



Dated eclogitic diamond growth zones reveal variable recycling of crustal carbon through time



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ABSTRACT

Monocrystalline diamonds commonly record complex internal structures reflecting episodic growth linked to changing carbon-bearing fluids in the mantle. Using diamonds to trace the evolution of the deep carbon cycle therefore requires dating of individual diamond growth zones. To this end Rb–Sr and Sm–Nd isotope data are presented from individual eclogitic silicate inclusions from the Orapa and Letlhakane diamond mines, Botswana. $\delta^{13}\text{C}$ values are reported from the host diamond growth zones. Heterogeneous $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.7033–0.7097) suggest inclusion formation in multiple and distinct tectono-magmatic environments. Sm–Nd isochron ages were determined based on groups of inclusions with similar trace element chemistry, Sr isotope ratios, and nitrogen aggregation of the host diamond growth zone. Diamond growth events at 0.14 ± 0.09 , 0.25 ± 0.04 , 1.1 ± 0.09 , 1.70 ± 0.34 and 2.33 ± 0.02 Ga can be directly related to regional tectono-magmatic events. Individual diamonds record episodic growth with age differences of up to 2 Ga. Dated diamond zones have variable $\delta^{13}\text{C}$ values (-5.0 to -33.6% vs PDB) and appear to imply changes in subducted material over time. The studied Botswanan diamonds are interpreted to have formed in different tectono-magmatic environments that involve mixing of carbon from three sources that represent: i) subducted biogenic sediments (lightest $\delta^{13}\text{C}$, low $^{87}\text{Sr}/^{86}\text{Sr}$); ii) subducted carbonate-rich sediments (heavy $\delta^{13}\text{C}$, high $^{87}\text{Sr}/^{86}\text{Sr}$) and iii) depleted upper mantle (heavy $\delta^{13}\text{C}$, low $^{87}\text{Sr}/^{86}\text{Sr}$). We infer that older diamonds from these two localities are more likely to have light $\delta^{13}\text{C}$ due to greater subduction of biogenic sediments that may be related to hotter and more reduced conditions in the Archaean before the Great Oxidation Event at 2.3 Ga. These findings imply a marked temporal change in the nature of subducted carbon beneath Botswana and warrant further study to establish if this is a global phenomenon.

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1. Introduction

Diamonds have formed from metasomatic reactions involving C–H–O–S-rich melts (Stachel et al., 2005) possibly over the last 3.52 Gyr (Smart et al., 2016) and therefore potentially record the evolution of the Earth's deep carbon cycle. Diamonds derived from the sub-continental lithospheric mantle (SCLM) are dominated by two distinct parageneses: peridotitic (P-type) and eclogitic (E-type) containing both silicate and sulphide inclusions (Stachel and Harris, 2008). Terrestrial diamonds have $\delta^{13}\text{C}$ values that range from -41% to $+5\%$ with P-type diamonds defining a narrower range with 96.7% between -10 and -1% with a mean at -5% (Cartigny et al., 2014). Carbon isotope compositions of eclogitic diamonds cover the full range with a mode at -5% (Cartigny et al., 2014). Models proposed to explain the variability of carbon iso-

tope compositions in diamond include: heterogeneous accretion of the Earth's mantle, crustal subduction recycling, and/or isotopic fractionation of carbon in the mantle (Cartigny et al., 2014; Sverjensky and Huang, 2015 and references therein). In addition to the extensive C-isotope diamond dataset, there is a large body of stable isotope data on diamond and its inclusions (Shirey et al., 2013), but spatial relationships between the inclusions and diamond growth zones are generally lacking as most isotopic analyses were undertaken by bulk combustion. The recognition of extreme S and O isotope compositions in sulphide and silicate inclusions respectively in E-type diamonds (e.g., Chaussidon et al., 1987; Ickert et al., 2015), as well as mass independent S isotope anomalies (Cartigny et al., 2009; Farquhar et al., 2002; Thomassot et al., 2009), provides unambiguous evidence of a recycled Archaean crustal component in a significant proportion of E-type diamonds, but as yet there are insufficient coupled and comprehensive stable isotope studies to determine the exact proportion of E-type diamonds that incorporate recycled material.

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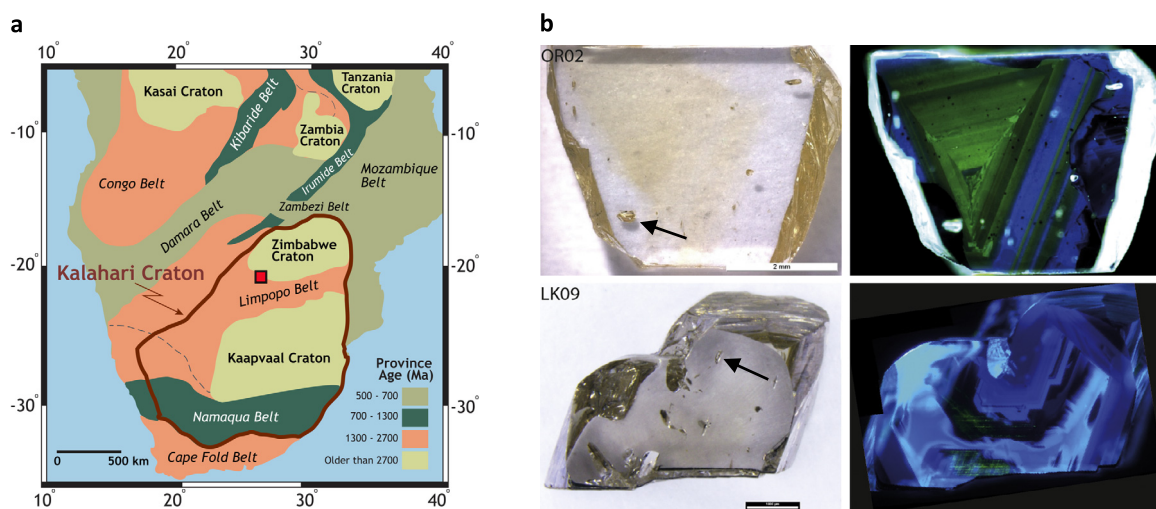


