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7 **The Evolution of Calcite-Bearing Kimberlites by Melt-Rock Reaction –**
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9 **Evidence from Polymineralic Inclusions within Clinopyroxene and Garnet**
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11 **Megacrysts from Lac de Gras Kimberlites, Canada**
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7
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9

10 11 12 **Abstract**

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15 Megacrystic (> 1 cm) clinopyroxene (Cr-diopside) and garnet (Cr-pyrope) xenocrysts within kimberlites
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17 from Lac de Gras (Northwest Territories, Canada) contain fully crystallized melt inclusions. These
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19 “polymineralic inclusions” have previously been interpreted to form by necking down of melts at mantle
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21 depths. We present a detailed petrographical and geochemical investigation of polymineralic inclusions
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23 and their host crystals to better understand how they form and what they reveal about the evolution of
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25 kimberlite melt. Genetically, the megacrysts are mantle xenocrysts with peridotitic chemical signatures
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27 indicating an origin within the lithospheric mantle (for the Cr-diopsides studied here ~4.6 GPa, 1015°C).
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29 Textural evidence for disequilibrium between the host crystals and their polymineralic inclusions (spongy
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31 rims in Cr-diopside, kelyphite in Cr-pyrope) is consistent with measured Sr isotopic disequilibrium. The
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33 preservation of disequilibrium establishes a temporal link to kimberlite eruption. In Cr-diopsides,
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35 polymineralic inclusions contain phlogopite, olivine, chromite, serpentine, and calcite. Abundant fluid
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37 inclusion trails surround the inclusions. In Cr-pyropes, the inclusions additionally contain Al-spinel,
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39 clinopyroxene, and dolomite. The major and trace element compositions of the inclusion phases are
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41 generally consistent with the early stages of kimberlite differentiation trends. Extensive chemical
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43 exchange between the host phases and the inclusions is indicated by enrichment of the inclusions in major
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45 components of the host crystals, such as Cr₂O₃ and Al₂O₃. This chemical evidence, along with phase
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47 equilibria constraints, supports the proposal that the inclusions within Cr-diopside record the
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49 decarbonation reaction: dolomitic melt + diopside → forsterite + calcite + CO₂, yielding the observed
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51 inclusion mineralogy and producing associated (CO₂-rich) fluid inclusions. Our study of polymineralic
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53 inclusions in megacrysts provides clear mineralogical and chemical evidence for an origin of kimberlite
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4 that involves the reaction of high pressure dolomitic melt with diopside-bearing mantle assemblages
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6 producing a lower pressure melt that crystallizes a calcite-dominated assemblage in the crust.
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10 **Keywords**

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13 Kimberlite; Cr-rich Megacrysts; Polymineralic Inclusions; Melt Inclusions; Decarbonation Reaction;
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15 Kimberlite Evolution
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20 **Introduction**

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23 Despite over 100 years of research on kimberlites, the nature and origin of their primary melt(s) and their
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25 magmatic evolution path to the observed low pressure mineral assemblages remain controversial and
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27 elusive issues. The entrainment of, and reaction with, xenoliths and xenocrysts spanning the entire
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29 lithosphere greatly complicates the search for the primary kimberlite melt, and the picture is further
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31 obscured by post-emplacement alteration (Mitchell 1986; Mitchell 1995; Price et al. 2000; Kopylova et al.
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33 2007; Sparks et al. 2009; Kjarsgaard et al. 2009). Two of the key issues in kimberlite research that are still
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35 debated are: 1) Whether the primary kimberlite is carbonatitic (Dawson 1971; Dawson and Hawthorne
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37 1973; Russell et al. 2012; Kamenetsky and Yaxley 2015; Kamenetsky 2016), a carbonated silicate melt
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39 (e.g., Nielsen and Sand 2008; Brey et al. 2008), or a highly saline carbonatite (e.g., Kamenetsky et al.
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41 2012); and: 2) The mechanism for generating the calcite-bearing kimberlites observed in the crust – all
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43 experimental evidence suggests that a primary melt formed from the melting of a peridotitic source must
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45 produce Mg-rich carbonate, namely dolomite (e.g., Wyllie and Huang 1975; Dalton and Presnall 1998a).
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51 The idea that kimberlites start off as end-member carbonatites that then react with mantle phases, mostly
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53 orthopyroxene (opx), to create a more Si- and Mg-rich melt has recently gained ground following
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55 experimental studies (Russell et al. 2012; Kamenetsky and Yaxley 2015). However, so far attention has
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57 focused on opx dissolution as a dominant process in kimberlite magma evolution and eruption from the
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59 base of the lithosphere, although this has not yet been validated through experiments at upper mantle
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4 conditions (e.g., Stone and Luth 2016; Sokol et al. 2016), and could be reproduced only for a limited
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6 pressure range (Kamenetsky and Yaxley 2015). Here, we focus on the reaction of early high pressure
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8 kimberlite melt with clinopyroxene (cpx) and garnet (grt), manifest as solidified melt inclusions within
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10 kimberlite-hosted xenocrysts from the Lac de Gras kimberlite field (including the Diavik and Ekati
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12 diamond mines). Using reconstructed bulk compositions of these inclusions that trace extensive reaction
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14 with the host xenocrysts, we try to further constrain the possible nature of kimberlite melt at depth. Most
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16 importantly, we present evidence for reactions between the ascending melt and mantle minerals that lead
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18 to the eventual low pressure calcite-dominated mineralogy of kimberlites emplaced in the crust.
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22 23 Polymineralic Inclusions in Kimberlite Xenocrysts

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25 This study focuses on polymineralic inclusions (composed of multiple discrete mineral phases) that
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27 represent crystallized melt inclusions trapped in kimberlite megacryst phases (cpx and grt). Such
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29 polymineralic inclusions, also referred to as “spheroids” or “globules”, have been reported to occur in a
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31 range of different megacryst minerals from kimberlites in different localities worldwide since the mid
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33 1970s. Commonly reported phases in such inclusions are: phlogopite, olivine, carbonates (dominantly
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35 calcite), spinel, and serpentine. Haggerty and Boyd (1975) first described polymineralic inclusions in
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37 olivine megacrysts from the Monastery kimberlite in South Africa and interpreted them as early
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39 kimberlite melt with an immiscible sulphide liquid. Schulze (1985), studying inclusions in grt and cpx
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41 megacrysts, adopted this interpretation and extended it to propose that the trapped kimberlite melt may
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43 also constitute the magma from which the hosts crystallized. More recent studies of melt inclusions in Cr-
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45 diopside from the Diavik Diamond Mine in the Slave Craton, Canada, established a link to carbonatitic
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47 melts not in equilibrium with the host phases (van Achtebergh et al. 2002). In follow-up studies on the
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49 same samples, melt differentiation towards more silicate-rich melts was proposed, based on the
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51 observation of two end-member types of inclusions; carbonate-rich and more silicate-rich (van
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53 Achterbergh et al. 2004). Araújo et al. (2009) challenged the need for different inclusion end-members
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55 and stated that the spectrum of melt inclusions could “have formed through melt differentiation after
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4 wall–rock interaction and fractionation from melts with kimberlitic composition”. The presence of pure
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6 calcite in the inclusions has previously been attributed to disequilibrium unmixing and quenching in the
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8 final stages of kimberlite emplacement (van Achterbergh et al. 2004). Pivin et al. (2009) reported similar
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10 polymineralic inclusions within grt megacrysts in kimberlites from the Democratic Republic of Congo,
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12 and tentatively interpreted them as products of destabilization of an unknown original phase due to
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15 metasomatism.
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20 Geological Setting

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22 The Archean Slave Craton forms the north-western part of the Canadian Shield and is exposed in the
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24 Northwest Territories and Nunavut (e.g., Kusky 1989; Padgham 1992). The Slave Craton formed by
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26 tectonic accretion of a pre-3 Ga nucleus in the west to a Neoproterozoic juvenile arc in the east, creating a
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28 north-south trending suture (e.g., Davis et al. 1996). Collisional events in the Paleoproterozoic led to the
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30 formation of the Thelon-Taltson and Wopmay orogens that now partially surround the craton (e.g.,
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32 Isachsen and Bowring 1994). The Slave Craton hosts multiple kimberlite fields with various emplacement
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34 ages ranging from Cambrian to Eocene (e.g., Creaser et al. 2004; Heaman et al. 2004). The central Lac de
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36 Gras (LDG) kimberlite field, which contains the Diavik and Ekati diamond mines, was principally active
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38 from 75 to 45 Ma (Cretaceous to Eocene) (Sarkar et al. 2015).
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43 The LDG field is located east of the inferred craton suture (Fig. 1). Its kimberlite pipes define a 100 km
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45 by 200 km northwest-trending zone (Lockhart et al. 2004). The generally small pipes (2-12 ha surface
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47 area) intrude into metamorphosed Archean basement rock and are overlain by Quaternary glacial deposits
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49 (Fedortchouk and Canil 2004; Nowicki et al. 2004). For a detailed description of Diavik and Ekati
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51 kimberlites, the reader is referred to Moss et al. (2008) and Nowicki et al. (2004), respectively. The
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53 geochemistry of the LDG kimberlite field has been discussed in detail by Kjarsgaard et al. (2009). We
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55 studied inclusion-bearing xenocrysts from the Diavik and Ekati kimberlites (Fig. 1), where Cr-diopside
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57 hosted polymineralic inclusions are common.
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Analytical Methods

Polished thin sections of megacrysts, some with surrounding kimberlite, were prepared avoiding the use of water, to prevent the dissolution of inclusion material. Samples with abundant and well-preserved inclusions were selected for imaging in back-scattered electron (BSE) mode using a scanning electron microscope (SEM) in the Canadian Centre for Isotopic Microanalysis (CCIM).

Major and minor element analyses of megacrysts and inclusion phases were collected in multiple analytical sessions by wavelength dispersive X-ray spectroscopy (WDS) using a JEOL 8900 electron probe microanalyzer (EPMA) at the University of Alberta. Analytical conditions and secondary standard results for the different minerals are given in Supplementary Data (Table S1). Trace element analyses for phlogopite were obtained by laser inductively coupled plasma mass spectrometry (LA-ICP-MS) at the CCIM Arctic Resources Laboratory, University of Alberta, using a RESOLUTION 193 nm ArF excimer laser ablation system coupled to a Thermo Scientific ELEMENT XR 2 mass spectrometer. Measuring conditions and secondary standard results are given in Supplementary Data (Table S1). LA-ICP-MS data reduction was performed using the Iolite software (Paton et al. 2011).

To obtain modal compositions of the inclusions, six megacrysts of cpx and seven of grt from the Point lake kimberlite, Ekati, were selected for automated mineralogical analysis using QEMSCAN® at the Universidad Católica del Norte, Chile. A detailed description of this technique is given in Ayling *et al.* (2011) and the operating conditions for these analyses were similar to those in Menzies *et al.* (2015). EPMA compositions were used in the elemental bulk calculations using mass balance.

Strontium isotopes were measured by thermal ionization mass spectrometry (TIMS) at the CCIM Arctic Resources Laboratory using a Thermo Scientific TRITON-Plus mass spectrometer. Carbonates in the inclusions were dissolved directly with 6N HCl. Host megacrysts were crushed and clear fragments were picked, undergoing two cleaning cycles (in 6N HCl at 120°C overnight, and rinsed in MQ water). The crystal fragments were dissolved in a mixture of HF and HNO₃. Column chemistry procedures to separate

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4 Sr from other potentially interfering elements are described by Sarkar *et al.*, (2015). A drop of H₃PO₄ was
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6 added to the sample solution before drying down at 90 °C. TaCl₅ was added as an activator to the sample
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8 solution, which was then loaded to Re filaments. Two analyses of the NBS987 standard yielded a ⁸⁷Sr/⁸⁶Sr
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10 ratio of 0.71027 with a standard error of 0.00001. No corrections were made to the measured data.

15 Results

18 Megacryst Hosts

21 The cpx and grt samples in this study commonly exceed 1 cm in size and thus are megacrysts in a non-
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23 genetic sense (Mitchell 1986).

26 Cpx megacrysts were collected directly from drill core at the Diavik Diamond Mine. The host kimberlite
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28 is coherent (hypabyssal) kimberlite from the A154-North pipe containing abundant fresh olivine, plus
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30 ilmenite and grt megacrysts, and calcite of different textural populations. The cpx megacrysts are
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32 generally veined/fractured but often contain zones where the cpx has a fresh, clear appearance (Fig. 2a-c).
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34 Some cpx megacrysts show signs of post-emplacement reaction with the host kimberlite in the form of
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36 embayments or reaction rinds (brownish discoloration of the surrounding kimberlite, Fig. 2a). Calcite
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38 segregations within the host kimberlite were often observed in direct contact with cpx megacrysts (Fig.
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40 2b). Four cpx megacrysts (> 2 cm in size) containing abundant inclusions were selected for further
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42 analysis.
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46 Diavik grt samples were obtained from ~1 cm mineral concentrates (Fig. 2e). Their original spatial
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48 relationship to the kimberlite is unclear, however, some grains have attached remnants of the host
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50 kimberlite (kimberlite rinds). Grt crystals in this study have different genetic origins, as suggested by a
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52 color range from orange (eclogitic or low-Cr megacrystic) to red (lherzolitic or high-Cr megacrystic) and
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54 purple (harzburgitic) (Fig. 2e) (McLean et al. 2007). Polymineralic inclusions are most common in red grt
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56 grains (lherzolitic or high-Cr megacrystic). A total of 14 separate grt grains with inclusions were
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58 analyzed.
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4 Megacryst minerals from the Point lake and Wolverine kimberlite pipes, located on the Ekati Mine claim
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6 block, were also examined. Both cpx and grt were obtained from mineral concentrates (> 6 mm) and are
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8 likely to represent fragments of even larger crystals. A total of nine cpx grains and 17 grt grains from
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10 Point Lake were found to contain inclusions. From the Wolverine sample suite, three cpx grains and six
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12 grt grains with inclusions were selected.
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14 15 16 Classification and Thermobarometry

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18 The majority of the cpx and grt mineral hosts to the polymineralic inclusions classify as Cr-diopside and
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20 Cr-pyrope, respectively (see Supplementary Table S2 for EPMA analyses). Their Cr-rich compositions
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22 may suggest a genetic link to the Cr-rich megacryst suite, first documented by Egger *et al.* (1979). To
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24 obtain P-T estimates of **equilibration** for Cr-diopside megacrysts, the single cpx thermobarometer of
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26 Nimis and Taylor (2000) was used in combination with the compositional filters suggested by Grütter
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28 (2009). This **exercise** yielded temperatures averaging around 1015°C and pressures around 4.6 GPa
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30 (Table 1).
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33 34 35 Polymineralic Inclusions

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37 Polymineralic inclusions can be very abundant, **especially in cpx**, with up to 10 inclusions observed in
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39 single sample sections (Fig. 2c) (see also van Achterbergh *et al.* 2002; 2004). In total, 80 polymineralic
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41 inclusions have been found and imaged in 20 cpx grains, and 51 inclusions in 29 grt grains. Polymineralic
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43 inclusions are usually rounded and up to 2 mm in diameter (Fig. 2). Depending on whether their
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45 mineralogy is carbonate- or silicate-rich, they appear brownish-white or dark in thin section. The
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47 inclusions are generally located where veins/fractures with abundant fluid inclusions converge in the host
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49 grain (Fig. 2d, f). **Optical** microscopy and preliminary Raman spectroscopy indicate that the fluid
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51 inclusions are CO₂-rich (**Gleeson, S.A., pers. comm.**). Larger inclusions along these trails are often filled
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53 by phlogopite, calcite, and/or serpentine indicating that the veins are genetically related to the
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55 polymineralic inclusions (Fig. 2d, f). Hence, the polymineralic inclusions strictly are secondary in nature,
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57 as defined by Roedder (1984). However, although the veins occasionally connect to the host kimberlite
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4 (where preserved) the appearance of polymineralic inclusions and kimberlite is markedly different (Fig.
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6 2f). Some megacryst grains contain dark blebs close to grain edges that are in direct contact with the host
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8 kimberlite (Fig 2b). On the basis of similar mineralogy to the host kimberlite groundmass, the blebs most
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10 likely represent late-stage kimberlite melt that infiltrated the grain rims.

