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Episodic diamond growth beneath the Kaapvaal Craton at Jwaneng Mine, Botswana

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Abstract
Important implications for the interior workings of the Earth can be drawn by studying diamonds and their inclusions. To better understand the timing and number of diamond forming events beneath the NW margin of the Kaapvaal Craton, a comprehensive reassessment of Jwaneng’s diamond populations has been undertaken. We report new inclusion abundance data from the visual examination of ~130,000 diamonds that validate the predominance of an eclogitic diamond suite (up to 88%) with on average 5% inclusion-bearing diamonds (with inclusions >10 μm in size). From this population, polished plates from 79 diamonds of eclogitic and peridotitic paragenesis have been studied with cathodoluminescence (CL) imaging and infrared spectroscopy (FTIR) traverses. The majority (80%) record major changes in N concentration and aggregation states, as well as sharp boundaries in the CL images of individual plates that are interpreted to demarcate discrete diamond growth events. In addition, bulk FTIR data have been acquired for 373 unpolished diamonds. Silicate inclusions sampled from distinct growth zones define 2 compositional groups of omphacites and pyrope-almandines associated with different N contents in their diamond hosts. These findings reinforce previous observations that at Jwaneng at least seven individual diamond forming events can be identified—3 peridotitic and 4 eclogitic. The results demonstrate that detailed examination of diamond plates by CL imaging and FTIR traverses is necessary to unveil the complex history recorded in diamonds.

Keywords Nitrogen aggregation · FTIR · Silicate inclusions · Cathodoluminescence

Introduction
Diamonds are considered ‘ancient messengers’ from Earth’s interior. Inclusions trapped during distinct diamond forming events provide insight into the temporal tectono-magmatic evolution of the subcontinental lithospheric mantle (SCLM) and the deep carbon cycle. However, the absolute number of diamond-forming events recorded in the mantle beneath an individual diamond mine and the scale of these events, remains unknown. Hence, it is vital to establish the timing of diamond forming events: e.g., by analysing individual sulphide inclusions for Re-Os isochron ages (Pearson et al. 1998; Wiggers de Vries et al. 2013). Initial attempts have also been made to date individual silicate diamond inclusions using the Sm-Nd isochron method e.g., at the Orapa kimberlite cluster in Botswana and at Finsch, Kimberley and Venetia diamond mines, South Africa (Koornneef et al. 2017; Smith et al. 1991; Timmerman et al. 2017). These data indicate multiple temporally-distinct diamond forming events at the same diamond mine. Similar conclusions have been reached in studies of the 240 Ma Jwaneng kimberlites on the NW rim of the Archaean Kaapvaal Craton (Kinny et al. 1989). Based on C and N stable isotope ratios, N concentration and N aggregation state data obtained by Fourier-transform infrared
(FTIR) spectroscopy and major element compositions of inclusions, multiple eclogitic and peridotitic populations have been inferred: the age of eclogitic diamond formation at Jwaneng was previously determined using a 2-point Sm-Nd isochron on composite silicate inclusions and yielded an age of 1540 ±20 Ma with indication of a second population formed at around 1 Ga (Richardson et al. 1999). Similarly, sulphide inclusions define 2 generations of eclogitic diamonds, one with a Proterozoic (1.5 Ga) age and another suite of Archean (2.9 Ga) age (Richardson et al. 2004; Thomassot et al. 2009). There are no isochron ages available for the peridotitic diamond suite at Jwaneng and only their relative age is constrained based on inclusion chemistry and C-isotope and FTIR data from the diamond hosts (Deines et al. 1997; Stachel et al. 2004). Those FTIR and C-N-isotope data for peridotitic diamonds were acquired on the same fragments but there are no references which part of the diamond (i.e., core or rim) these pieces represent (Cartigny et al. 1998; Deines et al. 1997; Thomassot et al. 2009).

The present study reports a detailed assessment of the diamond populations at Jwaneng to further constrain the number of diamond-forming events beneath an individual mine. New inclusion abundance data from the visual examination of ~130,000 diamonds refine proportions of different inclusion suites. Of these, bulk FTIR data were acquired for 373 whole diamonds, and polished plates from 79 diamonds of eclogitic and peridotitic paragenesis were studied through cathodoluminescence (CL) imaging, with FTIR traverses conducted and major element inclusion data interpreted based on recognised growth zones.

