg carbonatitic and high-Mg carbonatitic, both rich in Ca, Mg, Fe, K and CO_2; and hydrous-saline, rich in Cl, K, Na and H_2O. Radiogenic isotope compositions, which preserve their signature during mantle processes such as melting and immiscible separation, are an important tool in tracing the potential mantle sources of the HDFs. Ideally, such isotope data can be used to constrain the timing and the varying origins of HDFs in the context of large-scale tectonic processes.

Available HDFs Sr isotope data of diamonds from Canada, Siberia and Africa vary between 0.703 to 0.723, indicating sources ranging from 'depleted' oceanic mantle to old continental lithosphere (e.g. Klein-BenDavid et al., 2014; Smith et al., 2012; Weiss et al., 2015). While the limited available Sr-Nd-Pb isotope compositions from a handful of diamonds from Canada indicate the contribution of both relatively primitive (ε_{Nd} of −0.2, $^{87}Sr/^{86}Sr$ of 0.7044, and $^{206}Pb/^{204}Pb$ of 17.52) and more trace element enriched ($\varepsilon_{Nd} < -16$, $^{87}Sr/^{86}Sr > 0.713$, and $^{206}Pb/^{204}Pb$ of 18.3) sources within the continental lithosphere (Weiss et al., 2023). Here, we combine major, trace element and isotope compositions of HDFs from the southwestern Kaapvaal Craton, South Africa, to constrain their petrogenesis and investigate possible sources and ages.

Isotopic composition of microinclusion-bearing southwestern African diamonds

We report the Sr-Nd-Pb isotope compositions of 18 microinclusion-bearing diamonds from the neighboring De Beers Pool, Finsch and Koffiefontein mines (Figure 1). Their $^{87}Sr/^{86}Sr$ vary from 0.7063 to 0.7587, $^{143}Nd/^{144}Nd$ from 0.5103 to 0.5130 (ε_{Nd} = –44.7 to +6.6, respectively), and Pb isotopes vary from 15.66 to 19.03 for $^{206}Pb/^{204}Pb$, 15.01 to 15.75 for $^{207}Pb/^{204}Pb$, and 37.97 to 38.75 for $^{208}Pb/^{204}Pb$. These variations expand the available data to more unradiogenic Nd and radiogenic Sr and Pb isotopes, and overlap a large proportion of the isotopic spectrum of the sub-continental lithospheric mantle (SCLM), as recorded by whole rock xenolith data from cratons, and subducted sediments. In addition, the calculated Nd model ages (T_{DM}) of the analyzed samples range between 0.2 and 3 Ga.

Combining the Sr-Nd-Pb isotope compositions with previously published data for the analyzed diamonds reveals systematic variations. For example, saline HDFs T_{DM} ages vary between 0.2 to 2 Ga with an average of 0.9 ±0.5 Ga, compared to carbonatitic and silicic HDFs, which record older ages between 1.6 and 3 Ga with an average of 2.4 ±0.5 Ga (Figure 2). These differences are associated with distinct SCLM-like $^3He/^4He$ ratios of 2.7–4.4 Ra (where Ra is the atmospheric ratio of 1.39×10^{-6}) of the saline HDFs and low-aggregated nitrogen impurities (only in A centers) of their host diamonds, compared to much lower radiogenic $^3He/^4He$ ratios of 0.07–0.69 Ra of carbonatitic and silicic HDFs in diamonds with aggregated nitrogen in both A and B centers (with 25–35% B centers; Weiss et al., 2021). All parameters have a strong temporal control indicating episodic formation events and variable mantle residence times.