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14 It is important to distinguish polymineralic inclusions from other mineral inclusions. For example, cpx
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16 megacrysts can contain rounded inclusions of fresh olivine. In grt crystals (especially purple grains)
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18 mineral inclusions of olivine are common but often pervasively altered (Fig. 2e). The resulting alteration
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20 assemblage is remnant olivine, with serpentine, calcite, and Ni-rich sulphide (vaesite or pentlandite). The
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22 proportion of secondary calcite that replaces olivine can exceed 50%, so that altered olivine inclusions in
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24 grt can be mistaken for carbonate-rich polymineralic inclusions (described below).
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28 Mineralogy of Inclusions in Clinopyroxene

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30 Polymineralic inclusions in cpx are commonly surrounded by a reaction rim containing abundant micro-
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32 inclusions (Fig. 3a-c). EPMA element distribution maps (Fig. 4) show that these rims are enriched in Ca,
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34 and depleted in Al and Na (also see Fig. 9). The maps further highlight the presence of phlogopite, calcite,
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36 and serpentine in the micro-inclusions. Inside polymineralic inclusions, common phases are calcite,
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38 phlogopite, serpentine/chlorite, olivine, and chromite (Fig. 3a-c). Two end-member inclusion types are
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40 distinguished based on the groundmass mineralogy of the inclusions. The groundmass can be composed
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42 entirely of calcite, which may display bright and dark zones in BSE images (Fig. 3a). Alternatively, the
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44 groundmass is comprised of hydrated Mg-Fe-silicates (serpentine or chlorite) often with a distinctive
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46 vermicular texture (Fig. 3c). In such inclusions calcite appears as small euhedral crystals or as blebs. In
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48 intermediate inclusions calcite can have a colloform texture (Fig. 3b). Based on the observation that
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50 inclusions may either contain a calcite-rich or a serpentine/chlorite-rich groundmass, a division into
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52 'carbonate-rich' and 'silicate-rich' inclusions, as proposed by van Achterbergh et al. (2004), is justified.
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Mineralogy of Inclusions in Garnet

Phases observed in polymineralic inclusions in grt megacrysts are the same as those within cpx megacrysts, but further include Al-rich cpx, Al-spinel, and Ni-sulphides (vaesite or pentlandite) (Fig. 3d-f). Al-spinels can have chromite cores, evident in BSE images as bright inner zones. Phlogopite crystals commonly line the inclusion walls. Importantly, carbonates within polymineralic inclusions in grt can be more Mg-rich (dolomite and Mg-calcite) as well as calcite (Fig. 3e). Alteration rims showing an enrichment/depletion of the host grt around inclusions are absent. Instead, inclusions are surrounded by grt dissolution features akin to kelyphite rims usually observed at the grt-kimberlite interface (e.g., Canil and Fedortchouk 1999). Analyzing these kelyphite zones using energy dispersive X-ray spectroscopy (EDS) with a defocused electron beam yielded similar spectra to those of pristine grt. Both silicate-rich and carbonate-rich groundmasses are present, the latter being less frequent. Hence, a distinction into ‘carbonate-rich’ and ‘silicate-rich’ end-members also seems practical for polymineralic inclusions in grt megacrysts.

Polymineralic inclusions in both host phases can contain accessory apatite and sulfides. Representative QEMSCAN® maps of the end-member inclusion types in Cr-diopside and Cr-pyropite are shown in Supplementary Figure 1.

Mineral Chemistry of Inclusion Phases

Olivine

Within polymineralic inclusions from both Cr-diopside and Cr-pyropite megacrysts, olivine appears as typically euhedral, oblate to elongate crystals comprising up to 25% of the inclusion area. Individual olivine crystals are compositionally homogenous, whereas significant compositional variation can be observed among different inclusions (Supplementary Table S3). Olivine also occurs as single-phase mineral inclusions in some Cr-pyropite and in Cr-diopside megacrysts. For comparison, olivine in kimberlite rinds attached to megacryst hosts was also analyzed.

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4 Olivine compositions from polymineralic inclusions and as mineral inclusions in grt and cpx are
5 compared to LDG kimberlite olivine core and rim data (Fig. 5). The LDG kimberlitic olivine data can be
6 divided into two main trends: 1) cores with high NiO (and low CaO) at variable Mg/(Mg+Fe) (Mg#), and
7 2) rims with decreasing NiO (and increasing CaO) at relatively constant Mg# (arrow in Fig. 5a) (e.g.,
8 Fedortchouk and Canil 2004; Brett et al. 2009; Bussweiler et al. 2015).

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11 Olivines in kimberlite rinds attached to cpx and grt hosts plot along the expected trends for LDG
12 kimberlite olivine. In contrast, olivines within the polymineralic inclusions are distinguished by lower
13 NiO and higher MnO, CaO, and Cr₂O₃ contents with decreasing Mg# (Fig. 5). Olivines from
14 polymineralic inclusions in Cr-pyrope megacrysts have lower NiO, CaO, and Cr₂O₃ and higher MnO
15 compared to those in cpx megacrysts. Viewed together, olivines from inclusions in the two hosts define a
16 trend of decreasing NiO and Mg# that diverges off the olivine rim trend in kimberlites (Fig. 5a). A similar
17 trend has been documented for megacrystic olivine in kimberlites and attributed to crystallization from a
18 magma at depth (Boyd 1974).

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21 Single phase olivine inclusions all plot in the field for olivine from LDG peridotite xenoliths (Fig. 5d) and
22 partially overlap with compositions of olivines included in diamond (Stachel et al. 2003; Donnelly et al.
23 2007). Thus, altered olivine mineral inclusions in grt megacrysts, which can mimic polymineralic
24 inclusions in terms of mineralogy, are easily distinguished by the chemical composition of remnant
25 olivine, for example by their high NiO and Mg#.

26 Phlogopite

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28 Phlogopite (Supplementary Table S4) occurs in nearly all polymineralic inclusions, comprising up to 25%
29 of the inclusion area within Cr-diopside, and up to 50% of the inclusion area in Cr-pyrope. Phlogopite is
30 usually homogeneous, but crystals zoned in Cr₂O₃ are observed. Phlogopite also occurs as single-phase
31 mineral inclusions, and as intergrowths with other mineral inclusions.

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4 Major element compositions of phlogopite in polymineralic inclusions fall into the range of kimberlitic
5 phlogopite, but define a more restricted window in Mg# space (0.85 - 0.94; Fig. 6a). Phlogopite in
6 polymineralic inclusions is further distinguished by higher Cr₂O₃ and more restricted TiO₂. Such elevated
7 Cr contents have been observed in phlogopite from the Snap Lake kimberlite, NWT (Kopylova et al.
8 2010). Importantly, compared to global carbonatite- and kimberlite-derived phlogopite compositions
9 (Reguir et al. 2009), the phlogopite data from polymineralic inclusions fall into the kimberlite field (Fig.
10 6d), suggesting that the polymineralic inclusions considered to be ‘carbonate-rich’ are in fact also of
11 kimberlitic lineage.

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23 Compositional differences among phlogopite in polymineralic inclusions are controlled by the host
24 crystal rather than kimberlite pipe or inclusion type; phlogopite in polymineralic inclusions in Cr-pyrope
25 has higher Cr₂O₃ (up to 4.5 wt.%) and slightly lower Mg# (~0.90) than phlogopite in polymineralic
26 inclusions in Cr-diopside (Fig. 6). Phlogopite in polymineralic inclusions in Cr-pyrope also has higher
27 Al₂O₃ contents and extends to higher Na₂O (Supplementary Table S4). BaO contents in phlogopite are
28 relatively low in inclusions from either host phases, usually < 0.5 wt.% (0.2 wt.% on average), but tend to
29 be higher in inclusions in Cr-pyrope (up to ~1.4 wt.%) (Supplementary Table S4). Phlogopite as single
30 mineral inclusions, as megacrystic intergrowths (with grt, olivine and diopside), or cores of zoned
31 phlogopite have significantly lower Cr₂O₃ contents.

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43 Halogen contents in phlogopite were generally low (mostly F < 0.5 wt.%, Cl < 0.1 wt.%) (Supplementary
44 Table S4). The highest F concentration (0.86 wt.%) was measured in a phlogopite in a ‘carbonate-rich’
45 inclusion in Cr-diopside from Diavik. The highest Cl concentration (0.13 wt.%) was measured in a
46 megacrystic phlogopite intergrown with Cr-pyrope from Diavik.

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53 **Giuliani et al. (2016) performed a detailed investigation of phlogopite from the Bulfontein kimberlite,**
54 **South Africa. Compared to their compositional fields for groundmass phlogopite (late crystallization) and**
55 **high Ti-Cr cores (crystallization at depth), the trace element composition of phlogopite in polymineralic**
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4 inclusions are low in Zr, Ba, and Mn while extending to Cr-rich compositions and are thus clearly more
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6 similar to phlogopite crystallizing at depth (Fig. 6d-f).
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9 Carbonates

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11 Carbonate forms the groundmass in ‘carbonate-rich’ end-member inclusions. In intermediate to ‘silicate-
12
13 rich’ inclusions carbonate is present as distinct crystals with colloform texture, as blebs, or as euhedral
14
15 phenocrysts. Compositional zoning, evident as darker and brighter areas in BSE images, is common.
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17 Most analyzed carbonates are calcite with < 1.0 wt.% MgO; rarely calcite contains up to 5.0 wt.% MgO
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19 (Supplementary Table S5). Within ‘silicate-rich’ inclusions in Cr-pyrope megacrysts, dolomite (up to ~22
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21 wt.% MgO) is present. FeO contents are generally low in calcite (< 0.5 wt.%) but are higher in dolomite
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23 (up to ~4.5 wt.%).
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28 All analyzed carbonates in polymineralic inclusions plot within the reported compositional range of
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30 kimberlitic carbonates in terms of Ca# ($Ca/(Ca+Mg+Fe)$) and SrO contents (e.g., Armstrong et al. 2004)
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32 (Fig. 7a). In polymineralic inclusions, calcite has generally low SrO (< 1.0 wt.%). Higher SrO
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34 concentrations are restricted to bright zones (in BSE images) in either the groundmass of ‘carbonate-rich’
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36 inclusions in Cr-diopside or zoned single crystals in ‘silicate-rich’ inclusions in Cr-pyrope. Dolomite
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38 within inclusions in Cr-pyrope also has low SrO contents (< 0.5 wt.%) and thus overlaps dolomite
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40 compositions in kimberlite. Carbonates in polymineralic inclusions, especially in ‘silicate-rich’ inclusions
41
42 in Cr-pyrope, extend to higher MnO contents than kimberlitic calcite (Fig. 7b).
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46 Chromite and Spinel

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48 Chromite occurs in all polymineralic inclusions, whereas Cr-rich Al-spinel only occurs in polymineralic
49
50 inclusions in Cr-pyrope. No clear compositional differences were found between ‘carbonate-rich’ and
51
52 ‘silicate-rich’ inclusions (Supplementary Table S6).
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57 Kimberlite chromite data show a trend of decreasing Cr# and increasing Ti contents at relatively constant
58
59 Fe# ($Fe/(Fe+Mg)$), which is referred to as “magmatic spinel trend 1” according to the terminology of
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4 Mitchell (1986, 1995). The less common “magmatic trend 2” is characterized by Fe-enrichment prior to
5
6 an increase in Ti contents, which has been tentatively attributed to preceding crystallization of phlogopite
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8 (Mitchell 1995).
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11 Chromites in polymineralic inclusions classify as titanian-magnesian-chromite (TIMAC, Mitchell (1986))
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13 and overlap core compositions of chromite found in the kimberlite groundmass (Fig. 8). Such chromites
14
15 are characteristic for kimberlites and are not present in carbonatites (Mitchell 1986). Chromites in
16
17 polymineralic inclusions generally lie at the beginning of the two differentiation trends (Fig. 8). Analyses
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19 of chromites in polymineralic inclusions in Cr-diopsides, and some analyses in Cr-pyropes, plot along the
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21 beginning of magmatic trend 2 (Fig. 8b). This Fe-enrichment is in accordance with the abundant presence
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23 of phlogopite in the polymineralic inclusions. Co-crystallization of olivine may also play a role in Mg-
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25 depletion, as evident in the olivine differentiation trend (Fig. 5a).
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30 Cr-rich Al-spinels within inclusions in Cr-pyrope are characterized by low TiO₂ contents (usually < 2.0
31
32 wt.%). Such spinels have no counterpart in the groundmass of LDG kimberlites (Armstrong et al. 2004;
33
34 Roeder and Schulze 2008). However, some Cr-rich Al-spinels were found to contain cores of chromite.
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36 Such zoned spinels may also be the cause for mixed analyses of relatively low Cr# and elevated TiO₂
37
38 contents (Fig. 8c).
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42 Spinel in peridotitic mantle xenoliths (coarse spinel-grt peridotites from Jericho) overlaps spinel in
43
44 polymineralic inclusions in Cr# vs. Fe# space but can be distinguished by lower TiO₂ (Kopylova et al.
45
46 1999).
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49 Serpentine and Chlorite

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51 Serpentine/chlorite (Supplementary Table S7) is present in most polymineralic inclusions. In ‘carbonate-
52
53 rich’ inclusions, serpentine (Mg-rich) commonly replaces olivine, and chlorite (Al-bearing) replaces
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55 phlogopite. In ‘silicate-rich’ inclusions, an intergrowth of serpentine and chlorite forms the groundmass
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57 and can display a characteristic vermicular texture.
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4 Serpentine in kimberlites is defined by a broad range in Mg# (~0.80 - 0.95) and mostly has low Al₂O₃
5 contents (< 1.0 wt.%). However, Al₂O₃ concentrations up to ~8.0 wt.% have been reported in the
6 literature (e.g., Mitchell, 1986). Serpentine within inclusions in Cr-diopsides from LDG lies at the high-
7 Mg# end and extends to Al₂O₃ contents of up to ~5.0 wt.%, whereas serpentine within polymineralic
8 inclusions in Cr-pyrope can have even higher Al₂O₃ contents (up to ~9.0 wt.%) (Supplementary Fig.2).
9 Serpentine associated with altered olivine inclusions in Cr-pyrope has intermediate Mg# and Al₂O₃
10 contents, and low Cr₂O₃ and TiO₂ contents.
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20 Clinopyroxene in Polymineralic Inclusions

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23 Cpx is only present in polymineralic inclusions within Cr-pyrope hosts and occurs exclusively in ‘silicate-
24 rich’ inclusions. Cpx in polymineralic inclusions generally classifies as Al-rich, Cr-bearing augite and
25 less commonly diopside (Supplementary Table S8). It is chemically distinct from the Cr-diopside
26 megacrysts hosts and the modified cpx in the alteration rims around the polymineralic inclusions (Fig. 9).
27 There is some compositional overlap between cpx in inclusions in Cr-pyrope and the global cpx
28 megacryst trend, for example in Mg# and TiO₂ contents. However, cpx in inclusions is more enriched in
29 Cr₂O₃, Al₂O₃, and CaO, and has significantly lower Na₂O contents than typical cpx megacrysts (Fig. 9).
30 Most prominently, cpx in inclusions has higher MnO contents (clustering around 0.3 wt.%) than all other
31 cpx populations (Fig. 9f).
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43 Strontium Isotopes

