

Sources of Carbon in Inclusion Bearing Diamonds

Thomas Stachel¹, Jeff W. Harris² and Karlis Muehlenbachs¹

¹Earth and Atmospheric Sciences, University of Alberta, Edmonton, T6G 2E3, Canada

²Department of Geographical and Earth Sciences, University of Glasgow, G12 8QQ, U.K.

Introduction

Over the last four decades diamond source regions in the Earth's mantle have been characterized geochemically and mineralogically through the study of syngenetic mineral inclusions. Such studies provided overwhelming evidence that the principal source of diamonds is the subcratonic lithospheric mantle (e.g. Meyer, 1987; Gurney et al., 2005) with its highly depleted peridotitic and volumetrically much less abundant (<5%, Schulze, 1989; McLean et al., 2007) eclogitic and pyroxenitic reservoirs. The character (solid, fluid, melt), redox state (methane or carbonate bearing) and source (primordial versus subducted) of diamond forming carbon is, however, still the subject of debate (e.g. Kirkley et al., 1991; Cartigny, 2005). Diamonds of both peridotitic and eclogitic paragenesis show prominent modes in $\delta^{13}\text{C}$ at about -5‰ (± 1) (see Cartigny, 2005 for a recent review). This common mode is generally assumed to represent mantle derived carbon (Deines, 1980). Eclogitic diamonds show a blurred second "mode" at about $-14 \pm 5\text{‰}$ that has been interpreted as representing either subducted organic matter (Sobolev and Sobolev 1980; Kirkley et al., 1991; McCandless and Gurney, 1997, and references therein), primordial inhomogeneity of the mantle (Deines, 1980) or open system fractionation (Javoy et al., 1986; Cartigny, 2005). Irrespective of the carbon source, the preferred distribution of eclogitic diamonds along veins (Schulze et al., 1996; Taylor and Anand, 2004) indicates metasomatic precipitation.

In order to place additional constraints on the origin of diamond carbon we compiled a data base of 1646 carbon isotopic analyses that are all linked to the chemical composition of mineral inclusions recovered from the same diamonds. To evaluate the apparent common mode for lithospheric diamonds at about -5‰ on a more detailed level we use a class size for histograms of 0.25‰ (rather than the usual binning in 1‰ classes). This narrower binning interval is a much closer reflection of the high analytical precision in the determination of $\delta^{13}\text{C}$ in diamond, which typically is in the range of 0.05 to 0.10‰ (1 sigma, c.f. Deines, 1980; Donnelly et al., 2007) in the various studies comprising our data base.

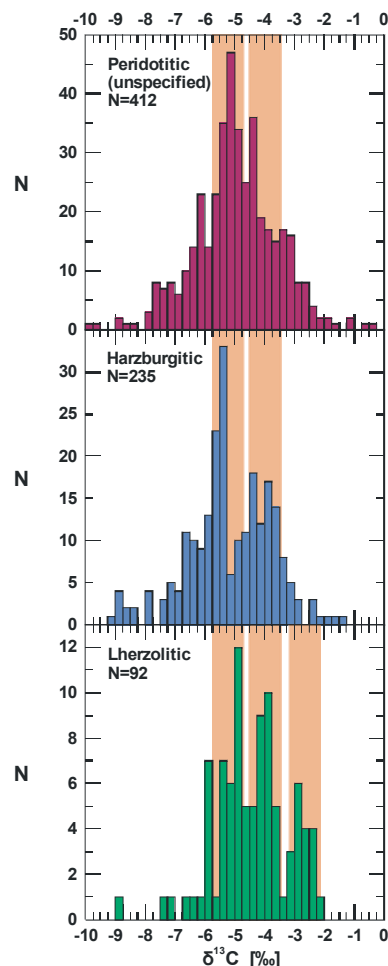


Figure 1: Carbon isotopic composition of diamonds with peridotitic inclusions. "Unspecified" paragenesis refers to diamonds without garnet and/or clinopyroxene inclusions.