Samples and methods

Over a 4-year period (2013 to 2017) run of mine production was examined from Jwaneng Mine (north, centre and south pipes) with diamonds ranging in size from 0.1 to 0.8 carat (Diamond Trading Company (DTC) sieve classes +07, +09, +11, and 3 grainer) at the Diamond Trading Company in Gaborone, Botswana (DTCB). In total, over 130,000 diamonds were characterised visually for their inclusion content using a binocular microscope. Inclusions were defined as recognisable minerals >10 μm in size. The main distinction criterion is based on differences in colour between, e.g., eclogitic pyrope-almandine garnet (orange), omphacitic clinopyroxene (cpx; pale green) and kyanite (blue) inclusions and peridotitic pyrope garnet (purple), olivine or orthopyroxene (yellow to colourless) and chromite (brown to black) inclusions. Sulphide inclusions were almost ubiquitously associated with a characteristic rosette fracture system around the inclusion. If not otherwise reported, the examined diamonds are of gem and near-gem quality.

Sample selection concentrated on diamonds containing multiple large (>50 μm) garnet, clinopyroxene and sulphide inclusions. After morphological characterization, 79 diamond plates of eclogitic (69) and peridotitic (8) paragenesis, as well as 2 sulphide-bearing diamonds were laser cut through the centre of the diamond along the (110) plane to produce plates of 0.3 to 1.8 mm thickness, followed by surface polishing on a cast iron scaife plate. The polished plates were photographed on a Nikon SMZ1500 microscope using its extended depth of focus (EDF) imaging software. Cathodoluminescence imaging was performed on the uncoated diamond plates on a Cambridge Image Technology Ltd. cold cathode luminescence 8200 mk3 instrument (attached to an optical microscope) operating with an acceleration voltage of 10 kV, a beam current of 400 μA and camera exposure times between 1 to 15 s.

Infrared absorbance spectra were collected on a number of instruments with different experimental settings. Bulk spectra of 373 randomly sampled diamonds from Jwaneng run-of-mine production (DTC + 09 gem and near-gem) were measured using Perkin Elmer and Bruker Alpha spectrometers during the sampling campaigns at DTCB. Spectra comprised 10 scans over the range 4000 to 400 cm−1 with a resolution of 8 cm−1.

Based on observations from the CL imaging, FTIR traverses were acquired from representative parts of the central plates. The majority of the plate analyses were performed using a FT/IR Jasco-470+ instrument equipped with a Jasco IRT-30 infrared microscope over the spectral range 4000 to 650 cm−1 with a resolution of 4 cm−1. The remaining plate analyses were performed using either a Thermo-Nicolet Nexus spectrometer attached to a continuum infrared microscope (4 cm−1 resolution) or a Thermo Nicolet iN10MX infrared microscope (2 cm−1 resolution). Depending on the thickness of the plate, the aperture was set between 50 to 100 μm. After background acquisition prior to every sample measurement, 5 to 20 spots were measured from core to rim on each plate.

All spectra were processed using the automated version of DiaMap (Howell et al. 2012a, b) to determine N content and aggregation state. The data collected using the Thermo Nicolet iN10MX were also processed using a Python program (Kohn et al. 2016; Speich et al. 2017). Given that both programs use a similar approach for N deconvolution (normalisation, subtraction of a Type IIa diamond spectrum, fitting of N components while adding a linear correction), they produced data in good agreement. Spectra with <50 atomic ppm (at.ppm) N were reprocessed manually using the Excel version of DiaMap, to provide greater confidence in reported values. Nitrogen concentrations were calculated from absorption coefficients at 1282 cm−1: [NA] = 16.5 ppm (Boyd et al. 1994) and [NB] = 79.4 ppm (Boyd et al. 1995). Errors for N content and N aggregation (expressed as %B = 100[B/(A + B)])
are typically ±10% and ±5%, respectively, but uncertainties increase with plate thickness (see Kohn et al. (2016) for more details) and the size of the aperture, especially for spectra recording low N contents (<50 at.ppm). In particular, it should be noted that if spectra of heterogeneous, zoned diamonds are collected using whole diamonds or thick plates it is likely that the analysed volume will contain diamond with variable N concentrations. If this is the case, the measured N will be the average within the analysed volume, but the N aggregation will be heavily weighted towards the high N regions (Kohn et al. 2016). Furthermore, the ‘points’ measured by FTIR of diamond plates do not correspond exactly with the zoning elucidated by CL that shows near-surface features of the plate, whereas FTIR absorption represents a conical volume through the sample. This is particularly important if data points from the transects are close to growth zone boundaries, resulting in spectra that reflect contributions from more than one growth zone. Hence, sampling points were positioned to avoid clear boundaries between growth zones.