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46 Sr isotopes were measured for ‘carbonate-rich’ inclusions and their Cr-diopside and Cr-pyrope hosts.
47 Because Rb concentrations were generally very low (commonly below detection limits), the measured
48 ratios were not corrected for radiogenic ingrowth of Sr since kimberlite eruption.
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54 Inclusions in Cr-diopside have ⁸⁷Sr/⁸⁶Sr ratios of 0.7049 to 0.7053, whereas inclusions in Cr-pyrope are
55 more radiogenic (0.7061 to 0.7071; Fig. 10). If a previous analysis of carbonate-rich globules in Cr-
56 diopside megacrysts by van Achterbergh et al. (2002) is included, there is overlap in the range of Sr
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4 isotopic values of the inclusions in the two hosts. Compared to groundmass calcite from the Jos
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6 kimberlite, Somerset Island (Malarkey et al. 2010), signatures of ‘carbonate-rich’ inclusions in both host
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8 minerals are less radiogenic than those of late-stage kimberlitic calcite that is presumed to be affected by
9
10 increased crustal contamination experienced by kimberlite melts during ascent plus subsequent low-T
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12 hydrothermal alteration. But there is overlap (Fig. 10) with a population of “early” kimberlitic calcite,
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14 interpreted to be the product of magmatic crystallization (Malarkey et al. 2010).
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18 Except for the most radiogenic ‘carbonate-rich’ inclusion in Cr-pyrope, the Sr signatures of the inclusions
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20 overlap with those of LDG kimberlites (Tappe et al. 2013). The host kimberlite (Diavik A154) has the
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22 most radiogenic signature of all LDG kimberlites and is slightly more radiogenic (0.70619) than
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24 inclusions in Cr-diopsides. The relatively radiogenic character has been previously attributed to derivation
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26 from a fertile peridotite domain within an otherwise highly depleted domain in the convecting upper
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28 mantle (Tappe et al. 2013). However, since LDG kimberlite isotope ratios were obtained from bulk
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30 samples, their more radiogenic Sr may be influenced by sampling of late-stage crustal components. The
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32 latter scenario seems plausible, because Sr isotope signatures measured in LDG perovskites - an early
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34 crystallizing phase in kimberlites - are lower (~0.70424) and have a narrower range (Sarkar et al. 2015).
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40 Importantly, the Sr isotope ratios of the LDG host megacrysts are significantly lower than those of their
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42 inclusions, indicating isotopic disequilibrium, as found also by van Achtebergh *et al.* (2002). In addition,
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44 it can be noted that the difference in Sr ratios between inclusion and host is smaller for Cr-diopside than
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46 for Cr-pyrope (Fig. 10).
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49 Reconstructed Bulk Compositions of Polymineralic Inclusions

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51 Major element bulk compositions of polymineralic inclusions were reconstructed using modal proportions
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53 obtained with QEMSCAN® or EPMA element maps and mineral compositions measured with EPMA.
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55 The overall uncertainty of the reconstructed bulk compositions is thus a function of 1) the uncertainty of
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57 the modal abundances and 2) the variability of the averaged mineral compositions used in the calculation.
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4 For 1), we have adopted a universal relative uncertainty of 5%, noting that this may be lower for major
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6 phases and higher for accessory phases. For 2), we observe that relative standard deviations of multiple
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8 EPMA analyses of a given component (in a given phase and in a given inclusion) are up to 10%. This
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10 uncertainty may be lower for major components (such as MgO, FeO and SiO₂) and higher for minor
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12 components (such as Na₂O and SrO). Thus, we report a propagated overall uncertainty of ~11% for each
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14 component of the reconstructed bulk compositions (Table 2).
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18 Oxide totals range from ~63 wt.% in ‘carbonate-rich’ inclusions to ~86 wt.% in ‘silicate-rich’ inclusions
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20 (Table 2). Low totals are attributed to major components that were not analyzed, such as CO₂ in
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22 carbonates and H₂O in phlogopite and serpentine. P₂O₅ and S hosted in accessory apatite and sulfides
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24 presumably have a minor effect on the bulk composition, as well.
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28 Notably, reconstructed bulk compositions of the inclusions plot along **nearly continuous** trends rather than
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30 forming discrete clusters of ‘carbonate-rich’ and ‘silicate-rich’ end-members (Fig. 11). The calculated
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32 bulk compositions of ‘carbonate-rich’ inclusions (>50% modal calcite) in Cr-diopside consistently lie
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34 below the threshold of 20 wt.% SiO₂ required for the designation of “carbonatite” (Le Maitre et al. 2002),
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36 whereas ‘silicate-rich’ inclusions extend to SiO₂ contents of up to ~40 wt.%. CaO steeply decreases with
37
38 increasing SiO₂ content (Fig. 11a) whereas MgO and FeO increase (Fig. 11b, c). Polymineralic inclusions
39
40 in the two hosts display similar general trends, but inclusions in Cr-pyrope are significantly more
41
42 enriched in the components FeO, Cr₂O₃, and Al₂O₃ (Fig. 11d), as expected from the mineral chemistry
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44 described in the previous sections.
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49 The Mg#s of the reconstructed bulk compositions have a large range from 0.76 to 0.98 (Table 2).
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51 Inclusions hosted in cpx have a mean Mg# of 0.93, whereas inclusions in grt have lower Mg# averaging
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53 around 0.81. It should be noted that Mg# is highest in inclusions where a great proportion of high-Mg
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55 serpentine and phlogopite is exposed. Mg# thus appears to be easily skewed by a sampling or cross-
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57 section effect (see Discussion).
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Discussion

Polymineralic Inclusions as Snapshots of Melt-Rock Reaction

Each polymineralic inclusion within Cr-diopside is surrounded by a halo of micro-inclusions of phlogopite and calcite within a matrix of recrystallized cpx depleted in the jadeite component (Fig. 4). We interpret this halo as a reaction rim between incoming melt and the original host phase. The textural and chemical character of these reaction rims is independent of whether the inclusion is of the ‘carbonate-rich’ or ‘silicate-rich’ type and is similar to the spongy rims commonly observed around the outside of cpx grains in mantle-derived xenoliths (Taylor and Neal 1989; Ionov 1998; Carpenter et al. 2002; Su et al. 2012; Lu et al. 2015). Similarly, Cr-pyropes display a kelyphitic reaction zone around polymineralic inclusions of both types. Kelyphite – typically as a fine-grained intergrowth of opx, spinel, olivine, phlogopite and glass – is a commonly observed product of reaction between kimberlite melt and grt xenocrysts (e.g., Canil and Fedortchouk 1999; Spetsius and Taylor 2002).

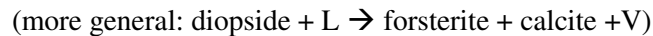
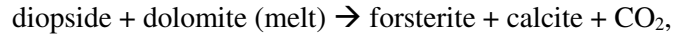
We interpret spongy textures in Cr-diopsides and kelyphite zones in Cr-pyrope as different textural representations of the same process, namely the reaction of a melt with mantle minerals during ascent. Thus, it seems likely that the original melt in the two different host minerals was the same, as previously suggested for polymineralic inclusions in cpx and grt megacrysts from Kentucky kimberlites (Schulze 1985).

New and published Sr isotopic data on the host megacrysts and inclusions support the reaction relationship deduced from textures (Fig. 10). The host megacrysts are less radiogenic than the carbonate within the polymineralic inclusions, but the range in Sr isotopic compositions of the inclusions trends towards their hosts. This trend is more evident for carbonate included within Cr-diopside, for which there are two possible explanations: 1) The reaction between melt and host scavenges more Sr from the Cr-diopside (usually containing ~200 ppm Sr) than from the Cr-pyrope (< 2 ppm Sr); 2) The reaction between melt and host Cr-diopside is stronger, as indicated by the presence of spongy rims around

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4 polymineralic inclusions that can be > 100 μm wide (Fig. 2d, 3, 4) and possibly by reaction rims around
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6 the outside of single Cr-diopside megacrysts (Fig. 2a). Polymineralic inclusions in Cr-pyrope tend to be
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8 smaller and their kelyphite zones narrower, so that the volume of interaction is smaller. **But both hosts**
9
10 **record clear evidence of reaction with the invading melt.**

14 Evidence for Decarbonation Reactions

16 Both textural and isotopic data indicate that the melt parental to the polymineralic inclusions has reacted
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18 with their **mantle-derived** hosts, with a potentially stronger reaction observed for Cr-diopsides. The
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20 possible driving factors behind these processes might lie in a suite of decarbonation reactions – reactions
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22 **that** release CO₂ – **long suspected to occur during the ascent of kimberlites** (e.g., Wyllie and Huang 1975;
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24 Egglar 1986). In addition to the commonly invoked reaction of melt with opx (enstatite + dolomite melt
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26 → forsterite + diopside + CO₂; “opx dissolution”), **the decarbonation reaction involving diopside** has the
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28 form:
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38 This reaction is expected to occur at pressures < 2.5 GPa (Stone and Luth 2016; Stone 2016).
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40 Polymineralic inclusions in Cr-diopside appear to preserve direct evidence of this reaction, as their
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42 mineralogy is dominated by the presence of pure calcite and forsteritic olivine, while the abundant fluid
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44 inclusion trails extending outward from the polymineralic inclusions could be the result of the
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46 accompanying exsolution of CO₂. General exsolution of a vapor phase (if H₂O is included in the system)
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48 may further be responsible for the crystallization of phlogopite and serpentine inside polymineralic
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50 inclusions, and/or in the surrounding micro-inclusions (e.g., Fig. 4).
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54 Polymineralic inclusions in Cr-pyrope also show evidence for extensive chemical interaction with the
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56 host (as evident from kelyphite zones), but commonly contain dolomite. **As there is no experimental**
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4 evidence for an analogous reaction between dolomite melt and grt to produce calcite, it seems logical that
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6 the dolomite preserved in polymineralic inclusions in Cr-pyrope could be pristine in nature.
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9 Process and Timing of Melt Entrapment

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11 A likely process for entrapment of a melt in mantle minerals is described as “necking down” by Roedder
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13 (1984) and has been proposed in previous studies on polymineralic inclusions in megacrysts (Schulze
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15 1985; van Achterbergh et al. 2002; van Achterbergh et al. 2004; Araújo et al. 2009). In this process, a
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17 melt enters the host crystal along fractures and cleavage planes by dissolution and recrystallization.
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19 Surface reduction then results in the entrapment of discrete inclusions that may coalesce to form larger,
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21 rounded inclusions. Hence, reaction and the production of new minerals are an integral part of the necking
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23 down process. This process of melt infiltration may be further aided by crack formation due to rapid
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25 decompression, as described for the formation of fractures in kimberlitic olivine lined with carbonate
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27 (Brett et al. 2015).
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33 Thermobarometry of the host Cr-diopsides indicates that they equilibrated at pressures of 4 - 5 GPa (~130
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35 - 160 km), deep in the lithospheric mantle (Table 1). The preservation of polymineralic inclusions and the
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37 striking compositional zoning around them, such as spongy rims in Cr-diopside, suggest that the melt
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39 infiltration occurred shortly prior to or during kimberlite ascent. Otherwise the compositional zoning
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41 would have equilibrated quickly at their original deep lithospheric mantle temperature conditions (e.g.,
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43 Schulze 1985). Similarly, the fine-grained texture of the kelyphite zones around inclusions within Cr-
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45 pyrope documents a rapid process during kimberlite ascent. In fact, dissolution experiments of grt
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47 xenocrysts in kimberlite melt suggest that this process occurs on a time scale of minutes (Canil and
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49 Fedortchouk 1999). Moreover, the decarbonation reaction proposed to account for the calcite-olivine(-
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51 plogopite) assemblage of the inclusions within Cr-diopsides should take place at pressures < 2.5 GPa,
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53 consistent with reaction during kimberlite ascent.
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Compositional Link to the Transporting Kimberlite Magma

Detailed investigation of individual mineral phases within polymineralic inclusions has shown that their compositions are similar to those crystallizing from kimberlite magma. Moreover, the phase compositions lie at the beginning of kimberlite mineral differentiation trends, suggesting that an early stage of kimberlite melt was trapped at mantle pressures. Particularly the evolution of spinels along magmatic trend 2 of the kimberlite compositional array (Fig. 8), and olivines falling along the megacryst trend (Fig. 5), suggests that polymineralic inclusions in both Cr-diopsides and Cr-pyropes from LDG kimberlites represent snapshots of kimberlite melt reacting with the lithospheric mantle during ascent. This model is further supported by the clear difference in the trace element compositions of phlogopite within the polymineralic inclusions versus groundmass phlogopite typically found in kimberlite (Fig. 6d-f). The phlogopite within the inclusions shows a striking similarity to high Ti-Cr phlogopite zones interpreted to have crystallized at depth (Giuliani et al. 2016).

No significant differences in mineral compositions exist between ‘carbonate-rich’ and ‘silicate-rich’ inclusion types, or between samples from different kimberlite pipes. Instead, mineral compositions appear to be controlled mainly by reaction and equilibration with the host megacrysts. Extensive chemical exchange between the invading melt and the host megacrysts is evident from the enrichment of individual inclusion phases in components such as Cr_2O_3 , Al_2O_3 , FeO , and MnO , in relation to the chemical character of their hosts; i.e. greater enrichment of these elements in Cr-pyropes compared to Cr-diopsides. In Cr-pyropes, elemental exchange may even lead to the stabilization of additional phases that are not typical of kimberlite, such as Al-rich cpx and Al-spinel by Al exchange.

The continuous range of bulk compositions from ‘carbonate-rich’ to ‘silicate-rich’ inclusions (Fig. 11) led van Achterbergh et al. (2002) to propose separate origins for the two inclusion types. However, we posit that the range in inclusion mineralogy and hence bulk compositions can be explained by a combination of: 1) The way in which the inclusions are exposed during sectioning will lead to random sampling of an assemblage dominated by carbonate and silicates (olivine, phlogopite, etc.), yielding the nearly

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4 **continuous** range in bulk compositions illustrated well by CaO-SiO₂ and MgO-SiO₂ relations (Fig. 11a,
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6 b). 2) Small-scale fractional crystallization of the melt in a small fixed volume will produce different
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8 compositions **that** are then exposed in two-dimensional sections. **This can explain observations** such as
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10 the two generations of calcite in the inclusions, with the later generation being more enriched in
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12 incompatible elements such as Sr (evident as bright zones in BSE images). 3) The polymineraleic
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14 inclusions represent trapped melts at various stages of **reaction and differentiation** prior to entrapment.
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16 Evidence for differentiation of the melt prior to entrapment comes from the range of olivine compositions
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18 seen in different inclusions that define a typical igneous fractionation trend (Fig. 5), as documented
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20 originally in olivine megacrysts by Boyd (1974). Fractionation is also clearly evident in the range of
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22 spinel compositions (Fig. 8). **Differentiation is likely to be associated with the progressive assimilation of**
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24 **mantle material, such as opx (Kamenetsky et al. 2008; Russell et al. 2012; Pilbeam et al. 2013; Brett et al.**
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26 **2015) and cpx – as observed here.**

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31 **Because bulk compositions are easily skewed by a combination of the above processes (in addition to**
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33 **reaction with the host), the Mg# of the reconstructed bulk compositions cannot be reliably compared to**
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35 **the expected range of Mg# for LDG kimberlite liquids (Canil and Bellis 2008). Instead, we think it is**
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37 **more instructive to examine the calculated melts in equilibrium with olivine inside the polymineraleic**
38
39 **inclusions. Using the equation by Canil and Bellis (2008) along with their lower and upper estimates for**
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41 **$K_{d_{FeMg}}^{ol/liq}$ (0.12 and 0.27) we arrive at average Mg# for all polymineraleic inclusions of 0.50 and 0.69,**
42
43 **respectively. Olivines within inclusions in cpx yield higher liquid Mg# (0.52 and 0.71, respectively) than**
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45 **those in inclusions in grt (0.47 and 0.66, respectively), as expected from the higher Mg# of the host cpx**
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47 **relative to grt. These estimates of liquid Mg# overlap those by Canil and Bellis (2008) for LDG**
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49 **kimberlites (mostly < 0.60) which we interpret as further evidence that the polymineraleic inclusions in**
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51 **cpx and grt megacrysts are linked to the early evolutionary stage of the transporting kimberlite magma.**
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A Glimpse at Primary Kimberlite Melt?

The reconstructed bulk compositions of polymineralic inclusions in Cr-diopside fall along a distinct trend in CaO-SiO₂ and MgO-SiO₂ space, produced by a combination of the three mechanisms described above (cross-section effect, internal fractional crystallization, and melt differentiation prior to entrapment), which essentially describes a mixing line between the end-member inclusion phases calcite and olivine (Fig. 12). The compositions of polymineralic inclusions in Cr-pyrope generally also fall along this trend, although they are more enriched in SiO₂, FeO, Cr₂O₃, and Al₂O₃ due to equilibration with the host (Fig. 11c, d).