Diamonds with peridotitic inclusions

Re-examining the prominent mode at -5‰ ($\delta^{13}\text{C}$) for peridotitic diamonds worldwide on a detailed level reveals that it is actually composed of multiple abundance peaks (Fig. 1). Harzburgitic diamonds have a distinct mode at about $-5.5 \pm 0.25\text{‰}$ but show two more abundance peaks between -4.5 and -3.75‰ . Lherzolititic diamonds have a mode at -5.0 to -4.75‰ , a prominent mode at about -4.5 to -3.75‰ and a third mode at -3.0 to -2.25‰ . Harzburgitic diamonds with $\delta^{13}\text{C} > -4.5\text{‰}$ are not restricted to, but are particularly

common in diamonds from the Central- and West-African cratons. For the lherzolitic paragenesis, the higher mode (-3.0 to -2.25‰) is clearly present in diamonds from the Kalahari, the Central- and the West-African cratons, but so far, has not been documented elsewhere (e.g. for the Slave Craton).

Radiometric dating of syngenetic mineral inclusions suggests that lherzolitic diamonds commonly form subsequent to generally Mesoarchean harzburgitic diamonds (Shirey et al., 2002; Gurney et al., 2005). The overall shift of lherzolitic diamonds towards isotopically heavier compositions, therefore, may reflect a temporal evolution in the isotopic composition of diamond carbon.

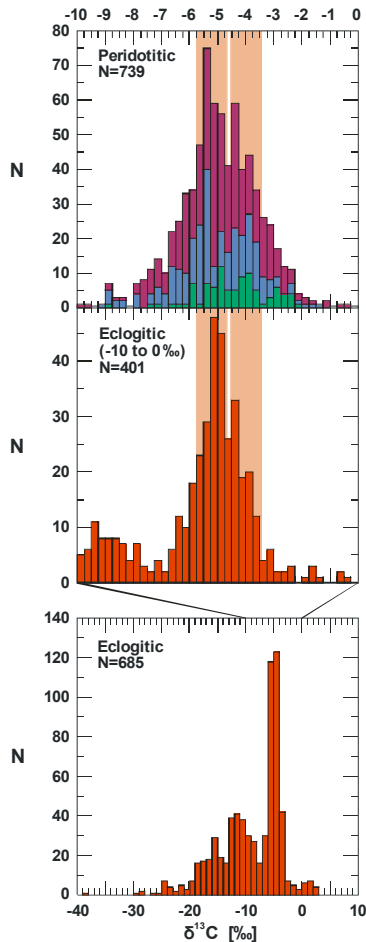


Figure 2: Carbon isotopic composition of diamonds with eclogitic inclusions. Peridotitic diamonds (see Fig. 1 for colour coding) are shown for comparison.

Diamonds with eclogitic and websteritic inclusions

Our database for eclogitic diamonds is strongly dominated by samples from the Kalahari Craton (402 of 685), where the prominent mode at around -5.0‰ is actually composed of two modes at -5.25 to -4.75‰ and -4.5 to -3.75‰ that correlate well with the two main modes for peridotitic diamonds worldwide (Fig. 2). In our eclogitic database the latter mode (-4.25 ± 0.25‰) is unique to the Kalahari Craton, the former

mode (-5.0 ± 0.25‰) is also seen for eclogitic diamonds from the Slave but is notably absent, e.g. among well represented eclogitic diamond populations from the Amazon and Kimberley cratons. The isotopically light tail for eclogitic diamonds extends to about -20‰ with scattered data down to -38.5‰. Visible modes in this tail reflect an overrepresentation of individual well studied localities: e.g. a mode at -13 to -9‰ is entirely due to a large data set from Argyle (Jaques et al., 1989). Isotopically heavy “eclogitic” diamonds (>0‰) appear to be limited to the New South Wales alluvial deposits and may derive from meta-rotundites (Group B of Davies et al., 2003).