Given the low spectral resolution at which some bulk analyses were performed, it is not always possible to accurately differentiate low N samples from those typically deemed as ‘nitrogen-free’ (Type II). The present study considers all measurements with <20 at.ppm N as ‘nitrogen below detection limit’. Since the diamonds show no blue colouration and B-centre related absorption in FTIR spectra these spectra are classified as ‘Type IIA’.

For a subset of samples encapsulated inclusions were liberated from the plates and off-cuts using a steel crusher. Single inclusions will be measured for Sm-Nd geochronology in the future, so inclusions were not polished before analysis to avoid sample loss. Garnet (gnt) and cpx inclusions were mounted on carbon tape and a horizontal surface used for electron microprobe analyses. The EMPA analyses were performed using a JEOL JXA-8350F instrument with an acceleration voltage of 15 kV, a beam current of 25 nA and a beam size of 1 μm at Utrecht University. The CITZAF correction method was applied to the raw data. Further analytical details follow the protocol of Timmerman et al. (2015) that stipulates normalisation of the data to 100% and only includes measurements within 2 standard deviations of the mean. The normalisation of the major element data, which is based on average compositions of individual analyses (n = 3 to 22) with totals from 92 to 100 wt%, allows direct comparison of the inclusion compositions.

Results

Relative proportions of inclusion suites

On average, 5% of the diamonds at Jwaneng contain inclusions (Table 1). Sulphide inclusions are the most abundant (~63%, Fig. 1). Diamonds containing multiple sulphides are more abundant but the sulphides are smaller in size compared to diamonds containing single sulphide inclusions. A visual distinction between eclogitic and peridotitic sulphide inclusions was only possible when silicate inclusions were also present in the diamond, and diamonds with combined eclogitic silicate and sulphide inclusions account for ~5% of the inclusion-bearing diamonds. Eclogitic inclusions dominate the silicate population, averaging 69% (Table 1). Six percent of inclusions were unidentified silicates, 6% contained P-type silicate inclusions (Fig. 1), <0.1% had combined peridotitic silicate/ sulphide inclusions. A detailed overview of the 15 individual production parcels examined for relative inclusion proportions can be found in the Online Resources (ESM 1).

Diamond morphology

Polished plates were mainly prepared from octahedral to dodecahedral diamonds, but irregular shapes and macles were also present. Surface features show resorption, stepped faces and trigons. The majority of these diamonds were colourless but some diamonds with different shades of yellow and brown, 2 green coated (from radiation staining) and one pink diamond were also present. Further details are presented in the Online Resources (ESM 2).

CL imaging

Cathodoluminescence imaging of the central plates (Fig. 2) reveals growth histories of the individual diamonds ranging from a single well-defined growth zone (Fig. 2b) to samples having multiple (2 to 4+) growth zones that are recognized by sharp, successive boundaries (Fig. 2e–i). A growth zone may be compromised of multiple similar (genetically related) growth layers. In contrast, resorption horizons show irregular geometry (Fig. 2d core) and can be difficult to distinguish from superimposition of different growth directions (Fig. 2f intermediate zone), especially when successive precipitation of growth layers was involved in diamond formation (Fig. 2f intermediate zone), and diamond growth progressed through intermediate zones.