The clear evidence of reactions between melt and host crystals makes an *original melt* composition difficult to obtain. The only possible result, using the inclusions themselves, is to arrive at an estimate for the composition of the melt after it has experienced reaction with the host. This composition will, necessarily, be subject to large uncertainties. Based on the observation that inclusions within Cr-pyrope have been more influenced by later equilibration with the host and noting that the reconstructed bulk compositions represent random samples of melt compositions, we use the average of the range of bulk compositions of inclusions in Cr-diopsides to obtain an average bulk composition. The resulting composition, which represents a melt *after* reaction with the host, has ~22 wt.% SiO₂, ~24 wt.% CaO and ~15 wt.% MgO (star symbol in Fig. 11, 12). This composition is compared to experimental melts and primary kimberlite magma estimates in order to explore possible precursor melts (Fig. 12).

Partial melts of carbonated peridotite produced in different experimental studies (at T ≤1500°C) generally lie at lower SiO₂ contents. However, a correlation of SiO₂ with T is observed in these experiments, so that the high T (1500°C) runs, e.g., by Gudfinnsson and Presnall (2005), approach our estimate of the average melt composition. These experiments are ~500°C above the equilibration temperature of the Cr-diopside megacrysts (Table 1), which seems excessively high. Alternatively, the experimental melts can be shifted onto the polymineralic inclusion array by reaction with the host crystals. In Fig. 12, this is demonstrated with reaction vectors towards cpx and grt compositions extending from one possible primary composition

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4 characterized by maximum overlap of the experimental fields at melting conditions of roughly 6 GPa and
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6 1400°C (question mark in Fig. 12). The digestion of other mantle silicates, especially opx, causes a
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8 similar displacement and may have started before the melt became trapped in the Cr-diopside and Cr-
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10 pyrope hosts (Kamenetsky et al. 2008; Russell et al. 2012; Pilbeam et al. 2013; Brett et al. 2015). Recent
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12 experimental studies, using carbonatite compositions obtained from previous low-degree partial melting
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14 experiments on mantle peridotite, suggest that opx dissolution does not occur until the kimberlite magma
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16 reaches pressures < 3.5 GPa (Stone and Luth 2016, and references therein). An indication that opx
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18 dissolution may be an important process during final (< 100 km) ascent and phenocryst crystallization of
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20 kimberlite magma – as opposed to the melt trapped as inclusions in cpx and grt megacrysts – lies in the
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22 mineral chemistry of olivine: Whereas olivines within the polymineralic inclusions define a fractionation
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24 trend (Fig. 5a), phenocrystic rims on olivines in kimberlite show a characteristic trend of decreasing Ni at
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26 buffered Mg# (arrow in Fig 5a) which has previously been attributed to the effect of opx dissolution (e.g.,
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28 Pilbeam et al. 2013; Bussweiler et al. 2015).

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34 Previous estimates of primary or parent kimberlite magma obtained through whole rock considerations
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36 (Le Roex et al. 2003; Kjarsgaard et al. 2009) also plot along the trend of reconstructed bulk compositions
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38 of polymineralic inclusions (Fig. 12). Compared to our average bulk composition, they are characterized
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40 by lower CaO and higher MgO and SiO₂ contents, which may suggest that these magma compositions, as
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42 well, have experienced some degree of mantle assimilation.

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45 While the polymineralic inclusions constitute snapshots of kimberlite melt reacting with mantle minerals
46
47 during ascent, it is important to note that they do not represent the primary or protokimberlite melt.
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49 Evidence for such precursor melts to LDG kimberlites may be preserved as melt inclusions in phenocryst
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51 phases (Kamenetsky et al. 2013), or as fluid inclusions in fibrous diamonds (Klein-BenDavid et al. 2007;
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53 Weiss et al. 2015), or can be gleaned from isotopic studies (Tappe et al. 2013). Together with the
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55 occurrence of sublithospheric diamonds at LDG (e.g., Tappert et al. 2005) these studies point towards an
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57 asthenospheric origin of the protokimberlite. The generally SiO₂-poor and CaO-rich compositions of the
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4 already reacted kimberlite melt, as represented by the now crystallized polymineralic inclusions, is
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6 consistent with suggestions of the carbonatite-like nature of the protokimberlite melt (e.g. Dawson 1971;
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8 Dawson and Hawthorne 1973; Russell et al. 2012; Kamenetsky and Yaxley 2015; Kamenetsky 2016).
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10 11 General Role of Decarbonation Reactions in Producing Calcite-Bearing Kimberlites 12 13

14 Oxybarometry studies applied to cratonic grt peridotites have shown that the lithospheric mantle below
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16 the LDG kimberlite field of the central Slave Craton tends to be more oxidized than the mantle below
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18 other cratons (Creighton et al. 2010; Luth and Stachel 2014), especially in the pressure-range of 4.5 to 6
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20 GPa (~140 – 180 km), corresponding roughly to the depth of formation of the host megacrysts (Table 1;
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22 Supplementary Fig. 3). This has been attributed to the oxidizing effects of carbonate-rich fluids/melts
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24 (e.g., Creighton et al. 2008). The result of this metasomatism is the oxidation of local zones in the mid- to
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26 lowermost lithosphere beneath the LDG kimberlite field to levels conducive to the stabilization of
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28 carbonate. The expected stable carbonate phase is Mg-rich in any four-phase peridotite assemblage, i.e.
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30 dolomite (e.g., Wyllie and Huang 1975; Dalton and Presnall 1998a) or magnesite at higher pressure (Brey
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32 et al. 1983), and the melt produced from such carbonated peridotites is always dolomitic in composition
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34 (e.g., Irving and Wyllie 1975; Brey et al. 2008). However, LDG kimberlites, and kimberlites globally, are
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36 mostly dominated by calcite (e.g., Skinner and Clement 1979; Armstrong et al. 2004), as are the early
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38 kimberlite melts trapped as polymineralic inclusions in LDG megacrysts.
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44 The reactions illustrated by the polymineralic inclusions provide an explanation for this discrepancy
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46 between expected high pressure melt compositions produced from carbonated peridotite and the calcite-
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48 rich nature of kimberlites in general, via the decarbonation reaction:
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54 This reaction is expected to begin during kimberlite ascent at depths around 70 km based on experimental
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56 and theoretical constraints (Wyllie and Huang 1975; Stone and Luth 2016; Stone 2016). The reaction may
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58 continue up to kimberlite emplacement in the crust, as indicated by the presence of reaction rinds (often
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4 containing calcite segregations) around single Cr-diopside megacrysts in their host kimberlite (Fig. 2a, b).
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6 The latter observation also indicates that the dolomite-diopside decarbonation reaction is not just relevant
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8 for melt differentiation within the limited volume of polymineralic inclusions, but affects the entire rising
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10 batch of kimberlite magma. The similarity of the phase composition variations present in the kimberlite
11
12 melt inclusions with those observed in kimberlites emplaced in the crust, indicates that this process of
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14 melt-rock reaction is critical in producing the observed compositions and mineralogy of kimberlites
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16 worldwide. In this scheme, the resulting exsolution of a vapor phase during the proposed reaction may be
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18 responsible for the crystallization of phlogopite and serpentine in the inclusions (and in the host
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20 kimberlite), as water may be present in the original melt and will accumulate during reaction and
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22 crystallization. In addition to driving the evolution of a dolomitic to a calcitic kimberlite melt at pressures
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24 in the shallower portion of the lithospheric mantle, the reaction provides a means of further CO₂
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26 exsolution, which is seen as an important process in driving kimberlite emplacement (e.g., Eggler 1986;
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28 Brey et al. 1991; Russell et al. 2012). The reaction could proceed as long as the kimberlite magma
29
30 contains a dolomitic melt component that can react with disaggregated diopside (either as Cr-diopside
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32 megacrysts or from peridotite xenoliths). While the abundance of diopside in lithospheric peridotites is
33
34 relatively low (~10%), the presence of lherzolites and discrete Cr-diopside megacrysts in kimberlites is
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36 ubiquitous, indicating the likelihood of this reaction occurring in any section of cratonic lithosphere.
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43 A schematic illustration of the formation of polymineralic inclusions and the concomitant evolution of the
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45 host kimberlite is summarized in Fig. 13. In sequence, the process is envisioned as (1) the formation of a
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47 dolomitic silico-carbonatite melt via melting of carbonated peridotite, (2) initial reaction with lithospheric
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49 mantle and early high pressure CO₂ exsolution, (3) lower pressure reactions with lithospheric wall rocks
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51 that change the bulk composition and release more CO₂, producing calcite, (4) the emplacement in the
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53 crust of the evolved kimberlite with the calcite-bearing mineral assemblage. The widespread occurrence
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55 of polymineralic inclusions in kimberlite megacrysts from numerous cratons (Haggerty and Boyd 1975;
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4 Schulze 1985; van Achterbergh et al. 2002; Pivin et al. 2009; Araújo et al. 2009) is evidence of the
5
6 general applicability of this process to the evolution of kimberlites.
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10 Conclusions

- 13 1. Lac de Gras Cr-diopside and Cr-pyrope megacrysts contain abundant, large (up to 2 mm)
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15 polymineralic inclusions formed by necking down of melts starting at lithospheric mantle depths.
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17 The megacrysts genetically are mantle xenocrysts with peridotitic chemical signatures.
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19 Geothermobarometry suggests that they originate from within the lithospheric mantle (~4.6 GPa,
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21 1015°C).
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- 24 2. There is clear textural and isotopic evidence for disequilibrium between the host crystals and their
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26 polymineralic inclusions, the preservation of which establishes a temporal link to the kimberlite
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28 eruption.
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- 31 3. The inclusion phases are broadly characteristic of kimberlite, with their compositions being
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33 overall consistent with the early stages of kimberlite differentiation trends. Enrichment of
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35 inclusion phases in components more abundant in the host crystals, such as Cr₂O₃ and Al₂O₃,
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37 points towards extensive reaction between inclusions and hosts. Trace element signatures of
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39 phlogopites within the inclusions constrain their crystallization from the melt inclusions at
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41 lithospheric mantle depths.
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- 44 4. Although scattered by sectioning effects and reaction with the host phases, reconstructed bulk
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46 compositions of the inclusions bear a general resemblance to the experimentally derived partial
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48 melts of carbonated peridotites, if allowance is made for the assimilation of mantle minerals
49
50 (including the host minerals cpx and grt). Thus, the polymineralic inclusions represent snapshots
51
52 of kimberlite melt reacting with the lithospheric mantle during ascent.
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- 55 5. Inclusions in Cr-diopside record direct evidence for a decarbonation reaction of the form:
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57 dolomitic melt + diopside → forsterite + calcite + CO₂, expected to begin at pressures < 2.5 GPa.
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This reaction may play an important role in the en-route transformation of an upward moving dolomitic silico-carbonatite melt into the calcite-bearing kimberlite observed at the surface. As such, it is one of many reactions likely to occur between carbonatitic melt and lithospheric mantle that drive the chemical and volatile evolution in kimberlite.

Figure Captions

Fig. 1 Geological overview map of the Slave Craton indicating the location of major diamond mines, modified from Bleeker et al. (2004). Dashed line indicates inferred suture based on Pb isotopic studies (Davis et al. 1996).

Fig. 2 a) Section through kimberlite drill core exposing cpx megacryst with brownish reaction rind. b) Thin section of cpx megacryst in direct contact with calcite segregations in coherent (hypabyssal) kimberlite from the A154N pipe, Diavik. c) Thin section of cpx megacryst with abundant polyminerale inclusions and remnant host kimberlite ('kimberlite rind'). d) Photomicrograph of a polyminerale inclusion in cpx megacryst (courtesy of V.S. Kamenetsky). Polyminerale inclusions are typically located where fractures/veins with fluid inclusions converge and are typically surrounded by a thick reaction rim (spongy rim). e) Sample mount section of grt megacrysts from < 1 cm concentrates from Diavik. Color range indicates different origins. Polyminerale inclusions are most common in red grt grains (Iherzolitc or megacrystic). f) Back-scattered electron (BSE) image of a polyminerale inclusion in a grt megacryst. Veins connect polyminerale inclusion to the host kimberlite, but mineralogy of inclusion and host kimberlite is markedly different.

Fig. 3 BSE images for polyminerale inclusions in Cr-diopside megacrysts (a-c) and in Cr-pyrope (grt) megacrysts (d-f). All inclusions in cpx are surrounded by spongy 'reaction rims', whereas inclusions in grt are surrounded by 'kelyphite zones'. a) 'Carbonate-rich' end-member inclusion with calcite (cc) groundmass composed of bright and dark zones, containing phlogopite (phl), olivine (ol), serpentine (srp), and chromite (chr). b) Intermediate inclusion with 'colloform' cc in Mg-Fe silicate groundmass, containing phl and chr. c) 'Silicate-rich' end-member inclusion with Mg-Fe-silicate groundmass composed of srp/chlorite (with distinctive vermicular texture), containing cc blebs and chr. d) 'Carbonate-rich' end-member inclusion with cc groundmass, containing phl, ol, srp, and chr. e) Intermediate inclusion with Mg-Fe silicate groundmass (vermicular texture) containing dolomite (dol), spinel (spl), and