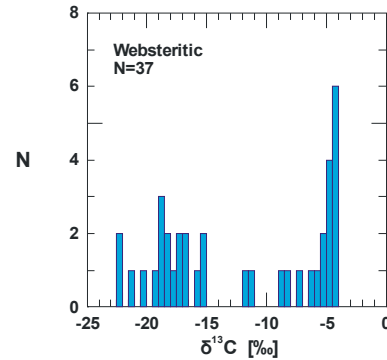


Figure 3: Carbon isotopic composition of diamonds with websteritic inclusions.

The carbon isotopic composition of websteritic diamonds (only 37 samples, Fig. 3) shows strong dependence to locality. Mantle-like carbon isotopic compositions are prominently represented by websteritic diamonds from Namibia and De Beers Pool, whereas at Jagersfontein and Orapa websteritic diamonds are isotopically light (~-20‰). For Jagersfontein there is also a distinct similarity in the isotopic ranges of websteritic and deep eclogitic diamonds (with majoritic garnet inclusions). Websteritic diamond sources are commonly assumed to relate to cumulate enriched deep portions of oceanic slabs or to result from interaction of slab derived melts with mantle peridotite (see Stachel and Harris, 2008). Organic matter should not be present in either scenario and hence the occurrence of exclusively light carbon isotopic values at Orapa and Jagersfontein requires a different explanation. Open system isotopic fractionation involving CO₂ is a viable option for olivine-free websteritic sources but does not explain the observed bias towards highly negative isotopic compositions. To the contrary, the similar ranges in δ¹³C for websteritic and sublithospheric diamonds from Jagersfontein point to a common carbon source possibly directly reflecting a slab signature (Tappert et al., 2005).

Discussion and Conclusions

Considering the overall variability in diamond carbon isotopic composition and mineral inclusion chemistry, the absence of discernible correlations between the two implies that inclusion chemistry reflects source

composition but – at least for major elements – is largely independent of a predicted evolution of the diamond precipitating fluids through Rayleigh type open system fractionation processes (e.g. Cartigny, 2005).

The observation of discrete modes in $\delta^{13}\text{C}$ for different diamond forming events through space and time may suggest that the isotopic composition of carbon in the Earth's mantle is heterogeneous (Deines, 1980) and possibly reflects secular trends. However, the existence of isolated primordial reservoirs with distinct carbon isotopic compositions cannot be evoked as the sole reason for the observed isotopic variations in diamonds since it would require that distinct lithospheric mineralogies (e.g. peridotite versus eclogite) consistently tap distinct primordial reservoirs. Alternatively, diamond precipitation, including peridotitic sources, involves at least some carbon that has been processed through a crustal cycle. The possible secular evolution in carbon isotopic composition, indicated, for example, by a shift in mode towards higher $\delta^{13}\text{C}$ values from harzburgitic to lherzolithic diamonds, may reflect an increasing or compositionally changing contribution of external (i.e. subducted) carbon sources. Progressive devolatilization of marine carbonates (initial $\delta^{13}\text{C}$ of $0 \pm 5\%$, Schidlowski et al., 1983) during subduction would drive their isotopic composition towards lower and hence mantle like $\delta^{13}\text{C}$ values, possibly rendering slab derived and mantle derived diamond carbon isotopically indistinguishable.

Similarities between eclogitic and peridotitic diamonds extending to a detailed level – e.g. the observation that at the Kalahari Craton the main mode at -5% actually comprises a second mode at -4.5 to -4.25 for both harzburgitic and eclogitic diamonds – suggests that carbon for both suites is derived from common sources. The observed ranges and particularities for the various diamond suites and parageneses then may be explained through a combination of slab signatures (e.g. different contributions of carbonates and organic matter) and isotopic fractionation (in the slab and the percolating metasomatic agent).

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