FTIR analyses of bulk diamonds

Nitrogen content and aggregation states of bulk (unpolished) diamonds within each of the 3-studied production parcels ranges from N below detection limit (<20 at.ppm) to 1466 at.ppm N and 0 to 100 %B, respectively (Fig. 3). Nine diamonds are pure Type IaA, 17 are Type IaB, 4 are Type IIa, the remaining are all Type IaAB (i.e. 10–90 % B) as listed in the Online Resources (Table ESM 3).
FTIR analyses of central plates

The majority of the diamond plates are Type IaAB (51), one diamond (JW407) is Type IaA, 17 have Type IIa growth zones but also record detectable N (>20 at.ppm) in at least one other major growth zone (Fig. 2h–j), 12 diamonds are entirely Type IIa (Fig. 2a). Together with insights from the CL imaging, each plate was subdivided into distinct growth zones (i.e., core, intermediate, rim) where N contents showed a variation >100 at.ppm between individual FTIR analyses as listed in the Online resources (Table ESM 4). Further, the plates were classified into 4 main groups: (A) Type IIa (15%; 10 E-, 2 P-types) as described above; (B) showing homogeneous growth with fairly constant N content and aggregation (3%; 1 E- & 1 P-type); (C) systematic changes in N content from core to rim with (C1) increasing (25%; 19 E-, 1 S-type), (C2) decreasing (38%; 25 E-, 4 P-, 1 S-types) and (C3) increasing and decreasing (16%; 12 E- & 1 P-type) between individual growth zones; and (D) showing non-systematic variations (3%; 2 E-types) in N content and aggregation state associated with irregular growth, possibly relating to plates not cut through the growth centre in some cases.

Individual eclogitic diamonds of group C (>80% of all studied diamond plates) record 2 (e.g., JW047, JW111; Fig. 2h, e), 3 (e.g., JW024, JW060; Fig. 2g, k) or 4 (e.g., JW033, JW194; Fig. 2d, l) distinct growth zones. Group C peridotitic diamonds have 2 (JW243; Fig. 2j) or 3 (JW083, JW089, JW102; Fig. 2i) distinct growth zones. Further transects of N content and aggregation state of the remaining 79 diamond plates are provided in the Online Resources (ESM 5).

Nitrogen content and aggregation state recorded in traverses range from N below detection limit to 1368 at.ppm N and 0 to 100 %B. These data cover most of the range of previously reported N contents (up to 1990 at.ppm N) and aggregation data for Jwaneng (Deines et al. 1997; Thomassot et al. 2009). The range of N contents and aggregation states within individual diamonds is, however, larger in the present dataset. In sample JW141, the N content varies from 1070 at.ppm in the core to 90 at.ppm in the intermediate zone and 400 at.ppm in the rim, while in JW351, the aggregation varies from 97 %B in the core to 2 %B in the rim. This compares with previous data that reported maximum differences of 216 at.ppm (Cartigny et al. 1998) and 402 at.ppm and 28% in JW0020 (Thomassot et al. 2009).

Deines et al. (1997) proposed that the N aggregation characteristics in their sample population (cleaved diamond chips, n = 68) formed 2 groups (0–10 %B; 50–70 %B). These groups are not observed in either the bulk or plate data presented here. Only a single peak at ~95%B for the plate data is observed (see Online Resources ESM 6).

Modelled mantle residence temperatures

The N aggregation state of a diamond in the mantle varies strongly with residence temperature (and to a lesser extent with time) and therefore can be used to obtain insight into a diamond’s thermal history. If the approximate formation time of a diamond (or a distinct growth layer) is known, a time averaged residence temperature can be calculated (Leahy and Taylor 1997).

Temperatures modelled for the FTIR data of plates and bulk diamonds in Fig. 3 were calculated for diamond genesis at 1, 1.5 and 2.9 Ga and kimberlite eruption at 240 Ma, based on ages reported in the literature (Richardson et al. 1999, 2004). Assuming diamond formation at 1540 Ma, mantle residence temperatures of bulk diamonds and the plates vary from ~1010 to >1250 °C. Some plates (Fig. 3) have a relatively homogeneous temperature record (JW020, JW033, JW047), while others seem to reflect a gradual shift (e.g., JW024, JW044, JW102) that in some cases might not be beyond the uncertainty in the data (e.g. JW044). In contrast, other examples imply distinct changes of up to several tens