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4 cpx in addition to phl and chr. f) ‘Silicate-rich’ end-member inclusion with Mg-Fe-silicate groundmass
5 containing cc as irregular blebs, phl, and chr.
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9 **Fig. 4** EPMA element distribution maps for Ca, Al, Na, Mg, K, and Sr of a polymineralic inclusion
10 (intermediate type) in a cpx megacryst. Warmer colors indicate higher element concentrations. Reaction
11 rims are enriched in Ca, and depleted in Al and Na. The maps highlight the presence of phlogopite (phl),
12 calcite (cc), and serpentine (srp) in micro-inclusions around the main inclusion.
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19 **Fig. 5** Bivariate plots for olivine in polymineralic inclusions resolved by megacryst host (Cr-diopside and
20 Cr-pyrope), and olivine in kimberlite rinds (a-c) and as mineral inclusions and intergrowths (d). Olivine
21 was analyzed in eight polymineralic inclusions in Cr-diopside from Diavik and Point Lake (seven
22 ‘carbonate-rich’, one ‘silicate-rich’), and in four polymineralic inclusions in Cr-pyrope from Diavik (two
23 ‘carbonate-rich’, two ‘silicate-rich’). 12 olivine mineral inclusions were measured in Cr-pyropes from
24 Diavik, one in Cr-pyrope from Point Lake, and one in Cr-diopside from Diavik. Reference data for LDG
25 kimberlite olivines divided into cores and rims are from Brett et al. (2009) and Bussweiler et al. (2015).
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34 **The arrow in (a) highlights the differentiation trend of phenocrystic rims (see text).** Data of olivine
35 inclusions in diamond from Lac de Gras, Slave Craton, Canada, are from Donnelly et al. (2007) and
36 Stachel et al. (2003).
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42 **Fig. 6 A-C:** Major element (EPMA data) bivariate plots for phlogopite in polymineralic inclusions
43 resolved by megacryst host (Cr-diopside and Cr-pyrope). Phlogopite was analyzed in polymineralic
44 inclusions in Cr-diopside from Diavik and Point Lake (five ‘carbonate-rich’, 19 ‘silicate-rich’), and in
45 polymineralic inclusions in Cr-pyrope from Diavik, Point Lake, and Wolverine (four ‘carbonate-rich’, 29
46 ‘silicate-rich’). Reference data for phlogopite in kimberlites are from Eccles et al. (2004), Armstrong et
47 al. (2004), and Kopylova et al. (2010). Mantle phlogopite data are from Menzies et al. (2004) and Giuliani
48 et al. (2014). **D-F: Trace element (LA-ICP-MS data) bivariate plots for phlogopite in polymineralic**
49 **inclusions. The inset in D shows compositional fields for global carbonatite- and kimberlite-derived**
50 **phlogopite (Requir et al. 2009). Phlogopite in polymineralic inclusions clearly falls into the kimberlite**
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4 field. Compositional fields for groundmass phlogopite and high Ti-Cr phlogopite cores are based on data
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6 from Giuliani et al. (2016). Phlogopite in polymineralic inclusions is similar to the high Ti-Cr group
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8 (crystallized at depth) in many trace elements.
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11 **Fig. 7** Bivariate plots for carbonates resolved by megacryst host (Cr-diopside and Cr-pyrope). Carbonate
12 was analyzed in polymineralic inclusions in Cr-diopside from Diavik (10 ‘carbonate-rich’, 26 ‘silicate-
13 rich’), Point Lake (four ‘carbonate-rich’, five ‘silicate-rich’), and in polymineralic inclusions in Cr-pyrope
14 from Diavik (two ‘carbonate-rich’, nine ‘silicate-rich’) and Point Lake (three ‘carbonate-rich’, 11 ‘silicate-
15 rich’). Carbonate was further analyzed in veins, kimberlite rinds, and altered olivine inclusions (in Cr-
16 pyrope). Reference data for kimberlitic carbonate are from Armstrong et al. (2004).
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26 **Fig. 8** Bivariate plots for chromite/spinel in polymineralic inclusions resolved by megacryst host (Cr-
27 diopside and Cr-pyrope) and in kimberlite rinds. Chromite/spinel was analyzed in inclusions in Cr-
28 diopside from Diavik (six ‘carbonate-rich’, 21 ‘silicate-rich’) and Point Lake (one ‘silicate-rich’), and in
29 inclusions in Cr-pyrope from Diavik (one ‘carbonate-rich’, 13 ‘silicate-rich’), Point Lake (one ‘carbonate-
30 rich’, 19 ‘silicate-rich’) and Wolverine (four ‘silicate-rich’). In addition, chromite was analyzed in veins,
31 kimberlite rinds, and around altered mineral inclusions. Reference data for spinel in LDG kimberlites are
32 from Armstrong et al. (2004) and Roeder and Schulze (2008), for spinel in Jericho mantle xenoliths
33 (peridotite/pyroxenite) from Kopylova et al. (1999). Shaded field represents magmatic spinel trend in
34 kimberlitic chromites from Mitchell (1986).
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47 **Fig. 9** Bivariate plots for major and minor elements of cpx in polymineralic inclusions in Cr-pyrope. Data
48 are compared against Cr-diopside hosts and spongy reaction rims (around polymineralic inclusions) from
49 this study, and against megacrysts worldwide (Eggler et al. 1979; Hunter and Taylor 1984; de Bruin
50 2005; Pivin et al. 2009).
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57 **Fig. 10** $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for ‘carbonate-rich’ inclusions and their Cr-diopside and Cr-pyrope hosts. Data
58 points marked with * are previous analyses from van Achtebergh et al. (2002). Reference data for calcite
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4 in the Jos kimberlite, Somerset Island, Nunavut, Canada are from Malarkey et al. (2010). LDG kimberlite
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6 whole rock data are from Tappe et al. (2013). LDG perovskite data are from Sarkar et al. (2015).
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9 **Fig. 11** Reconstructed bulk compositions of polymineralic inclusions resolved by megacryst host (Cr-
10 diopside and Cr-pyrope). Inclusions in Cr-diopside are further resolved by end-member type based on
