3 4 5	1	Redox state of the Dharwar craton root as inferred from eclogite and peridotite
6 7 8	2	sourced mantle cargo, with implications for kimberlite and lamproite magma
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58 59	22	Despite over 400 occurrences of kimberlites and related rocks in India, mantle-derived xenoliths
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kimberlites has hampered investigations of the subcontinental lithospheric mantle (SCLM). Using a valuable selection of the rare xenolith inventory, we here report Fe³⁺/ Σ Fe measurements for garnets using the electron microprobe (EPMA) flank method, targeting six mantle eclogite xenoliths (KL2 pipe) and fourteen peridotitic garnet xenocrysts (P9 and P10 hypabyssal intrusions) from the Wajrakarur kimberlite field (WKF) on the Eastern Dharwar craton (EDC). These data provide some of the first direct constraints on the oxygen fugacity (*f*O₂) of the lithospheric mantle beneath the Indian subcontinent.

The measured Fe³⁺/ Σ Fe ratios vary between 0.02 and 0.05 (±0.01) for the eclogite xenoliths and between 0.02 and 0.10 (±0.01) for the peridotitic garnets. Calculated $\Delta \log fO_2$ values for the KL2 eclogites show a wide range from FMQ-3.9 to FMQ-0.9 (±0.6), straddling the boundary between the diamond and carbonate stability fields. In terms of redox compositions, it appears that the KL2 eclogites are able to host diamond, which is consistent with the diamondiferous nature of this particular WKF locality and the presence of eclogitic garnet inclusions in diamonds from the nearby TK4 kimberlite body.

The peridotitic garnet xenocrysts from the P9 and P10 kimberlite bodies, which were entrained between ~125 and 170 km depth, reveal $\Delta \log fO_2$ values between FMQ-4.5 and FMQ-2.6 (±0.9). Garnet xenocrysts with 'normal' REE patterns exhibit higher $Fe^{3+}/\Sigma Fe$ ratios compared to garnets with 'sinusoidal' REE patterns. Importantly, the Fe³⁺/ Σ Fe ratios of garnet xenocrysts with 'normal' REE patterns (~125-160 km depth) correlate with metasomatic Ti-Y-Zr-V enrichment, which suggests metasomatism-driven oxidation of the cratonic mantle at mid-lithospheric depths. Such melt-related mantle metasomatism was probably diamond-destructive within the otherwise diamond-fertile lithospheric keel.

The observed wide range of $\Delta \log fO_2$ values for the Dharwar cratonic mantle lithosphere allows for stabilization of various metasomatic phases (e.g., amphiboles, micas, carbonates) that may have formed (or concentrated in) distinctly different metasome assemblages within the continental root that underpins Peninsular India. Changing the relative contributions from such highly diverse volatile-rich metasomes may explain the spatiotemporal association of kimberlites and various diamond-bearing potassic magma types such as orangeites, ultramafic lamprophyres and lamproites, a scenario that is influenced by the redox composition of the Dharwar craton root.

Keywords: Continental lithospheric mantle, Eclogite, Garnet, Oxygen fugacity, Mantle redox, Wajrakarur kimberlites, Southern India

Introduction

Oxygen fugacity (fO_2) has significant implications for the speciation of CHONS-volatiles and the stability of diamond/graphite in the Earth's mantle (Stagno, 2019). Oxygen fugacity also influences (i) melting and metasomatism of mantle rocks, thereby influencing fluxes of various magma types, and (ii) recycling of volatiles, thereby affecting global volatile cycles (Yaxley et al., 2017). Therefore, understanding fO_2 in Earth's mantle is crucial for comprehending terrestrial magmatism and volatile cycles (Foley, 2011; Tappe et al., 2018). The fO₂ compositions of the subcontinental lithospheric mantle (SCLM) have been shown to vary with depth and time (Woodland and Koch, 2003; Creighton et al., 2009, 2010; Yaxley et al., 2017; Tappe et al., 2021; Aulbach et al., 2022), which has the potential to shift melting regimes over geological timescales (Foley, 2011). The fO₂ values can be estimated for mantle-derived peridotite (Ballhaus et al., 1991;

68 Stagno et al., 2013) and eclogite (Stagno et al., 2015) xenoliths by measuring the Fe³⁺/ Σ Fe ratios 69 of their constituting garnet fractions.