Table 1 Relative inclusion proportions in diamonds at Jwaneng

| DTC sieve class | Diamonds | Silicates [%] | Sulphides [%] | Eclogitic silicates and sulphides [%] | Eclogitic inclusions* [%] | Total inclusions** [%] |
|----------------|----------|---------------|---------------|--------------------------------------|--------------------------|-----------------------|
|                |          | E  | P  | UN | Single | Multiple |                |                        |                        |
| +07            | 27,500   | 23.1 | 6.1 | 6.9 | 24.4  | 29.2     | 10.4            | 72.0                   | 4.6                     |
| +09            | 48,800   | 18.3 | 5.6 | 5.3 | 28.0  | 39.4     | 3.4             | 66.7                   | 5.2                     |
| +11            | 37,700   | 21.1 | 6.5 | 5.9 | 23.2  | 39.6     | 3.7             | 66.7                   | 5.1                     |
| 3Gr            | 10,300   | 27.1 | 7.0 | 6.7 | 33.1  | 42.0     | 6.4             | 70.9                   | 4.3                     |
| Total          | 130,000  | 20.4 | 5.7 | 5.8 | 25.9  | 37.3     | 4.8             | 68.7                   | 5.0                     |

Details on the proportions of individual production lots can be found in the Online Resource (ESM 1). E, eclogitic inclusions; P, peridotitic inclusions; UN, unidentified silicate

*Percentage of eclogitic inclusions calculated from eclogitic silicates plus combined eclogitic silicates and sulphides over the total number of silicate and combined eclogitic silicate and sulphide inclusions

**Percentage of total amount of inclusions calculated by total number of inclusions over number of diamonds
of degrees between individual growth zones that cut across isotherms (e.g., JW033, JW060, JW114, JW147, JW175, JW194). Overall, the FTIR data of the plates (Fig. 3) show clustering of diamond cores at higher N aggregation (70 to 90 %B; ~1200 to 1250 °C for all 3 calculated ages) and to a lesser extent of lower aggregation states (10 to 30 %B; ~1050 to 1150 °C), where also the majority of rims is located. Data from intermediate growth zones span the whole range. It should be noted that if the analysed volume is not homogeneous in N concentration (which is certain to be the case for most bulk diamonds and some points from traverses on plates), there will be a systematic and predictable overestimate of the mantle residence temperatures (Kohn et al. 2016) that may have some effect on the distribution of the FTIR data in Fig. 3.

**Inclusion compositions**

**General remarks**

Inclusion compositional data are summarized in Table 2 for 30 omphacite and 15 pyrope-almandine inclusions liberated from 16 diamonds. All omphacite inclusions (Fig. 4a) are characterised by moderate molar Mg# (71 to 83) and low molar Cr# (<5). Pyrope-almandine inclusions (Fig. 4b) have low Cr₂O₃ (0.5 wt%) with CaO between 4 and 14 wt%. These compositions fall in the eclogite (G4 and G5) garnet fields as defined by Grütter et al. (2004) and are comparable with previous studies of Jwaneng inclusions (Richardson et al. 1999; Richardson et al. 2004; Stachel et al. 2004). The compositional variation between inclusions, liberated from the same diamond, is between zero to 2.3 wt% in CaO and < 0.3 wt% in Cr₂O₃ for garnet and between 0.3 to 5.6 in Mg# and < 0.7 in Cr# for cpx.

**Coupled diamond-inclusion relationships**

The omphacite inclusions define 2 subgroups with (1) low-Mg# (71 to 83) and low-Cr# from 0.2 to 1.4 (JW030, JW010, JW111, JW137, JW138, JW147, JW194) from diamond growth zones with ≥ 50 to 700 at.ppm N, and (2) omphacite inclusions with high-Mg# (79 to 87) and high-Cr# from 2.8 to 4.7 from diamond growth zones having ≤ 50 at.ppm (JW048, JW058, JW070, JW080, JW112, JW116, JW124). The majority of low-Ca (3.8 to 6.1 wt% CaO) pyrope-almandine inclusions (JW058, JW137, JW147 core) are from growth zones characterized by ≤ 50 at.ppm N (Fig. 4b), only JW175 shows high N contents (~1000 at.ppm). The pyrope-almandine inclusions (JW068, JW147 intermediate, JW194) from diamond growth zones with N contents from 50 to 300 at.ppm are all characterised by high CaO contents (7.9 to 13.5 wt%).