11 carbonate-rich or silicate-rich groundmass. The star symbol represents an estimate of the average bulk
12 composition as an average of all inclusions hosted in Cr-diopside (see Discussion).
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18 **Fig. 12** Schematic diagram relating the observed range of reconstructed bulk compositions of
19 polymineralic inclusions (dashed, grey, double-arrow field) to partial melts of carbonated peridotite
20 produced in high-P-T experiments, by reaction with the host phases Cr-diopside (cpx) and Cr-pyrope
21 (grt), and to previous estimates of primary kimberlite magma based on whole rock geochemistry of
22 hypabyssal kimberlites from Kimberley, South Africa (Le Roex et al. 2003) and from Lac de Gras
23 (Kjarsgaard et al. 2009). Only experimental melts produced at $T \leq 1500^\circ\text{C}$ are included in the fields, so
24 that experimental conditions for the different melts are as follows, Gudfinnsson and Presnall (2005): 3.2-
25 5.9 GPa, 1340-1500°C; Dalton and Presnall (1998a): 3-7 GPa, 1245-1430°C; Dalton and Presnall
26 (1998b): 6 GPa, 1380-1480°C; Brey et al. (2008): 6-10 GPa, 1300-1500°C; Foley et al. (2009): 4-6 GPa,
27 1090-1290°C.
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42 **Fig. 13** Schematic model of formation for polymineralic inclusions in Cr-diopside and Cr-pyrope
43 megacrysts and the concomitant evolution of the host kimberlite. 1) Formation of a dolomitic silico-
44 carbonatite melt via partial melting of carbonated peridotite close to the base of the lithosphere. 2)
45 **Ascending** kimberlite melt infiltrates megacrysts after interaction with other mantle minerals (above 150
46 km) **and is trapped** by process of necking down (Roedder 1984) and/or decompression cracking (Brett et
47 al. 2015). 3) Decarbonation reactions cause chemical exchange between inclusions and hosts during
48 kimberlite ascent, forming spongy rims around inclusions in Cr-diopside, whereas kelyphite zones around
49 inclusions in Cr-pyrope form by decompression reactions. In Cr-diopside, all Mg-carbonate is
50 transformed to calcite, whereas primary dolomite may be preserved in inclusions in Cr-pyrope. Some
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4 trails of fluid inclusions and veins form outwards due to the release of CO₂. Importantly, the reaction also
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6 occurs around the outside of the megacryst hosts, thereby releasing calcite and CO₂ into the transporting
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8 kimberlite. 4) Upon kimberlite emplacement globules have fully crystallized to polymineralic inclusions.
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10 ‘Carbonate-rich’ and ‘silicate-rich’ zones, formed by internal fractional crystallization within the
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12 inclusions, are exposed in two-dimensional sections and create the impression of compositional
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14 bimodality. Reaction rims and calcite segregations around Cr-diopsides and kelyphite rims around Cr-
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16 pyropes may suggest that they react with the host kimberlite until emplacement.
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20 **Supplementary Fig. 1** QEMSCAN® maps of polymineralic inclusions in Cr-diopside (a = PL_CPX_03
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22 In01; b = PL_CPX_03 In10) and Cr-pyrope (c = PL_GRT_04 In13; d = PL_GRT_04 In05). Inclusions a)
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24 and c) are of the ‘carbonate-rich’, and b) and d) of the ‘silicate-rich’ end-member type. Modal proportions
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26 of the inclusions as obtained with QEMSCAN® are as follows: a) 10.3% ol; 11.4% srp; 11.2% phl; 65.8%
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28 cc; 0.1% ap. b) 4.0% ol; 60.4% srp; 16.5% phl; 15.2% cc; 0.1% ap. c) 8.2% ol; 0.2% cpx; 15.0% srp;
29
30 30.2% phl; 3.7% spl; 40.7% cc; 0.9% dol; 0.1% py. d) 0.8% ol; 2.0% cpx; 45.7% srp; 31.1% phl; 6.2%
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32 spl; 0.1% cc; 6.0% dol; 0.1% ap; 0.1% py. Mineral abbreviations are as follows: ol = olivine; cpx =
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34 clinopyroxene; srp = serpentine; phl = phlogopite; spl = spinel; cc = calcite; dol = dolomite; ap = apatite;
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36 py = pyrite.
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41 **Supplementary Fig. 2** Bivariate plots for major and minor elements in serpentine/chlorite in
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43 polymineralic inclusions resolved by megacryst host (Cr-diopside and Cr-pyrope) and in altered olivine
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45 mineral inclusions in Cr-pyrope. Reference data for kimberlitic serpentine are from Hayman et al. (2009)
46
47 and Mitchell (1986).
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51 **Supplementary Fig. 3** $\Delta\log f_{O_2}$ (FMQ) values for grt peridotites from different cratons (modified from
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53 Luth and Stachel 2014). Samples from the central Slave Craton (Creighton et al. 2010) are notably more
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55 oxidized than those from other cratons.
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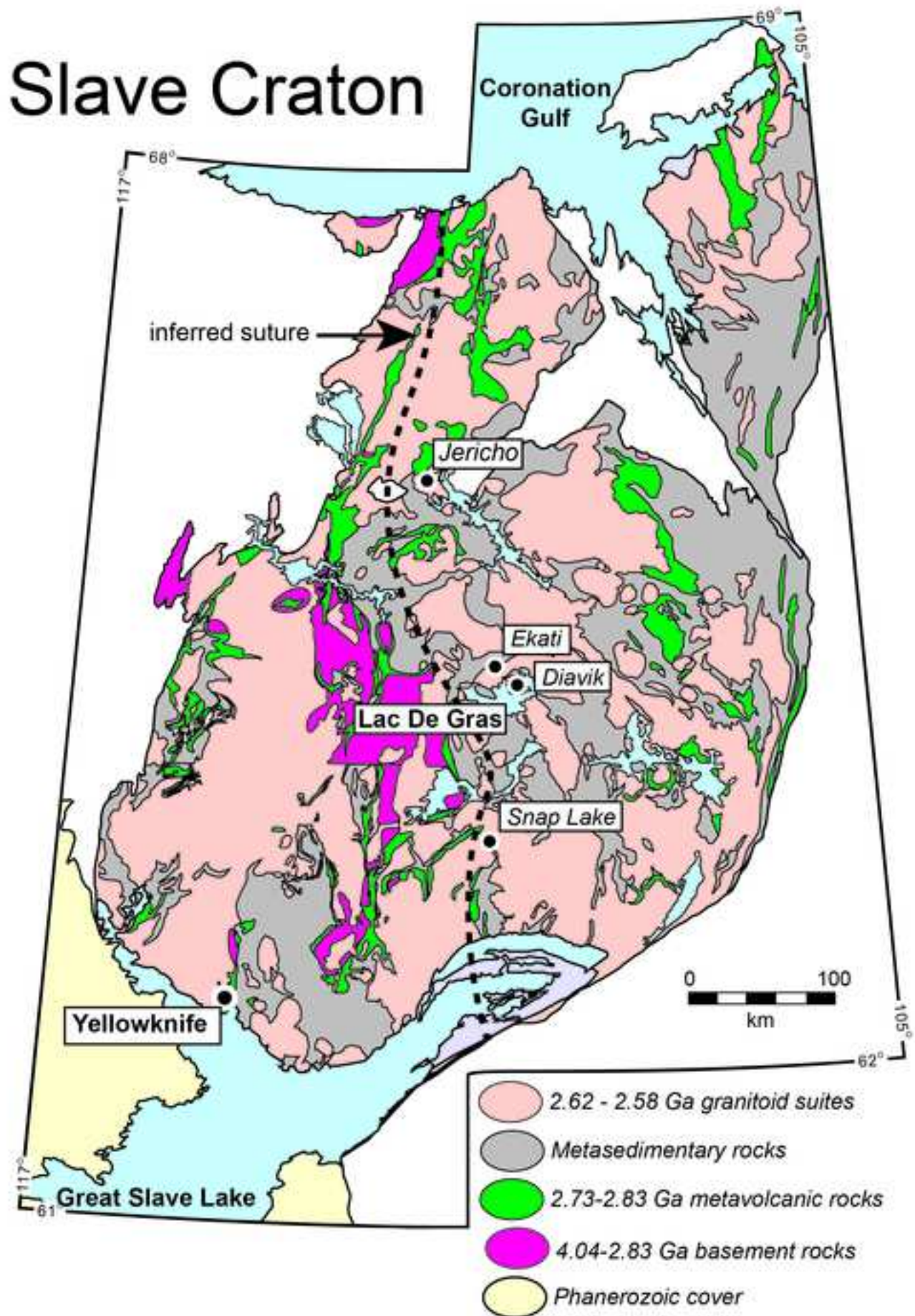
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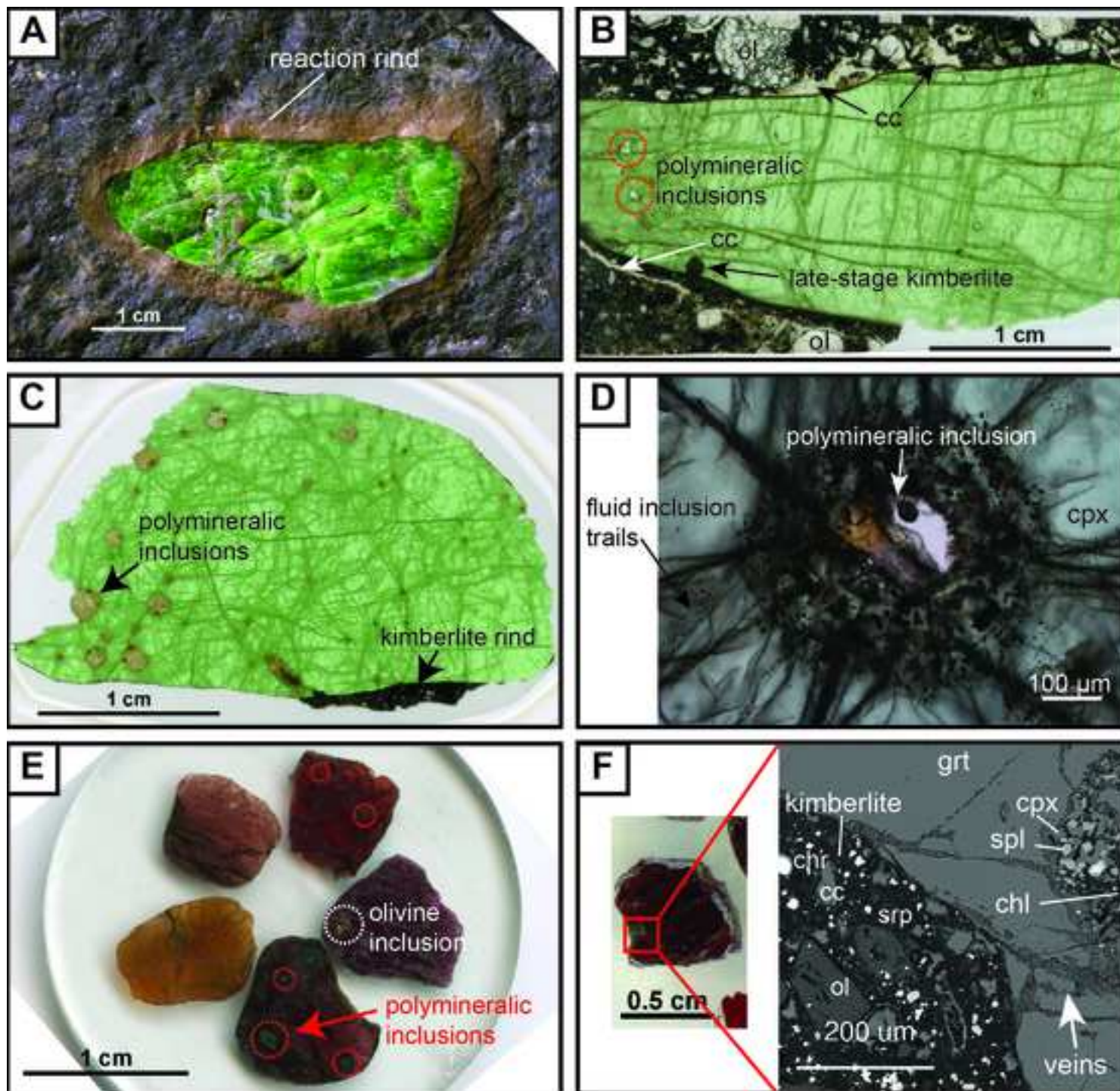
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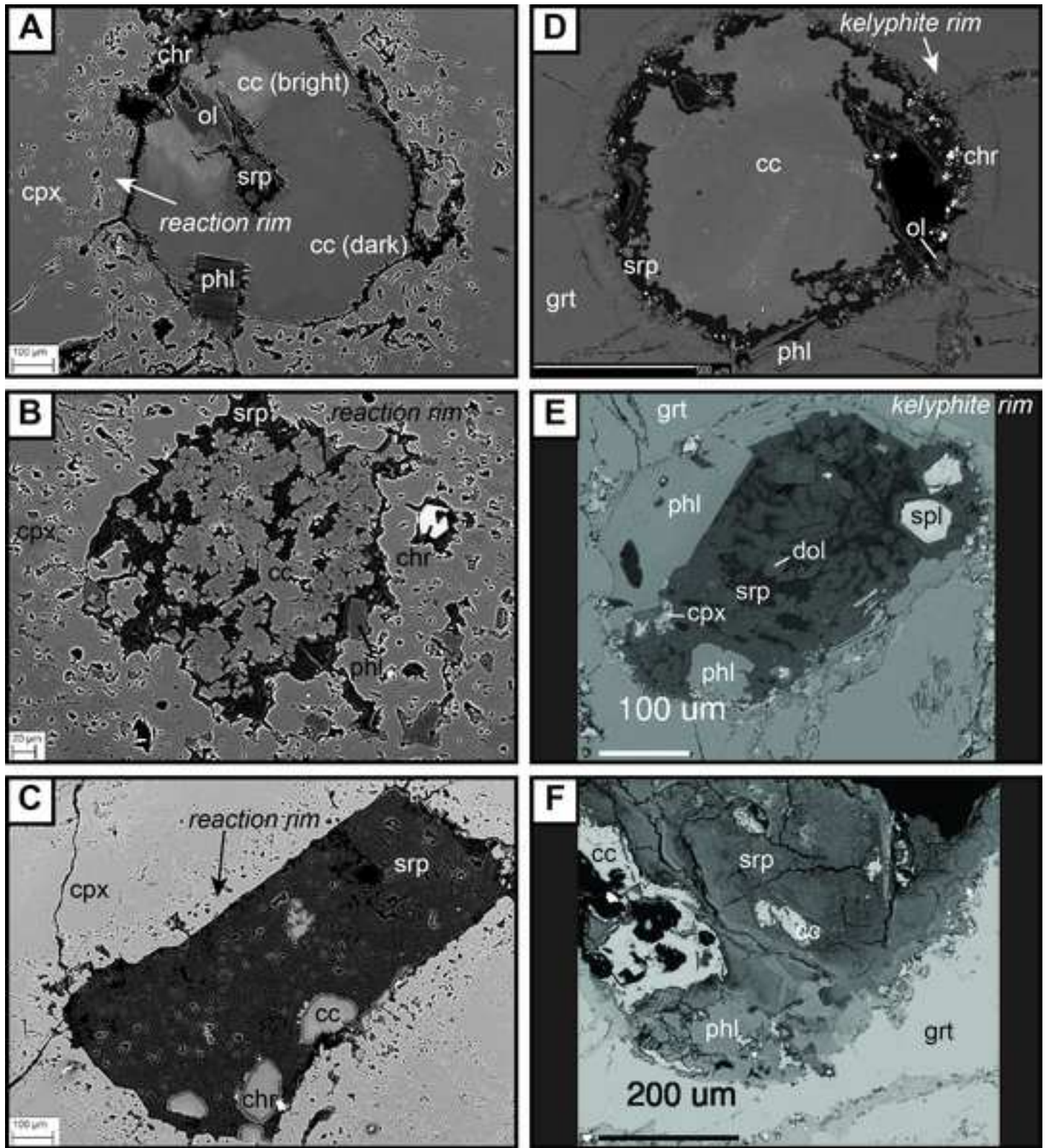
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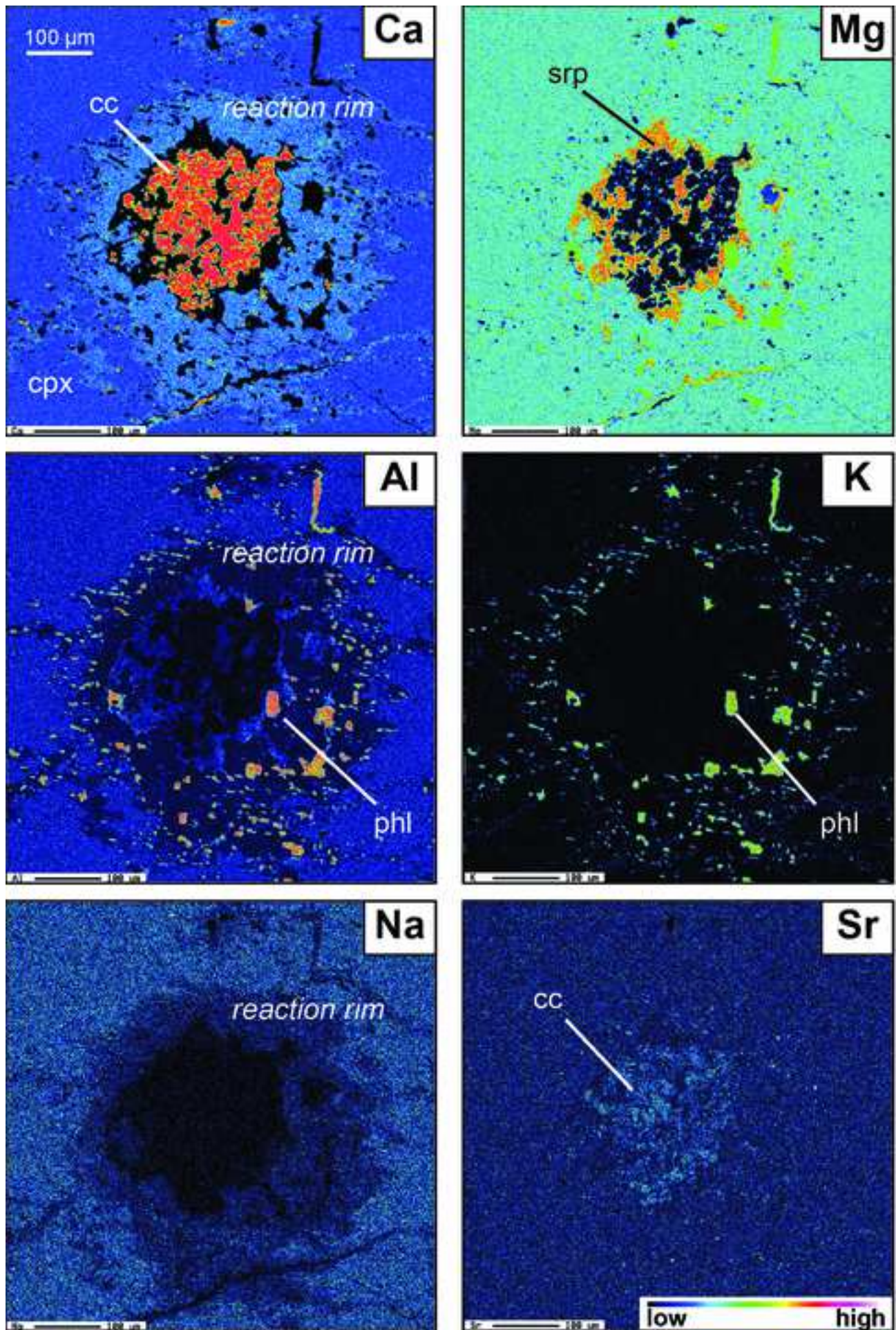
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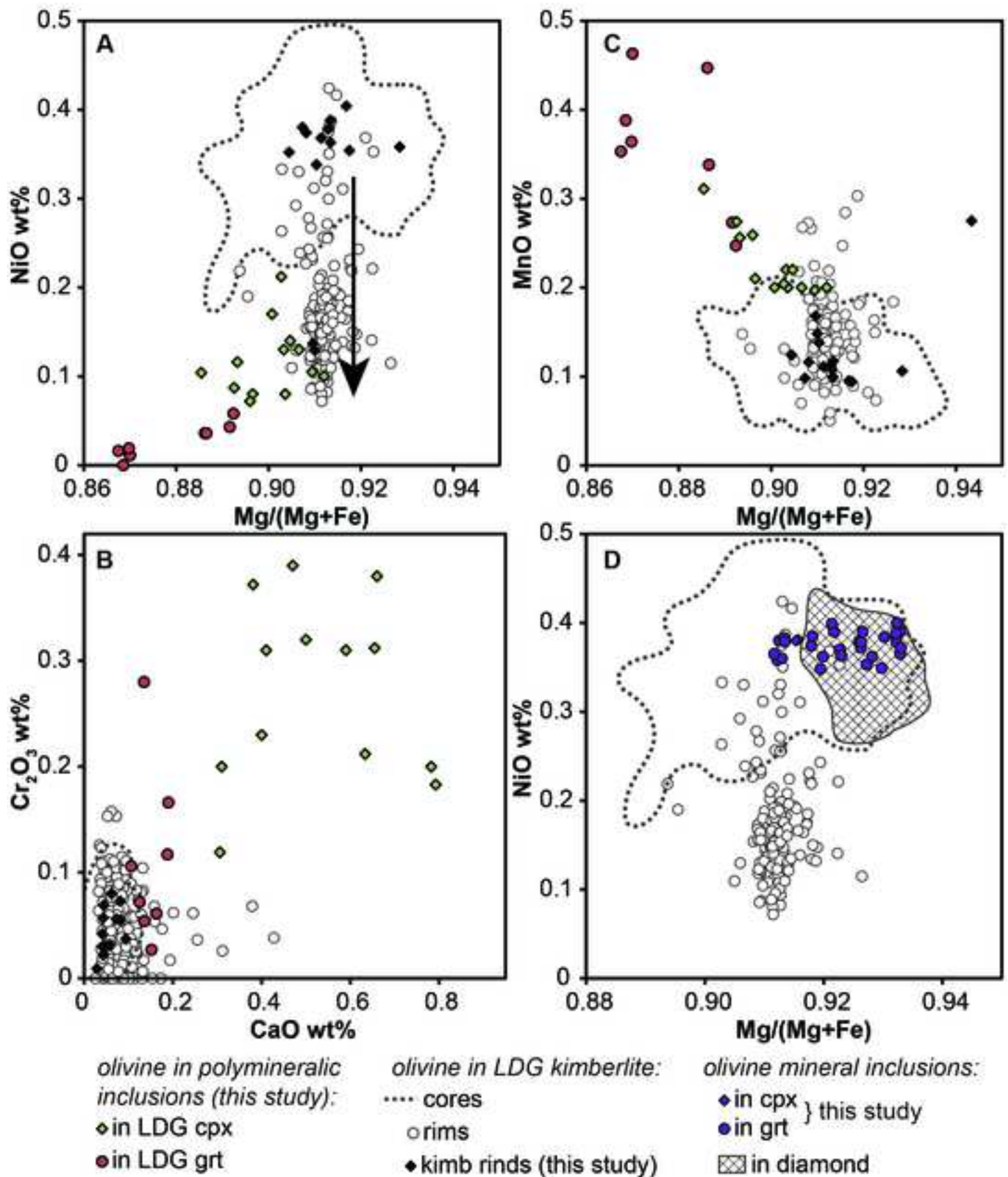
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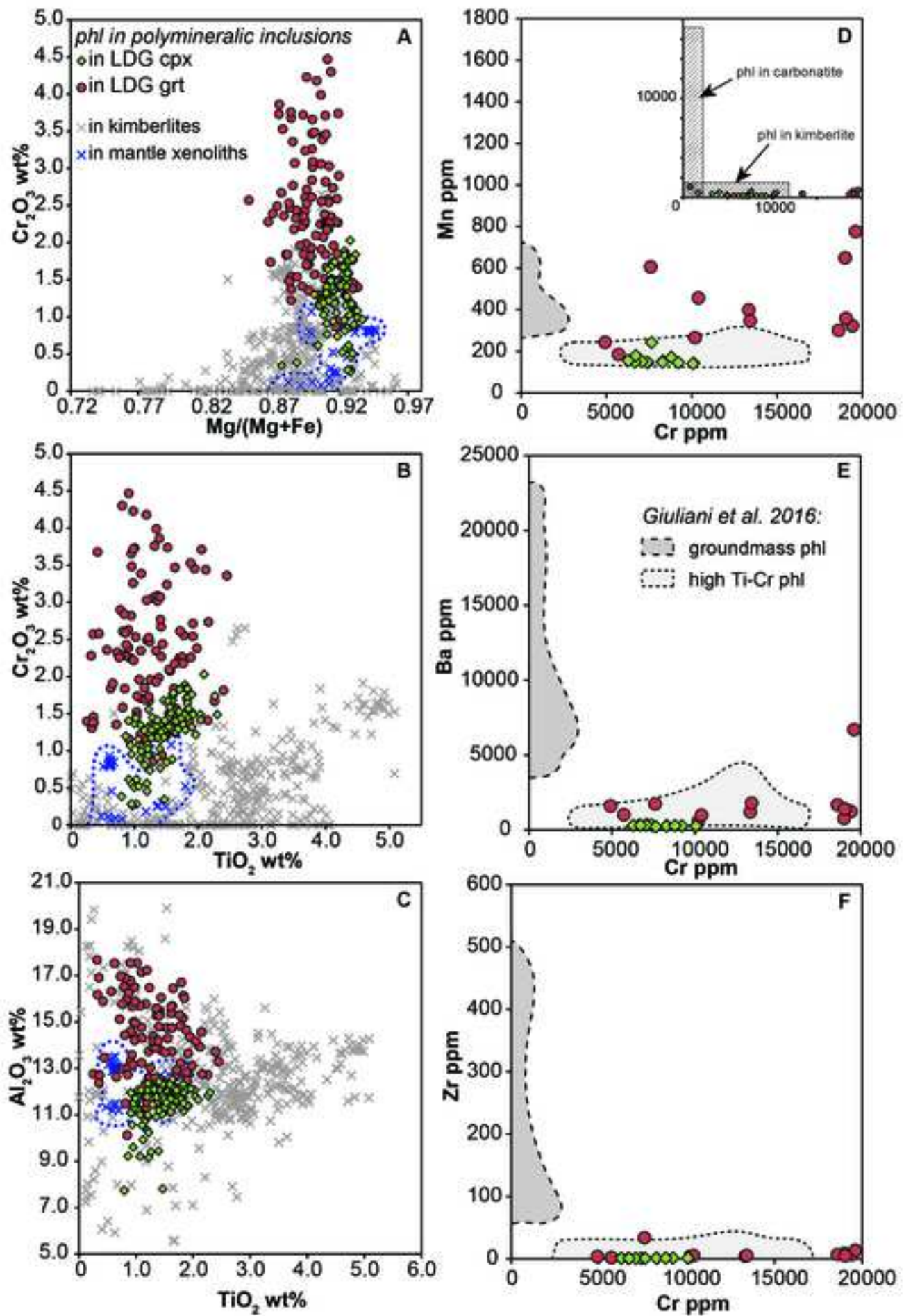