This contribution presents the first measurements of Fe³⁺/ Σ Fe in garnets from six eclogite xenoliths from the KL2 hypabyssal intrusion and fourteen peridotitic garnet xenocrysts from the P9 and P10 intrusions of the Wajrakarur Kimberlite Field (WKF) located on the Eastern Dharwar craton (EDC) in southern India. We aim to constrain the redox state of the regional SCLM that underpinned Peninsular India at ca. 1.1 Ga. The KL2 intrusion forms part of the Kalyandurga cluster, which consists of seven minor kimberlite intrusions (KL1 to KL7) and is located in the southern part of the WKF (Fig. 1). The P9 and P10 minor intrusions form part of the Wajrakarur-Lattavaram cluster, situated in the north. Shaikh et al. (2020) constrained the architecture and thermal state of the regional SCLM using major and trace element compositions of garnets from P9 and P10, some of which were re-analyzed here for their Fe³⁺/ Σ Fe compositions. According to Shaikh et al. (2020), lithospheric thinning beneath the Dharwar craton, from ~190 km depth at 1.1 Ga to ~ 120 km depth at present, was aided by extensive melt metasomatism at the bottom of the SCLM.

The ca. 1.1 Ga Kalyandurga kimberlites are special in several ways. For instance, (i) the diamondiferous KL2 minor intrusion is distinguished by an overabundance of eclogite xenoliths (>95% of the mantle xenolith population are eclogite nodules: Rao et al., 2001; Neelakantam, 2001; Patel et al., 2006); (ii) the KL4 minor intrusion exhibits an exceptional abundance of mantle-derived ilmenite macrocrysts (Sastry et al., 2005); (iii) the Kalyandurga kimberlites intruded the 2.6–2.5 Ga Closepet granite in close proximity to the Chitradurga Boundary Fault, which presents an important structure for the understanding of the evolution of the Dharwar craton (e.g., Chadwick et al., 2000). KL2 eclogite nodules have previously been investigated for petrography, in-situ

mineral major and trace element contents, and oxygen isotopic compositions to constrain their origins (Ganguly and Bhattacharya, 1987; Rao et al., 2001; Patel et al., 2006, 2009; Griffin et al., 2009; Dongre et al., 2015). Four eclogite xenoliths were examined by Ganguly and Bhattacharya (1987), while two eclogite nodules were examined by Rao et al. (2001) to determine mineral major element compositions. The first detailed petrographic observations and mineral major element data for a suite of eleven KL2 eclogite xenoliths were provided by Patel et al. (2006). In order to review the P-T conditions and origins of eclogites and pyroxenites from the WKF, Patel et al. (2009) combined published xenolith data with nine newly analyzed xenoliths and argued against the involvement of subducted ancient oceanic crust in the formation of KL2 eclogites. These authors used textural and mineral compositional evidence to support a magmatic cumulate origin for the KL2 eclogites. Babu et al. (2008) presented a much larger eclogite dataset (35 xenoliths) at the 9th International Kimberlite Conference, and Griffin et al. (2009) utilized these results to argue that this eclogite xenolith suite originated from mafic melts that ponded and crystallized at lithospheric mantle depths. Mineral major and trace element compositions, as well as the lateral distribution of mantle-derived xenoliths and xenocrysts, were used to support this interpretation (Griffin et al., 2009). In opposition to this notion, Dongre et al. (2015) examined 28 KL2 eclogite xenoliths and reported the first δ^{18} O data (+5.3 to +7.8%), which are best explained by a subduction-recycling model. Recently, a few more KL2 eclogite nodules were studied by Chatterjee et al. (2023) supporting the subduction model. However, no attempt has been made so far to use the eclogites and other mantle-derived materials to determine the oxidation state of the SCLM beneath southern India.