**Discussion**

**Inclusion abundance**

The average proportion of inclusion-bearing diamonds for 130,000 Jwaneng diamonds, comprising DTC sieve sizes +7, +9, +11, and 3 grainers (equivalent to a combined range from 0.1 to 0.8 carats), is 5%. Previous studies of inclusion-
A general predominance of the eclogitic inclusion suite for Jwaneng (Table 1) was also noted in previous studies (Gurney et al. 1995). Stachel et al. (2004) and references therein) link the eclogitic predominance to Proterozoic magmatic and tectonic events that triggered eclogitic diamond formation, potentially resorbing parts of any pre-existing peridotitic diamond suite.

Growth characteristics

General remarks

Studies of diamond populations worldwide almost always reveal variations in colour and morphology (Gurney et al. 2004). Deines et al. (1997) suggested a rough classification of Jwaneng diamonds based upon their colour, morphology and paragenesis. They concluded E-type diamonds were more often colourless, partially resorbed and of irregular shape, compared to the P-type diamonds that were more commonly octahedral and green coated or brown and deformed. No coherent relationships were reported between N characteristics and diamond paragenesis. The same lack of relationship is observed in the present dataset. While bulk FTIR analyses in

![Image](https://example.com/image.png)

**Fig. 2** Cathodoluminescence images and N traverses of representative polished plates. The diamonds illustrate common growth features (in groups A to D defined in the text) with samples a) JW116 (A), b) JW044 (B), c) JW062 (D), d) JW033 (C1), e) JW111 (C1), f) JW114 (C1), g) JW024 (C2), h) JW047 (C2), i) JW102 (C2), j) JW243 (C2), k) JW060 (C3) and l) JW194 (C3). White symbols correspond to the location where FTIR spectra were acquired and are subdivided into major growth zones: core (filled circles), intermediate (diamonds) and rim (squares). Type Iia spectra (<20 at.ppm N) are marked as below detection limit (b.d.l.)
Table 2  Averaged major element composition (EPMA results quoted in wt%) for inclusions in diamonds from Jwaneng

| Sample   | Zone* | n** | SiO₂  | TiO₂  | Al₂O₃ | Cr₂O₃ | FeO   | MnO   | MgO   | CaO   | Na₂O  | K₂O   | Total |
|----------|-------|-----|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| JW030 cpx A | core  | 15  | 51.4  | 0.27  | 5.4   | 0.05  | 4.7   | 0.06  | 12.6  | 18.0  | 0.07  | 2.9   | 0.28  | 95.8  |
| JW048 cpx A | rim   | 12  | 50.8  | 0.16  | 4.6   | 0.20  | 6.4   | 0.10  | 15.1  | 16.3  | 0.06  | 2.2   | 0.41  | 96.3  |
| JW058 cpx A |        | 10  | 52.2  | 0.12  | 5.1   | 0.24  | 5.9   | 0.12  | 16.8  | 13.8  | 0.08  | 2.6   | 0.16  | 97.1  |
| JW058 cpx B | core  | 15  | 53.4  | 0.13  | 5.0   | 0.22  | 5.8   | 0.11  | 15.1  | 14.2  | 0.08  | 3.0   | 0.16  | 97.2  |
| JW058 cpx C | core  | 9   | 50.2  | 0.13  | 5.2   | 0.25  | 5.9   | 0.11  | 17.3  | 13.9  | 0.08  | 2.3   | 0.16  | 95.5  |
| JW070 cpx B | core  | 11  | 51.9  | 0.15  | 4.1   | 0.27  | 6.4   | 0.10  | 14.8  | 15.8  | 0.07  | 2.2   | 0.39  | 96.1  |
| JW070 cpx C | core  | 7   | 54.5  | 0.15  | 3.9   | 0.27  | 6.3   | 0.10  | 14.4  | 16.0  | 0.07  | 2.3   | 0.44  | 98.5  |
| JW070 cpx D | core  | 9   | 51.0  | 0.14  | 4.0   | 0.26  | 6.4   | 0.10  | 15.4  | 16.1  | 0.08  | 2.1   | 0.38  | 95.9  |
| JW070 cpx G | core  | 10  | 48.5  | 0.15  | 4.0   | 0.26  | 6.3   | 0.10  | 15.4  | 15.8  | 0.07  | 2.2   | 0.42  | 93.3  |
| JW070 cpx H | core  | 11  | 53.7  | 0.14  | 4.3   | 0.26  | 6.3   | 0.09  | 15.6  | 16.3  | 0.06  | 2.4   | 0.39  | 99.5  |
| JW070 cpx A | rim   | 11  | 54.3  | 0.12  | 3.6   | 0.26  | 5.4   | 0.11  | 15.3  | 16.2  | 0.08  | 2.4   | 0.29  | 98.0  |
| JW101 cpx A | int   | 11  | 53.9  | 0.27  | 4.8   | 0.07  | 7.0   | 0.11  | 13.8  | 15.3  | 0.06  | 2.3   | 0.43  | 98.1  |
| JW111 cpx A | core  | 12  | 51.5  | 0.41  | 11.9  | 0.04  | 8.4   | 0.10  | 15.6  | 12.3  | 0.02  | 6.4   | 0.09  | 95.7  |
| JW111 cpx B | core  | 9   | 53.2  | 0.45  | 9.1   | 0.05  | 4.4   | 0.04  | 6.0   | 11.9  | 0.01  | 8.2   | 0.09  | 93.5  |
| JW111 cpx C | core  | 13  | 52.1  | 0.44  | 12.3  | 0.05  | 4.6   | 0.05  | 8.2   | 12.2  | 0.02  | 5.9   | 0.09  | 95.9  |