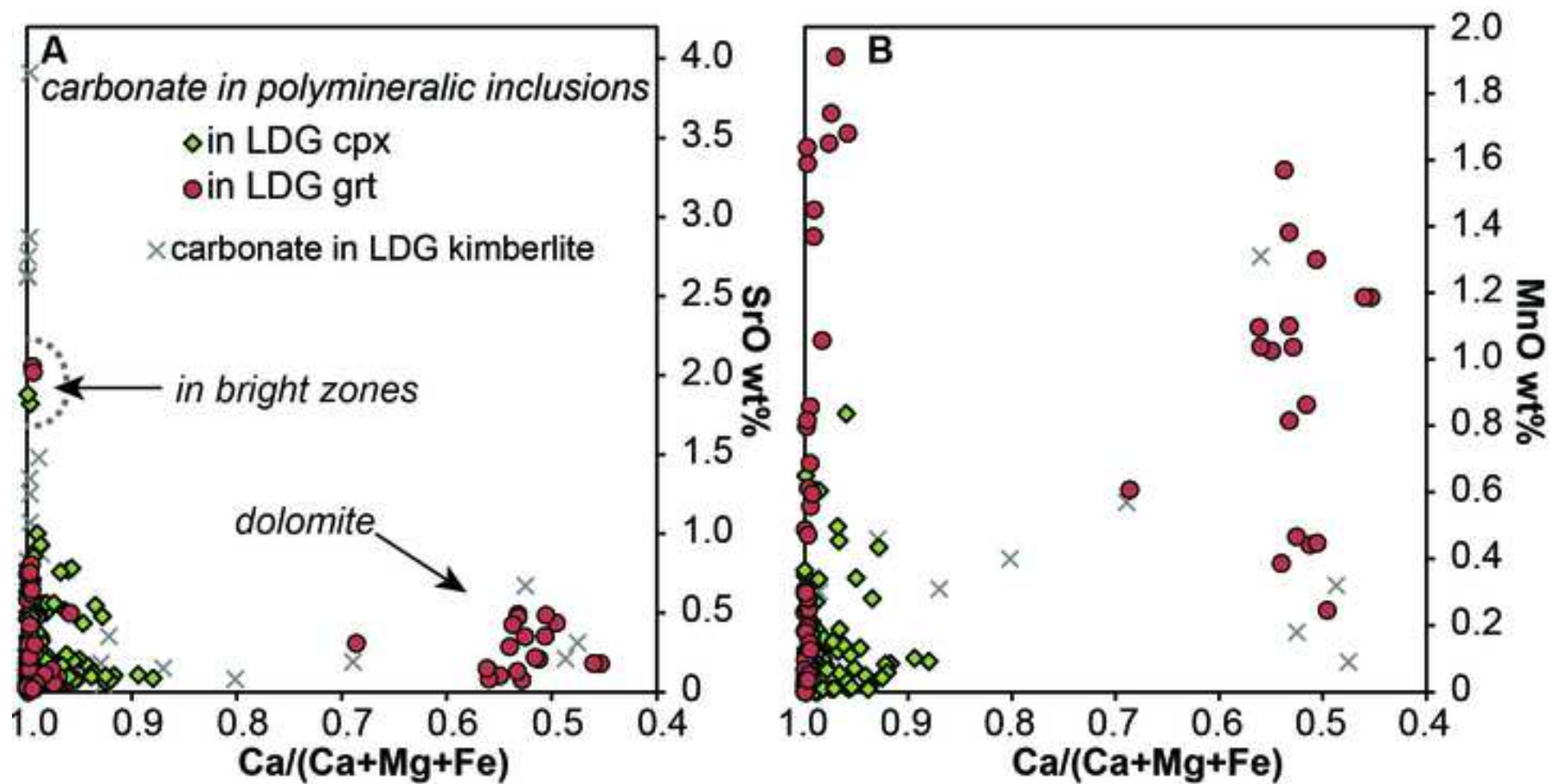


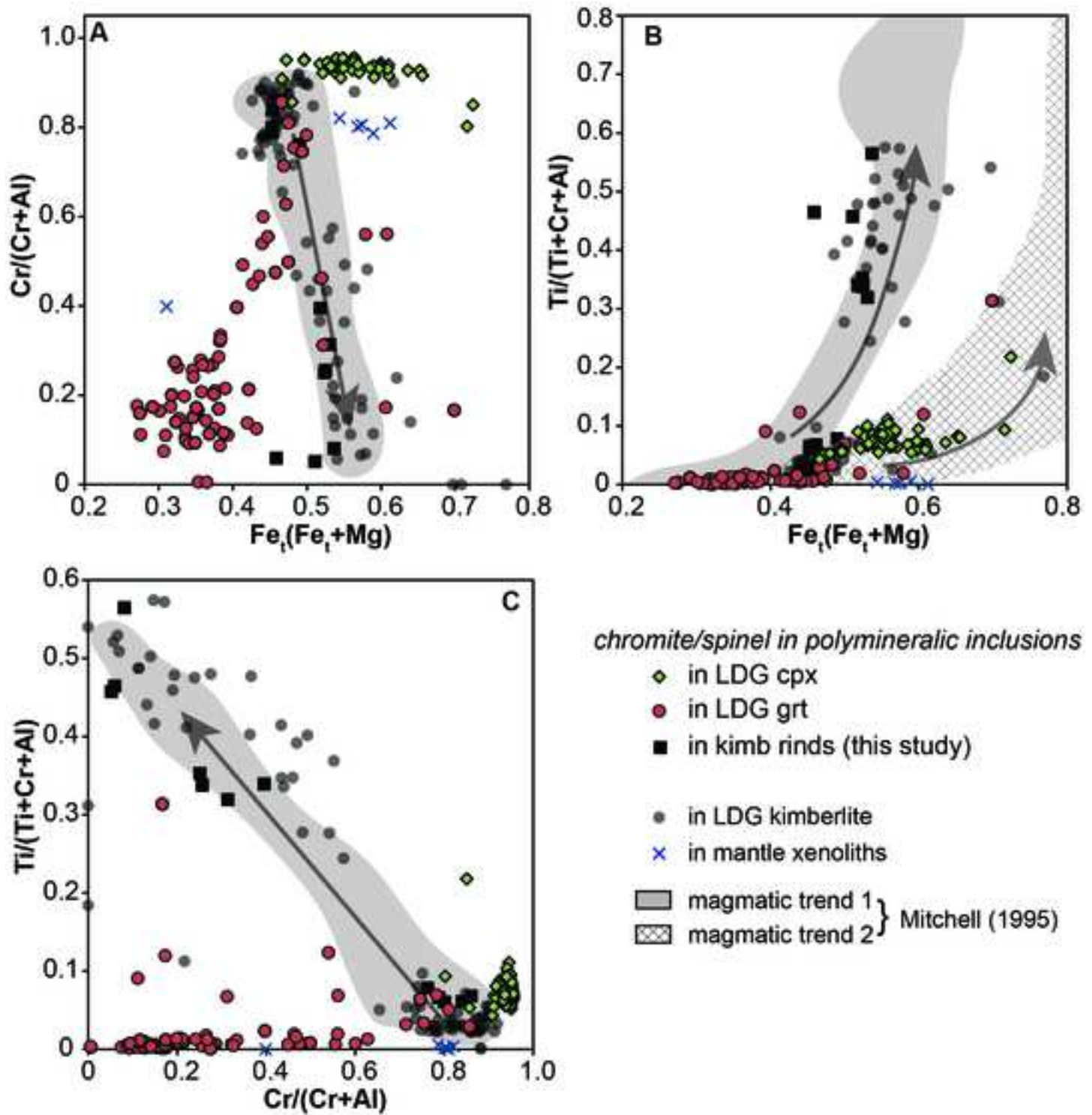


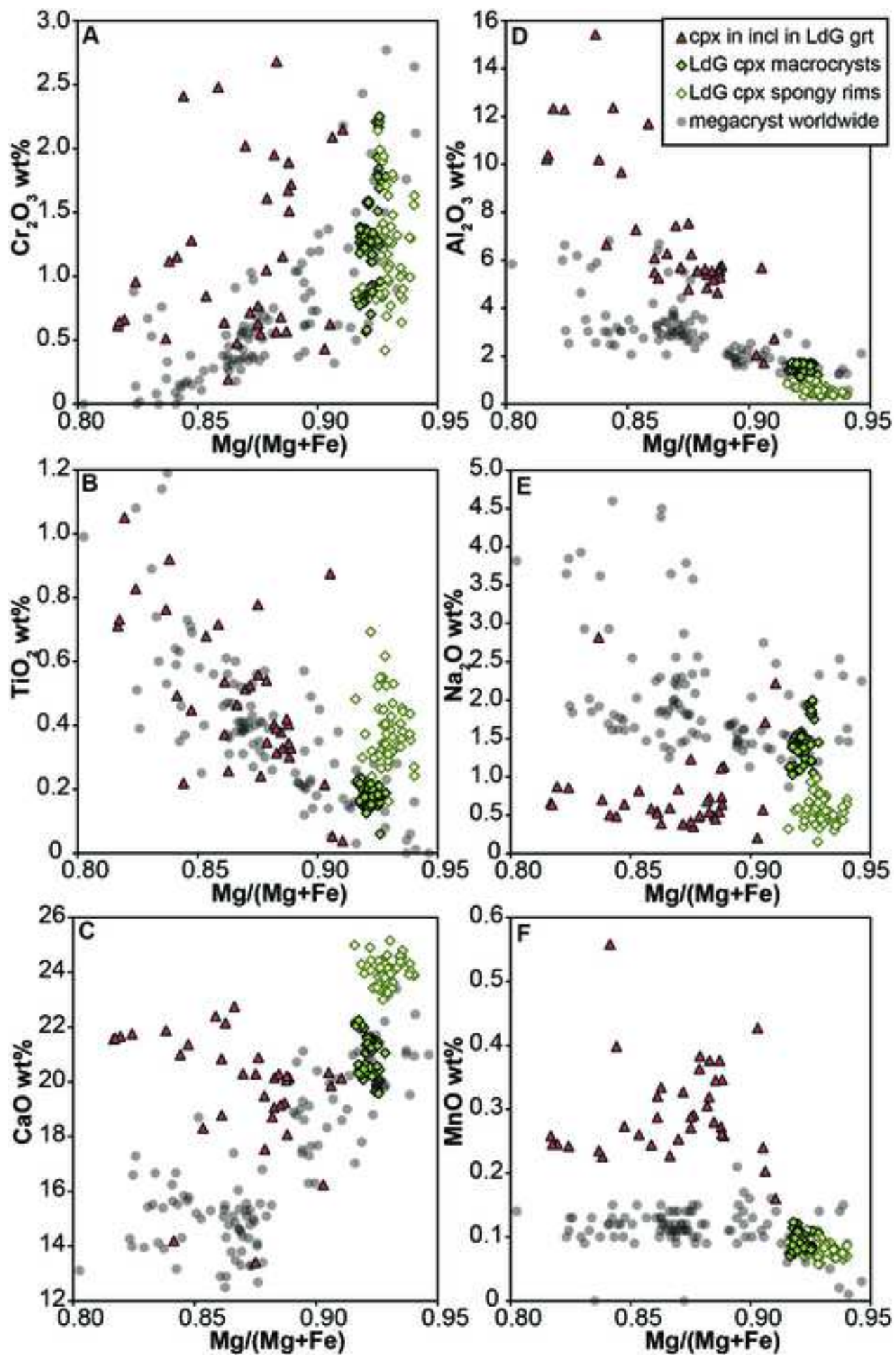


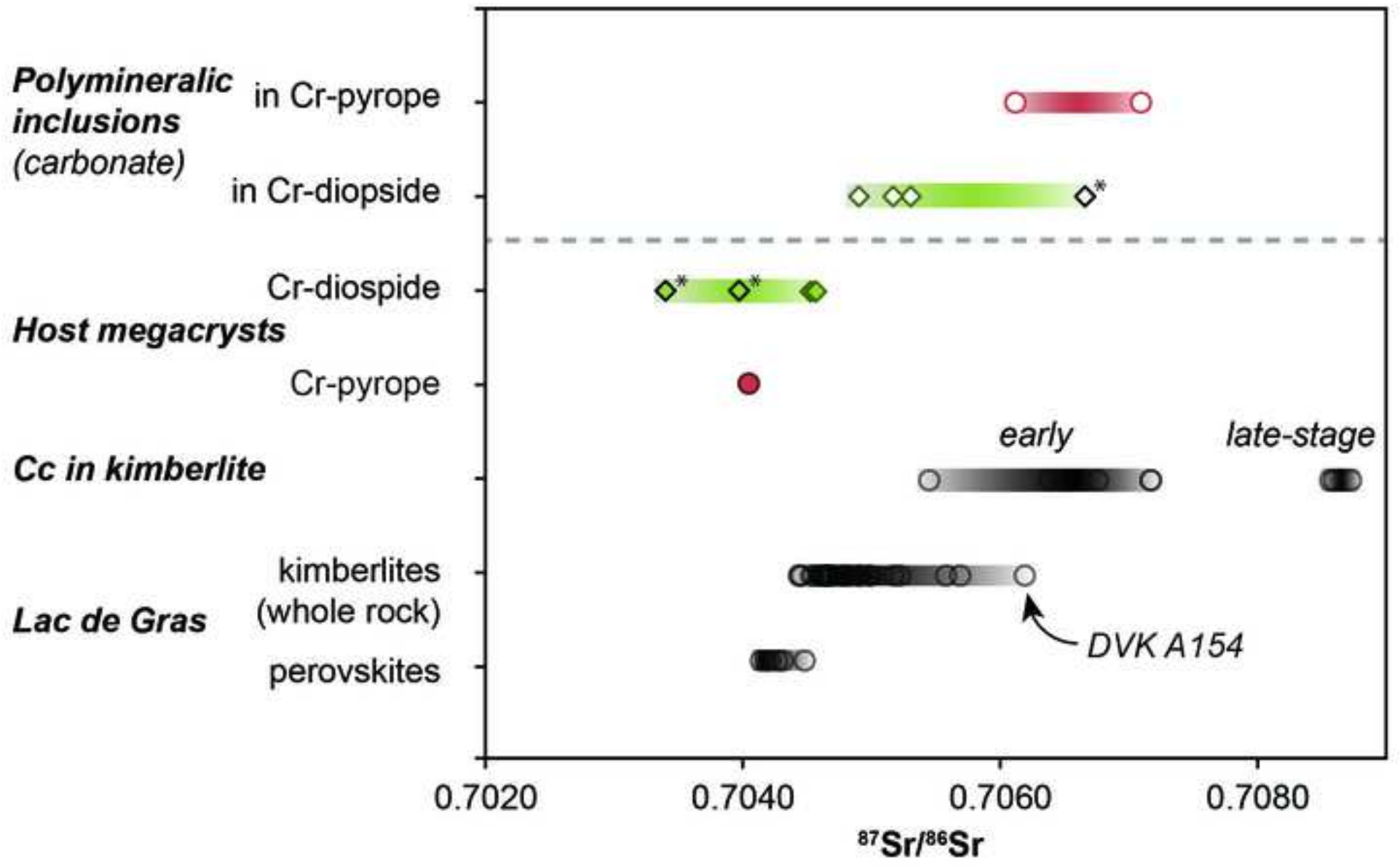


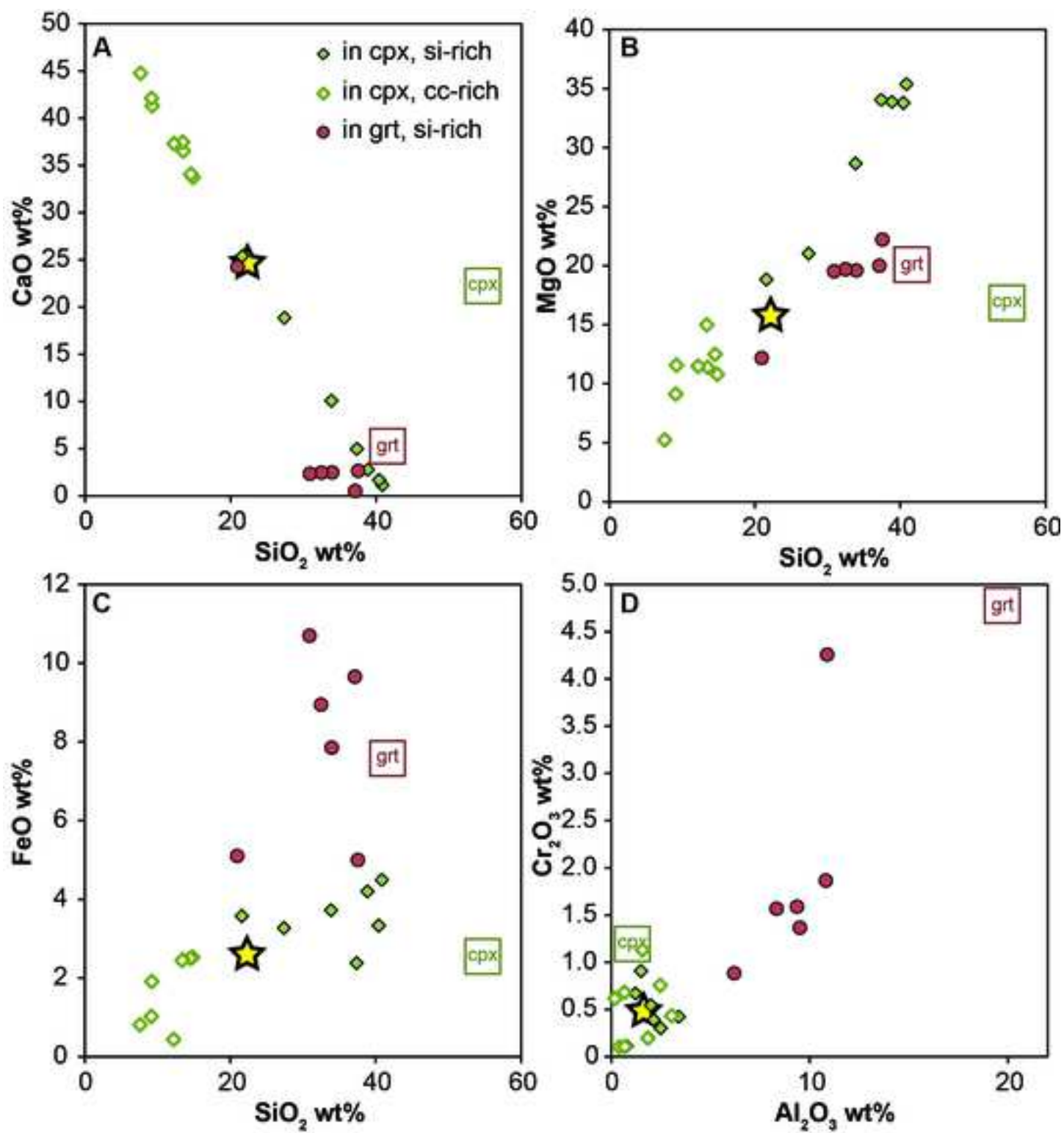


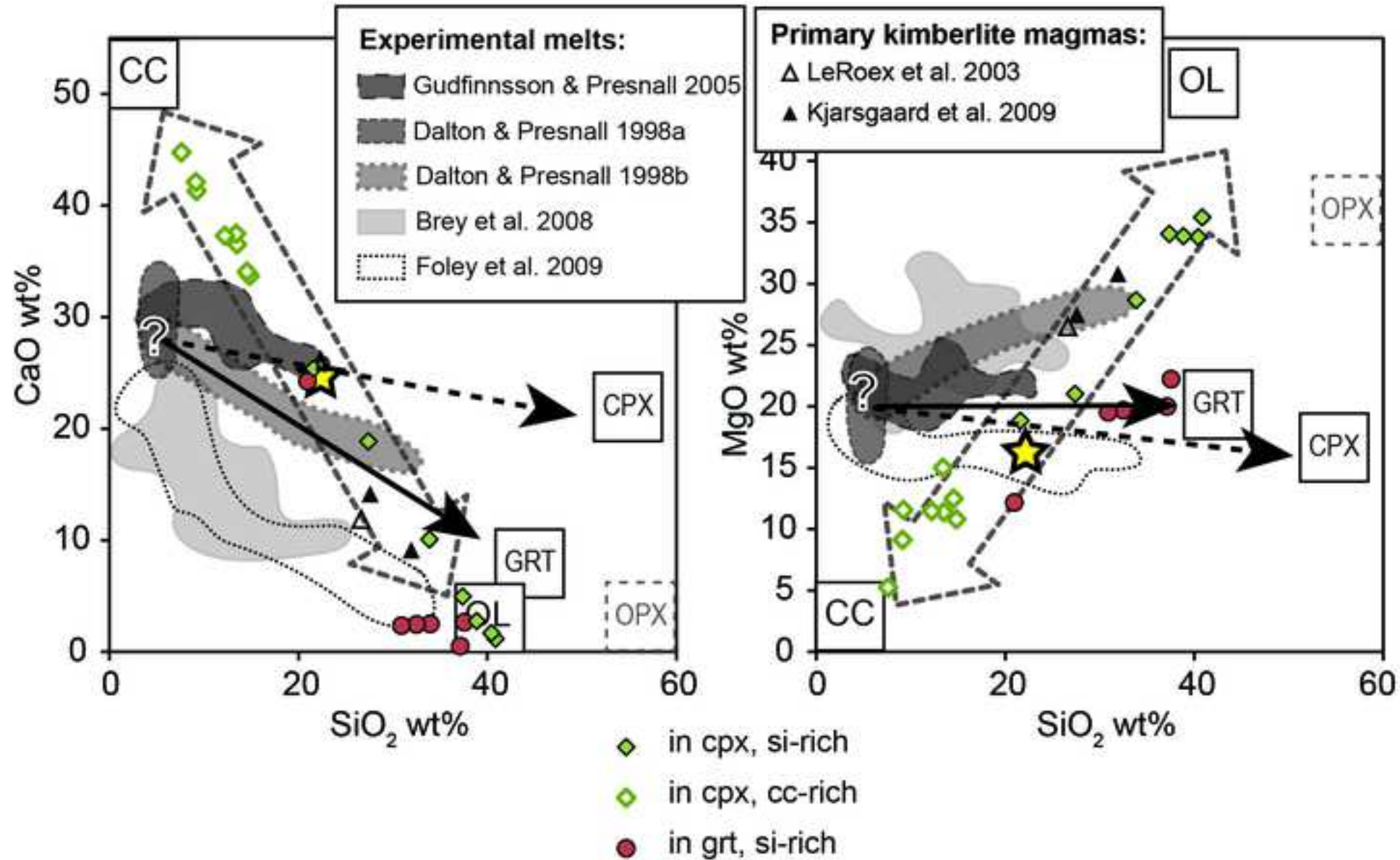












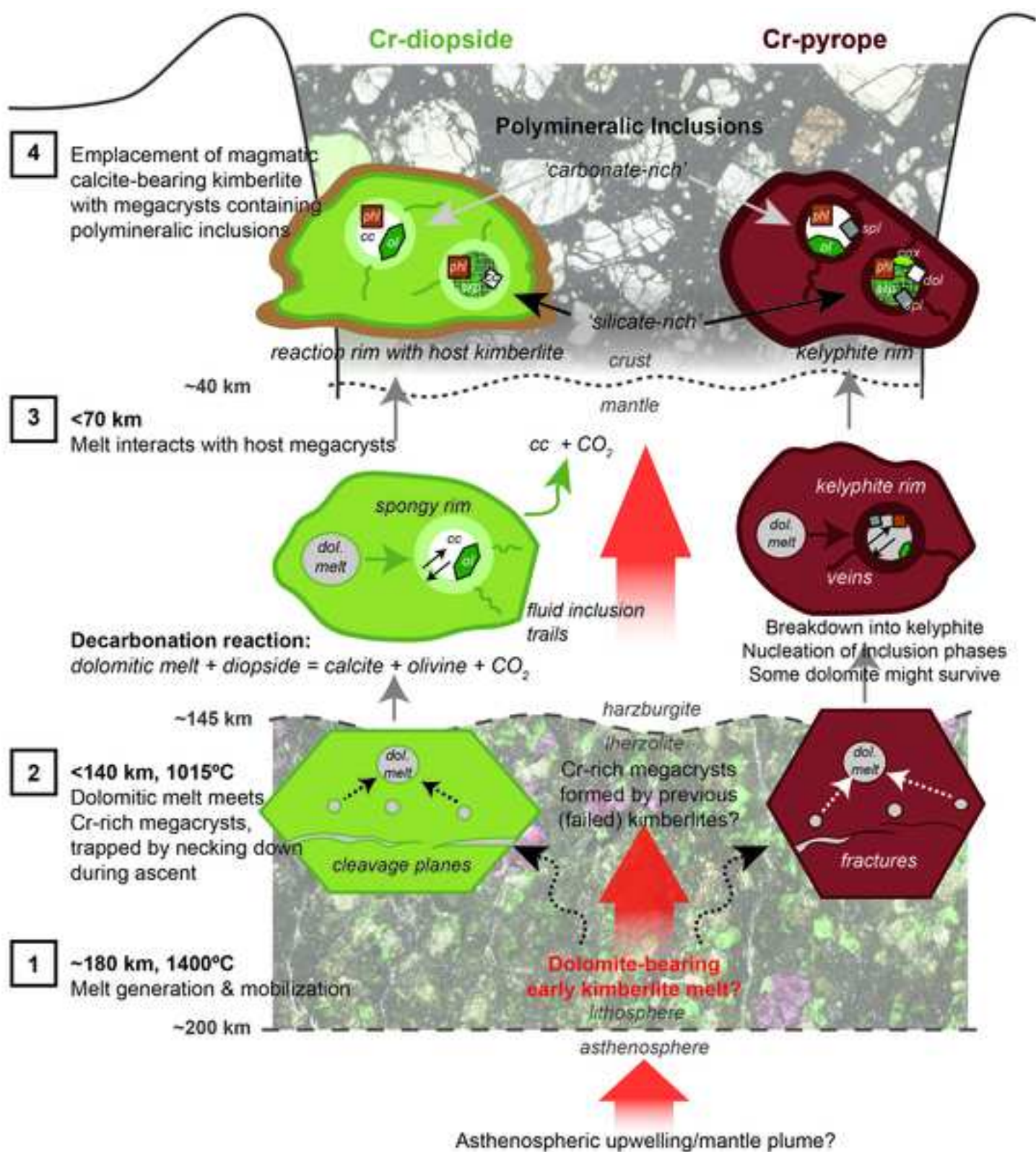


Table 1: P-T results for Cr-diopside megacryst hosts using single-cpx thermobarometry (Nimis and

Location	Diavik - A154N					
Sample	DVK_CD_04B	DVK_CD_06A	DVK_CD_06B	PL_CD_03 (In01 - 04)	PL_CD_03 (In05)	PL_CD_03 (In06)
T [°C]	1043	1009	1031	1025	965	1040
P [GPa]	5.3	4.6	4.7	4.5	4.2	4.6

Taylor, 2000)

Ekati - Point lake

PL_CD_03 (In07 - 08)	PL_CD_03 (In09)	PL_CD_03 (In10)	PL_CD_06 (In01, 04)	PL_CD_06 (In02, 02b)	PL_CD_06 (In03)	PL_CD_07 (In01 - 03)	PL_CD_07 (In04 - 06)
1025	938	1046	1017	1014	997	1043	1011
4.6	4.4	4.5	4.5	4.7	4.5	4.7	4.5

Table 2: Reconstructed bulk compositions of polymineralic inclusions in wt%

Location	DVK_CD_01		DVK_CD_01		DVK_CD_01		DVK_CD_01	
Sample	_In07		_In04		_In02		_In01	
Type	cc-rich		cc-rich		cc-rich		cc-rich	
		1 σ		1 σ		1 σ		1 σ
SiO ₂	14.8	1.66	14.5	1.63	13.4	1.50	12.2	1.37
TiO ₂	0.2	0.02	0.2	0.02	0.1	0.01	0.2	0.02
Al ₂ O ₃	1.6	0.18	3.1	0.34	0.2	0.02	0.7	0.07
Cr ₂ O ₃	1.1	0.13	0.4	0.05	0.6	0.07	0.7	0.08
MnO	0.1	0.01	0.1	0.01	0.5	0.06	1.1	0.12
FeO	2.5	0.28	2.5	0.28	2.4	0.27	0.4	0.05
NiO	-		-		-		-	
MgO	10.8	1.21	12.5	1.40	15.0	1.68	11.5	1.28
CaO	33.7	3.77	34.1	3.81	37.5	4.19	37.3	4.17
BaO	1.3	0.14	-		-		-	
Na ₂ O	-		0.1	0.01	-		-	
K ₂ O	0.7	0.08	2.8	0.31	-		0.2	0.03
SrO	0.8	0.09	0.1	0.01	0.1	0.01	0.1	0.01
F	-		0.1	0.01	-		-	
Cl	-		-		-		-	
Total	67.6		70.3		69.9		64.4	
Mg/(Mg+Fe)	0.88		0.90		0.92		0.98	

*overall uncertainty is ~11% and includes uncertainty on modal proportions obtained by QEMSCAN (~

Polymineralic inclusions in clinopyroxene

Diavik - A154N

DVK_CD_01 _In08 cc-rich	DVK_CD_01 _In03 cc-rich	DVK_CD_01 _In05 cc-rich	DVK_CD_6A _In05 si-rich	DVK_CD_6A _In06 si-rich
1 σ	1 σ	1 σ	1 σ	1 σ
9.2	9.1	7.6	40.9	40.4
1.03	1.02	0.85	4.57	4.52
0.1	0.1	0.2	0.3	0.4
0.01	0.01	0.02	0.03	0.05
0.4	0.7	1.9	2.0	2.5
0.05	0.08	0.21	0.22	0.28
0.1	0.1	0.2	0.5	0.3
0.01	0.01	0.02	0.06	0.03
0.3	-	-	0.1	0.1
0.03	-	-	0.02	0.01
1.9	1.0	0.8	4.5	3.3
0.21	0.11	0.09	0.50	0.37
-	-	-	-	-
11.6	9.1	5.2	35.4	33.8
1.29	1.02	0.59	3.96	3.78
41.3	42.1	44.8	1.2	1.7
4.62	4.71	5.01	0.13	0.19
-	0.1	0.1	-	-