Herein, we apply the calibration of Stagno et al. (2015), which estimates eclogite fO_2 using the major element compositions of garnet and clinopyroxene, together with Fe³⁺/ Σ Fe ratio

measurements for garnet, an approach taken in several previous eclogite xenolith studies from cratons worldwide (e.g., Smart et al., 2017, 2021a; Aulbach et al., 2019, 2022; Burness et al., 2020; Mikhailenko et al., 2020). Our garnet Fe³⁺/ Σ Fe measurements are complemented by in-situ major and trace element data for garnet crystals from the KL2 eclogites. For peridotitic garnet xenocrysts, we used the calibration of Stagno et al. (2013) and combined garnet $Fe^{3+}/\Sigma Fe$ data with their modeled $Fe^{2+}/(Fe^{2+}+Mg)$ compositions, assuming equilibrium with olivine and orthopyroxene in the original peridotite host rock (O'Neil and Wood, 1979; Harley, 1984; Ryan et al., 1996; Gaul et al., 2000) at estimated equilibrium P-T conditions (Canil et al., 1999). Our results provide the first fO_2 estimates for eclogites and peridotitic garnets from southern India entrained by ca. 1.1 Ga old Mesoproterozoic kimberlites that represent one of the earliest global emplacement events of diamond-bearing deep-sourced magmas (Tappe et al., 2018).

Analytical techniques

Seven eclogite xenoliths (KL2A to KL2G), measuring 3 to 5 cm in diameter, were collected from the KL2 kimberlite body by S. Ravi. They were cut and polished into a total of 18 standard petrographic thin sections for this study. The eclogite xenoliths are commonly rounded to subrounded (oval) nodules with medium-grained textures. Honey brown pyrope garnet crystals are embedded in a light green to white matrix formed mainly by altered omphacitic clinopyroxene.

A CAMECA SX100 electron microprobe housed in the Spectrum lab at the University of Johannesburg was used for quantitative in-situ mineral analyses of major and minor elements. The analyses were conducted with a nominal beam size of 1 µm, an accelerating voltage of 20 kV, and a beam current of 20 nA. Trace elements were measured for garnet, clinopyroxene and kyanite in representative eclogite thin sections at the University of Johannesburg using a Thermo Scientific

iCAP RO inductively coupled plasma mass spectrometer (ICP-MS) coupled to a 193 nm ArF RESOlution SE155 excimer laser. NIST SRM 612 was used as the calibration reference material, and ²⁹Si was used for internal standardization. To verify accuracy of the data, various USGS basaltic glasses (BCR-2G, BHVO-2, BIR-1G) and the in-house GHR1 megacrystic garnet from the Monastery kimberlite were analyzed as secondary standards, similar to the setup reported in Tappe et al. (2023). Data quality is documented in Supp. Table 1. When compared to recommended values for the USGS glasses and the GHR1 garnet (e.g., Jochum et al., 2016; Tappe et al., 2021), the majority of trace elements analyzed reproduced with a variance of less than 10 to 15%. To avoid any potential contamination from the host kimberlite during laser ablation, we rejected ICP-MS trace element measurements with >1 ppm Ba (see Shaikh et al., 2020). The analytical techniques and data are described in more detail in the Appendix 1 and Supp. Table 1, respectively. The analytical techniques applied to measure the major and trace element concentrations for fourteen peridotitic garnet xenocrysts from Wajrakarur kimberlites P9 and P10 are given in Shaikh et al. (2020).

The atomic Fe³⁺/ Σ Fe proportions in garnet crystals from six eclogite nodules and fourteen xenocrysts of peridotitic affinity were determined with the flank method as developed by Höfer et al. (1994) and further refined by Höfer and Brey (2007). Measurements were conducted with a JEOL JXA-8530F Plus electron microprobe at Goethe University Frankfurt, Germany. The flank method and the quantitative elemental analyses were conducted simultaneously using WDS at 15 kV and 120 nA, with a beam diameter of 1 μ m. Two spectrometers with TAPL crystals for high intensities and the smallest detector slit (300 μ m) were used, with 100 s counting time for FeLa and FeL_{β}. The Fe³⁺/ Σ Fe ratios for garnets were determined by applying the correction for self-absorption using natural and synthetic garnet crystals with variable total Fe and Fe³⁺/ Σ Fe, as