*Zone = approximate location of inclusion in core, intermediate (int) and rim zones based on visual observation only
**n = number of individual analyses per grain

* M. U. Gress et al.
this study and the literature provide broad insights into the N characteristics of the prevalent diamond populations (Online Resources ESM 6), this approach averages the significant N concentration variation found in diamonds that record multiple growth events and systematically overestimates the N aggregation.

Further information on growth relationships between Jwaneng diamonds and their eclogitic inclusions can be found in a complementary study of Davies et al. (2018). The diamonds of that study were also sampled during the campaign at DTC.

**Peridotitic diamonds**

Deines et al. (1997) state that peridotitic diamonds \((n = 20)\) tend to have lower N contents compared to the eclogitic suite but are similar in N aggregation state and postulate 3 different growth environments for olivine-bearing diamonds based on N-content, C-isotopes and mineral composition. We make similar observations based on the FTIR data from the eight studied peridotitic plates with 2 that are pure Type IIa, 4 that include Type IIa growth zones and 2 with 330 to 760 at.ppm N and highly variable aggregation; 0–88 %B. The combined CL and FTIR characteristics of the individual peridotitic diamonds establish that they were formed in a single growth event (e.g., JW020, Fig. 3) or contain 2 (e.g., JW243, Fig. 2j & Fig. 3) to 3 (e.g., JW102, Fig. 2i & Fig. 3) distinct growth zones that record changes in modelled residence temperatures of several tens of degrees from core to rim. Clearly, an expanded sample set and detailed chronology of these growth zones is required to establish the intervals between individual growth zones and whether the timescale of P-type diamond growth is comparable to that of the more common E-type diamonds or distinct as suggested by Stachel et al. (2004).
Eclogitic diamonds

By combining the N-characteristics obtained from FTIR with the CL imagery of diamond plates, the Jwaneng diamonds are seen to represent a highly varied population. Five samples that exhibit multiple growth zones plot along a single isotherm (e.g., JW083, JW455, JW94, JW196, JW210 in Fig. 3). As the different growth zones of individual diamonds exhibit a near constant thermal history, this suggests that these individual diamonds were formed by pulses of fluid in a single diamond-forming event or formed within a short period of time (100’s Myr, i.e. the effective resolution obtained in Fig. 3). Samples JW044 (Fig. 2b) and JW058 also potentially formed in a single event. In contrast, the majority (>90%) of the eclogitic diamond plates record 2 (e.g., JW111, Fig. 2e; JW047; Fig. 2h), 3 (e.g., JW024, Fig. 2g; JW060, Fig. 2k) or 4 (e.g., JW033, Fig. 2d; JW194, Fig. 2l) changes in modelled mantle residence temperatures, ranging from 10s of °C (JW024) to >100 °C (e.g., JW033, JW111, JW175, JW194; Fig. 3), between individual growth zones of single diamonds. Assuming near-constant temperatures throughout the whole residence period, the cores of these plates typically show higher N aggregation compared to the rims, which most probably implies that there is a significant time gap between growth of the core and rim. If the core experienced a hotter period of residence prior to rim formation, this inferred time gap decreases. In principle, there is an indefinite number of combinations of time and temperature possible if the corresponding inclusion ages are unknown. Hence, accurate dates for individual growth zones (i.e. core and rim) are required to more accurately model their residence temperatures (Kohn et al. 2016) and to constrain the time gap between individual growth zones. In a few cases there also appears to be evidence for higher model temperatures in diamond rims (JW003, JW030, JW045). This cannot be correct, as the cores must be at least as old the rims and any high temperature event experienced by the rim would also have been experienced by the core. The most likely explanation to these observations are modelled temperature anomalies resulting from overlapping zones (Kohn et al. 2016). Other possible explanations may include a deviation from the expected kinetics of A to B centre aggregation, perhaps caused by some additional defect incorporation (e.g., see Fisher and Lawson (1998) for the effect of Ni and Co on N-aggregation in diamond).