-	0.01	0.01	-	-
-	-	-	-	0.1
0.3	0.5	1.6	0.7	1.3
0.03	0.06	0.18	0.08	0.15
0.1	0.1	0.1	-	-
0.01	0.01	0.01	-	-
-	-	0.1	-	-
-	-	0.01	-	-
-	-	-	-	-
65.4	63.0	62.5	85.6	84.0
0.92	0.94	0.92	0.93	0.95

5%) and variability of mineral compositions of inclusion phases obtained by EPMA (~10%).

DVK_CD_6A _In03 si-rich		DVK_CD_01 _In06 si-rich		DVK_CD_6A _In02 si-rich		DVK_CD_6A _In01 si-rich		DVK_CD_6A _In04 si-rich	
	1 σ		1 σ		1 σ		1 σ		1 σ
38.9	4.35	37.4	4.18	33.9	3.79	27.4	3.06	21.6	2.41
0.2	0.02	0.2	0.02	0.2	0.03	0.5	0.05	0.2	0.02
1.2	0.14	0.8	0.09	2.2	0.24	3.4	0.38	1.5	0.17
0.7	0.07	0.1	0.01	0.4	0.04	0.4	0.05	0.9	0.10
0.1	0.02	0.1	0.01	0.1	0.01	0.1	0.01	0.1	0.01
4.2	0.47	2.4	0.27	3.7	0.42	3.3	0.37	3.6	0.40
-		-		-		-		-	
33.9	3.79	34.1	3.81	28.7	3.20	21.0	2.35	18.8	2.11
2.8	0.31	5.0	0.56	10.1	1.13	18.9	2.11	25.4	2.84
-		-		-		0.1	0.01	-	
0.1	0.01	-		-		0.1	0.01	-	
0.3	0.04	0.2	0.02	0.8	0.08	2.7	0.30	0.5	0.06
-		-		0.1	0.01	0.1	0.01	0.2	0.02
-		-		-		0.1	0.01	-	
-		-		-		-		-	
82.4		80.1		80.0		77.9		72.9	
0.93		0.96		0.93		0.92		0.90	

Ekati - Point lake

PL_CD_03

_In01

cc-rich

1 σ

13.5 1.51

0.4 0.04

2.5 0.28

0.8 0.08

0.3 0.04

2.5 0.28

- -

11.4 1.27

36.5 4.08

- -

- -

2.3 0.25

0.1 0.02

0.4 0.05

- -

70.6

0.89

PL_GRT_04

_In14

si-rich

1 σ

37.6 4.20

0.6 0.06

8.3 0.93

1.6 0.18

0.1 0.01

5.0 0.56

0.1 0.01

22.2 2.48

2.7 0.30

- -

0.1 0.01

3.2 0.36

- -

0.1 0.01

- -

81.5

0.89

PL_GRT_04

_In7

si-rich

1 σ

37.1 4.15

0.4 0.05

9.5 1.06

1.4 0.15

0.1 0.01

9.7 1.08

0.4 0.04

20.0 2.24

0.5 0.06

- -

0.1 0.02

5.1 0.57

- -

0.1 0.01

- -

84.4

0.79

Polymineralic inclusions in garnet

Ekati - Point lake

PL_GRT_04

_In04

si-rich

1 σ

33.9 3.80

0.5 0.05

10.8 1.21

1.9 0.21

0.1 0.01

7.9 0.88

0.3 0.03

19.6 2.19

2.5 0.28

- -

0.2 0.02

5.1 0.57

- -

0.1 0.02

- -

82.8

0.82

PL_GRT_04

_In05

si-rich

1 σ

32.5 3.63

0.2 0.03

9.4 1.05

1.6 0.18

0.1 0.02

8.9 1.00

0.4 0.04

19.7 2.20

2.5 0.28

- -

0.3 0.04

2.9 0.33

- -

0.1 0.01

- -

78.6

0.80

PL_GRT_04 _In03 si-rich		PL_GRT_04 _In13 si-rich	
	1 σ		1 σ
30.9	3.46	20.9	2.34
0.3	0.04	0.6	0.06
10.9	1.22	6.2	0.69
4.3	0.48	0.9	0.10
0.1	0.02	0.1	0.01
10.7	1.20	5.1	0.57
0.3	0.03	0.2	0.02
19.5	2.18	12.2	1.36
2.4	0.26	24.3	2.71
-		-	
0.4	0.04	0.1	0.01
1.5	0.17	3.1	0.35
-		-	
-		0.3	0.03
-		-	
81.3		73.9	
0.76		0.81	