independently determined by Mössbauer spectroscopy (Höfer and Brey, 2007). We used Damknolle garnet megacrysts from Nigeria (Rankenburg et al., 2004) and the McGuire almandine from the U.S.A. (McGuire et al., 1992) along with three in-house Cr-pyrope standards (UA5, UA10, UA17) as reference materials. The Cr-pyrope crystals have a wide range of total Fe, which is ideal for calibration (Supp. Table 1) to obtain fit parameters derived exclusively from these three natural garnets. The other three spectrometers, not involved in flank method measurements, carried out elemental analysis of Si, Ti, Al, Cr, Fe, Mn, Ni, Mg, Ca, Na, K and P during the same measurement cycle. Appropriate silicate minerals [pyrope (Mg, Al, Si), albite (Na), CaSiO₃ (Ca)] and a phosphate mineral [KTiOPO4(Ti,K,P)], as well as metals or metal oxides [iron metal (Fe), NiO (Ni), MnTiO₃ (Mn), Cr₂O₃ (Cr)] were used as calibration standards. A PRZ routine was used for matrix correction. The analytical uncertainty in $Fe^{3+}/\Sigma Fe$ ratio measurements by the EPMA flank method is approximately ± 0.01 (1 σ), and it is noted that in general the uncertainty is inversely correlated with garnet FeO content. A recent discussion about the applicability of the flank method for determining ferric-ferrous iron ratios can be found in Rzehak et al. (2020).

Results

Petrography

KL2 Eclogites

Secondary alteration is prominent in the KL2 eclogite nodules, with a general decrease in the level 51 179 of alteration from omphacite to kyanite to garnet (Fig. 2, 3), which was also observed previously (Sastry et al., 2005; Patel et al., 2006; Dongre et al., 2015). Garnet crystals are least altered but show minor secondary carbonate and hydro-garnet replacement along grain boundaries and fractures. Despite the alteration of several of our KL2 eclogite samples, the secondary minerals

preserve the original rock textures, such as elongation texture with substantial micro-fracturing in garnet and clinopyroxene relics (e.g., Fig. 2, 3). Among the examined eclogite nodules, substantial clinopyroxene and kyanite alteration was observed in five samples (KL2C, KL2D, KL2E, KL2F, KL2G), whereas these phases were completely altered in sample KL2B. The freshest sample KL2A does not contain kyanite.

Subhedral to spherical garnet grains are interlocked with anhedral clinopyroxene and bladed kyanite in relatively fresh samples with minor alteration along grain boundaries. Inclusions of garnet and clinopyroxene are frequent in kyanite (Fig. 3 d, e, f). A few eclogite nodules (KL2D, KL2F) contain needles of corundum, which are generally spatially associated with kyanite (Fig. 4a). Rutile is preserved as an inclusion in kyanite within the KL2A eclogite nodule (e.g., Fig. 4b). In the majority of samples analyzed, the relative modal proportions of garnet, omphacite and kyanite are ~45:45:10, and ~50:50 in bimineralic kyanite-free eclogite nodules with only very minor kyanite and corundum in a few samples. Carbonate and chlorite are the most prevalent alteration products of all primary minerals. Omphacite is typically replaced by hornblende and diopside, kyanite by celsian feldspar (along grain boundaries and fractures), and garnet by chlorite, epidote, celsian and grossular. The matrix of the eclogites contains rare sulfides (NiFeS) and pure nickel oxide (NiO). Mukherjee et al. (2021) reported native gold particles and Au-Pt alloys from KL2 eclogite xenoliths. A very detailed petrographic report for KL2 eclogites is given by Patel et al. (2006).

Peridotitic garnet xenocrysts

Fourteen peridotitic garnet xenocrysts (3-7 mm in size) from the P9 and P10 minor intrusions were mounted on thin sections and polished for in-situ mineral chemical examination of major and trace

elements. Garnet grains exhibit micro-fracturing and host rare inclusions of Cr-diopside consistent with their peridotitic mantle origins.

Major and trace element compositions of minerals

KL2 eclogites

The five primary minerals in KL2 eclogites (garnet, clinopyroxene, kyanite, corundum, rutile) were analyzed for major and trace elements. Secondary phases such as celsian, and radite, grossular, serpentine and carbonates were also analyzed for major elements. Supp. Table 1 lists representative major, minor and trace element compositions of primary phases essential for this work, as well as the compositions of secondary phases for documentation purposes. We only address the compositions of primary phases here, and readers are referred to Patel et al. (2006) for a detailed description of secondary phases in the KL2 eclogite xenoliths.