The presence of different inclusion populations in diamonds of distinct N contents is interpreted to record modifications in the lithospheric mantle beneath the Kaapvaal Craton involving different metasomatic fluids. Omphacites with low Mg# and Cr# and high-Ca pyrope-almandines (Fig. 4) are included in diamond growth zones with 50 to 700 at.ppm N whereas omphacites with high Mg# and Cr# and low-Ca pyrope-almandines come from diamond growth zones with low N (generally ≤50 at.ppm). The data from these inclusions suggests that either the original mantle protoliths have different C-buffering capacities or that fluids of different composition metasomatized the lithosphere at different depths and, potentially, times. This may explain why JW175 has high N contents (1000 at.ppm) although the garnets are low-Ca pyrope-almandines. The interpretation of multiple diamond forming events is consistent with observations based on coexisting garnet and clinopyroxene equilibration temperatures and FTIR data described in Deines et al. (1997) as well as the inclusion dating studies of Richardson et al. (1999, 2004) that argued for diamond formation at 2.9, 1.5 and 1.0 Ga.

We conclude that our FTIR dataset is very useful in revealing the varied time-temperature characteristics of a diamond population, but that either temperature or age constraints from inclusion studies are required for a robust and quantitative interpretation. The dataset emphasizes the complex growth histories recorded by the diamond suite. The record of up to 4 distinct growth zones in single silicate inclusion-bearing diamonds (JW033, Fig. 2d; JW194, Fig. 2l; Fig. 3) requires further chronology to establish the time intervals between the genesis of these individual zones. Such data would also allow for more rigorous interpretation of the FTIR dataset to determine whether the diamond populations reflect a few large-scale diamond-forming events (i.e. samples of the same age with varied thermal histories, indicating diamond growth over a significant depth profile of the mantle), or alternatively more frequent diamond formation on a much smaller, local scale as a result of confined movements of metasomatic fluids.

Conclusion

The data from this study support five main conclusions. (i) Counting statistics for 130,000 diamonds from Jwaneng emphasise the predominance of an overwhelming (>88%) eclogitic diamond suite with individual production parcels containing up to 10% inclusion-bearing diamonds (5% on average). (ii) Over 80% of the eclogitic and peridotitic diamond plates record changing FTIR characteristics with 2, 3 or 4 individual growth zones potentially spanning extended timescales during the evolution of the Kaapvaal Craton, while a minority of inclusion-bearing diamonds were likely formed in a single event. This observation has not been quantified for inclusion-free diamonds. (iii) FTIR data acquired for bulk diamonds integrate N defects from multiple growth zones and consequently do not record the total internal variability, especially when major changes between individual growth zones are present. (iv) Two compositional subgroups of eclogitic garnet and cpx inclusions with different characteristics in N content of their diamond hosts were identified and imply different diamond forming conditions (i.e. a combination of different protoliths, formation depth, fluid composition
and most probably age). (v) Detailed examination of diamond plates by CL imaging and FTIR identifies at least 7 individual diamond-forming events recorded in peridotitic (3) and eclogitic (4) diamonds. The scale and timing between these individual events is yet to be fully resolved, as is their distribution with depth.

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