Fig. 1. a) Large scale tectonic map of Southern Africa showing the location of the Orapa and Lethakane diamond mines on the edge of the Zimbabwe Craton; indicated by a red square. Modified after Priestley et al. (2006). b) Reflected light, and Diamond ViewTM and cathodoluminescence images of Orapa diamond O2 and Lethakane diamond O9 respectively. Diamond ORO2 has a yellow core and garnet inclusions in the colourless rim. The yellow core is associated with green CL layers, whereas the colourless rim corresponds to the almost non-luminescent homogeneous growth zone after a resorption event. Diamond O9 is characterised by tangential octahedral growth, with dominantly blue CL colours and two dark CL zones that contain yellow–green slip lines. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Diamond formation in the mantle involves precipitation from metasomatic media at P – T conditions where the amount of solute in the fluid and the solubility of H_2O in silicate melts have converged along a miscibility gap making the distinction between fluid and melt disappear (Salters and Stracke, 2004). In this context the terms melt and fluid are interchangeable and we use fluids from here on. Due to the complex growth histories recorded in many diamond populations (Bulanova et al., 2005; de Vries et al., 2013a), determining any temporal variation in sources of diamond-forming fluids requires knowledge of $\delta^{13}C$ values from diamond growth zones of known age. Initial geochronology of diamonds involved pooling several hundred inclusions from different diamonds to obtain Sm–Nd isochrons on garnets and clinopyroxenes (Richardson et al., 1990), Nd model ages on pooled garnets (Richardson et al., 1984) and Pb–Pb model ages on pooled sulphides (Kramers, 1979). In the 1990s the geological meaning of the “average” ages of these pooled inclusions was questioned (e.g., Shimizu and Sobolev, 1995). Therefore, a dating technique for single sulphides using Re–Os (Pearson et al., 1998) was developed. The recent application of 10^{13} Ohm current resistors to thermal ionisation mass spectrometry now makes it possible to measure small (>40 μm) individual silicate inclusions from diamond with Nd amounts down to >10 pg (Koornneef et al., 2014). This advance allows direct comparison of ages produced from earlier studies, mostly of pooled silicate inclusions and allows individual growth zones to be dated. We report here the first Rb–Sr and Sm–Nd isotope systematics of small (2.1 to 110.5 μg) individual eclogitic garnet ($n = 10$) and clinopyroxene ($n = 9$) inclusions from 16 diamonds from the Orapa and Lethakane diamond mines, Botswana. Each diamond was first characterised for growth structure (Fig. 1b) and individual growth zones for nitrogen content, nitrogen aggregation state and $\delta^{13}C$ values. With this integrated methodology we determine the carbon isotope composition of individual dated diamond zones to evaluate potential temporal changes in carbon isotope composition of eclogitic diamond-forming fluids. The Orapa and Lethakane mines (Fig. 1a) were chosen because of the reported large range in carbon isotope compositions (Cartigny et al., 1999; Deines and Harris, 2004) that is ideal for investigating temporal carbon isotope variation in the mantle.

2. Orapa and Lethakane mines

The Orapa and Lethakane diamond mines located in North-Eastern Botswana on the edge of the Zimbabwe Craton are ~ 40 km apart (Fig. 1a). Despite abundant peridotitic xenoliths, the mines contain predominantly eclogitic diamonds as identified by mineral inclusions. A small percentage (2–6%, dependent on size) of the diamonds from the Lethakane mine ($\sim 62,500$ visually examined in this study) have inclusions. The majority are sulphides ($\sim 90\%$) and only $\sim 10\%$ silicates. Of the latter, eclogitic silicates predominate ($>80\%$). Counting inclusion abundances of $\sim 30,000$ diamonds from Orapa (this study) suggest E-type diamonds are similarly dominant. Previous studies have shown Botswanan diamonds have a wide range of nitrogen contents and carbon isotope compositions. Eclogitic and websteritic diamonds of Lethakane have nitrogen contents between 12 and 1846 ppm and IaB aggregation from 0 to 100% and $\delta^{13}C$ values of -3.9 to -33.5% , of which between 20 and 50% have $\delta^{13}C$ values below -14% (Deines and Harris, 2004; this study). Previous work on diamonds from Orapa shows similarly large variations in N and C systematics as reported from Lethakane with 24–1351 ppm N, 0–78% IaB and a $\delta^{13}C$ of -3.4 to -22.3% of which $\sim 30\%$ have $\delta^{13}C$ below -14% (Cartigny et al., 1999; Deines et al., 1993, 1991). Based on nitrogen aggregation state, previous research on Botswanan diamonds suggested that low $\delta^{13}C$ values were associated with greater depth (Deines et al., 1993, 2009) and/or older eclogitic diamonds (Deines et al., 1997).

3. Methods

3.1. CL and FTIR

Central plates of the diamonds were characterised for growth structure by cathodoluminescence imaging (CL; Lethakane) and Diamond ViewTM (Orapa). Representative images of two diamonds are given in Fig. 1b and detailed diamond images and interpretations of growth structures are given in SI Fig. 1. Nitrogen content and aggregation state of individual growth zones were measured on a FT/IR Jasco-470 plus equipped with a Jasco Irtron IRT-30 infrared microscope at the Utrecht University. Transmission spectra were collected over the range 4000 – 650 cm^{-1} with a resolution of

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