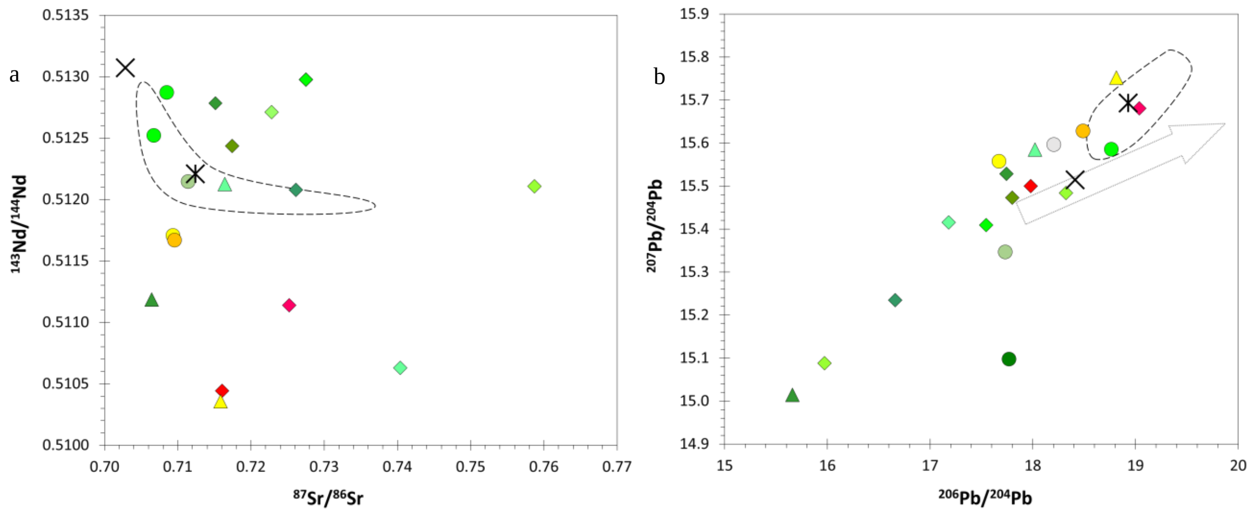

Figure 1: Sr-Nd-Pb isotope compositions of HDF from the southwestern Kaapvaal Craton, South Africa. (a) ^{143}Nd/^{144}Nd *vs.* ^{87}Sr/^{86}Sr and (b) ^{207}Pb/^{204}Pb *vs.* ^{206}Pb/^{204}Pb. Mine locality and HDF type are distinguished by data point shape and color, respectively: De Beers Pool – diamonds symbol, Koffiefontein – circles, Finsch – triangles; saline HDFs – green symbol, carbonatitic HDFs – yellow/orange symbol, silicic HDFs – red symbol (one Koffiefontien diamond not yet analyzed for HDF type – grey circle). For comparison, the range and average of subducting sediments are shown (dashed area and asterisk, respectively; Plank, 2014), as well as the average composition of mid-ocean ridge basalts (x symbol, MORB; Gale et al., 2013) and the direction of Pb mantle array (dotted arrow).

Temporal variation and source contributions to diamond-forming fluids

The He isotope signature of the saline HDFs and their host diamonds' unaggregated nitrogen (Figure 2) indicate limited mantle residence and probable formation during the Cretaceous, close in time to kimberlite eruption (Weiss et al., 2021). Moreover, their initial ^{3}He/^{4}He value of 4.6 ± 0.7 Ra strengthen the connection between these young-age saline HDFs and recycled subducted surface material (comparable to 'low-^{3}He/^{4}He' OIBs of young ages). However, their Sr-Nd-Pb isotope compositions reveal the complex involvement of several distinct sources of different age.

The ^{87}Sr/^{86}Sr *vs.* ^{87}Rb/^{86}Sr trend of the saline HDF inclusions may be interpreted as an age that corresponds to 1194 ± 291 Ma. This age could reflect a connection with the Laurentia-Kalahari collision and subduction event at ~1150–950 Ma, however, mixing between two sources is indicated by a ^{87}Sr/^{86}Sr *vs.* 1/Sr relationship. Considering the Nd isotope variation of these HDFs, we interpret these two sources as an Archaean LREE enriched lithospheric mantle (i.e. low ^{143}Nd/^{144}Nd), and a less enriched component with more radiogenic Nd and unradiogenic Sr signature that approaches MORB values (Figure 1a). An important observation, however, is that the Pb isotope ratios of most saline HDFs have significantly higher ^{207}Pb/^{204}Pb than the compositions of recent ocean island basalts, indicating high U/Pb ratios in early Earth history that points towards subducting sediments (Figure 1b). In contrast, the Pb isotope trend records mixing with a source that corresponds to Archaean lithosphere that has lost its U and Th (Figure 1b). Thus the combined data suggest that saline HDFs-bearing young diamonds in the southwestern Kaapvaal Craton formed from fluids that record an isotopic signature of the involvement of three components – an LREE-enriched Archaean lithosphere that has lost its U and Th, and subducting surface material that includes both recent sediments and a mantle-like component (i.e. subducting slab). This confirms the involvement of subducting surface material in the formation of saline HDFs and highlights the reworking of old lithospheres by younger metasomatic fluids during diamond formation events.

Radiogenic ^{3}He/^{4}He <1 Ra and aggregated nitrogen in B centers of carbonatitic and silicic HDFs and their host diamonds, respectively (Figure 2), indicate an older Paleozoic to Proterozoic formation event compared to the event(s) that produced saline HDFs. The Sr-Nd-Pb isotope signature of these HDFs also indicates an old lithospheric source but one that was not U and Th depleted (Figure 1). However, T_{DM} ages and He model ages do not simply correlate; this is expressed by old T_{DM} ages and young He ages for the silicic HDFs compared to the carbonatitic ones. While, such discrepancy suggests a scenario of reworking old lithospheres by younger metasomatic fluids, similar to the observed isotopic signature for saline HDFs, the exact nature of possible sources responsible for the silicic and carbonatitic compositions is still vague and the subject of future research.

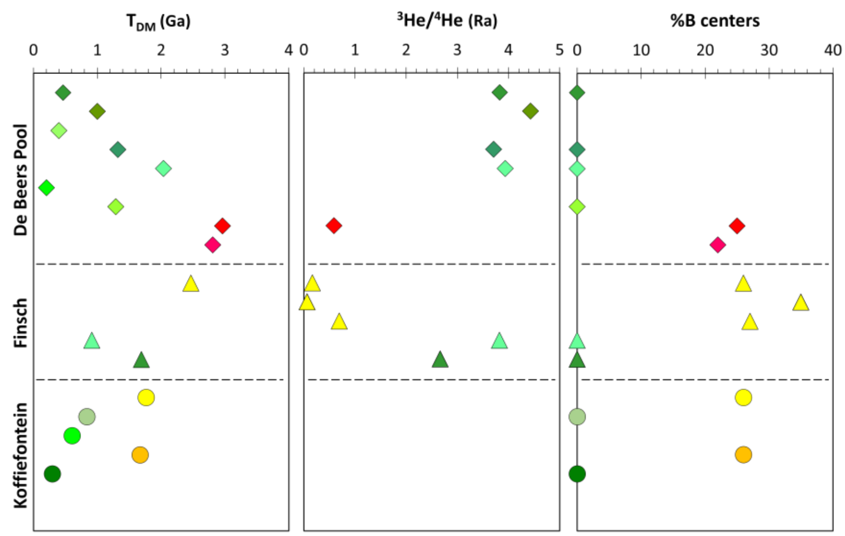

Figure 2: Nd model ages (T_{DM}; this study), $^3He/^4He$ ratios and nitrogen aggregation (Weiss et al., 2021) of HDF-bearing diamonds from the southwestern Kaapvaal Craton, South Africa. Mine locality and HDF type are distinguished by data point shape and color, respectively, as in Figure 1.

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