Despite the secondary alteration along grain boundaries and fractures, garnets in KL2 eclogites show relatively uniform compositions (Supp. Table 1). The eclogitic garnet compositions determined here vary between Alm_{24–27}Grs_{25–58}Pyp_{18–48} falling within the range of published data for KL2 (e.g., Patel et al., 2006; Dongre et al., 2015). Garnets have low Na₂O and TiO₂ contents (<0.3 wt.%; Supp. Table 1), which is typical for low-pressure eclogites (Gurney and Moore, 1993). Chondrite-normalized (after Sun and McDonough, 1989) REE patterns of garnets from KL2 eclogites show LREE-depletion, a positive Eu anomaly, and flat MREE-HREE distributions (Fig. 5c), which is characteristic for garnets from cratonic eclogites worldwide (e.g., Jacob, 2004; Aulbach and Jacob, 2016; Aulbach and Smart, 2023). Garnets show variable trace element concentrations: Sc (29-35 ppm), Ti (828-1135 ppm), V (48-71 ppm), Cr (109-518 ppm), Ni (15-

Clinopyroxenes are omphacitic in nature $(Ae_{7-10}Jd_{31-43}Di_{50-61})$ similar to the known compositions of KL2 eclogitic clinopyroxene (Ae₃₋₁₇Jd₁₃₋₅₂Di₄₁₋₇₉). Contents of Na₂O and Al₂O₃ range from 2.9-8.2 wt.% and 5.8-16.2 wt.%, respectively. Cr₂O₃ content is low at 0.1 wt.%, rarely approaching 1.6 wt.%. Clinopyroxene shows a clear negative correlation between MgO and Na₂O (inset in Fig. 5b), typical for kimberlite-borne eclogites. Their chondrite-normalized REE patterns exhibit high LREE/HREE with a notable positive Eu anomaly (Fig. 5d).

Kyanite in KL2 eclogite xenoliths contains trace amounts of FeO (<0.5 wt.%) and Cr₂O₃ (<0.9 wt.%) (Supp. Table 1). Most trace elements have concentrations below the EPMA detection limit (<30 ppm), except for Ti (106-294 ppm), V (35-70 ppm), Cr (157-604 ppm) and Ga (7-10 ppm). Corundum is a pure aluminous phase with a mean Al_2O_3 content of ~99 wt.% and minor amounts of FeO (<0.5 wt.%) and Cr₂O₃ (<0.16 wt.%). Rutile is a pure Ti-oxide phase (~99 wt.% TiO_2) with minor amounts of FeO (<0.12 wt.%) and Cr_2O_3 (<0.18 wt.%).

Peridotitic garnet xenocrysts

Major and trace element compositions of peridotitic garnet xenocrysts were discussed in detail by Shaikh et al. (2020). For this redox study, we included 11 lherzolitic G9 grains, two harzburgitic G10 grains, and a single Ti-metasomatic G11 grain (garnet classification of Grütter et al., 2004; Fig. 6a). Overall, these garnet crystals show a wide range of Cr_2O_3 (1.8–11.8 wt.%), moderate CaO (4.3-5.9 wt.%) and very low TiO₂ contents (<0.35 wt.%). Most of these garnets show 'normal' chondrite-normalized REE patterns, however, xenocrysts with >5 wt.% Cr₂O₃ are characterized

by 'sinusoidal' REE patterns (Fig. 6b). Clinopyroxene inclusions in the garnet xenocrysts are Crdiopsides (1.1–2.1 wt.% Cr_2O_3 ; $En_{50-55}Wo_{41-46}Fs_4$) with consistently high Mg# values (0.92–0.94).

Bulk eclogite reconstruction and fO₂ estimates based on redox-sensitive elements

Due to pervasive alteration and kimberlite melt infiltration of the KL2 eclogite nodules (see Fig. 3), bulk-rock compositions were reconstructed from the measured chemical compositions of the primary minerals (clinopyroxene, garnet, corundum, rutile, \pm kyanite) in combination with their estimated modal mineral abundances. This method of bulk-rock reconstruction has been widely used in the past and continues to be a valuable tool in the study of xenolithic eclogites (e.g., Jacob, 2004; Tappe et al., 2011; Smart et al., 2016; Aulbach and Smart, 2023). EPMA data were used for the major elements and LA-ICP-MS data for the trace elements. The calculations were performed taking into account the specific gravities (Deer et al., 2013) of the primary minerals (3.3 g/cm³ for clinopyroxene, 3.6 g/cm³ for garnet, 3.3 g/cm³ for kyanite, 4.04 g/cm³ for rutile, 4.02 g/cm³ for corundum) and for bulk eclogite (3.45 g/cm^3) . The specific gravity was utilized to calculate the relative weight proportions. Given that the variability of the average chemical compositions of the samples studied is low (generally <10% RSD), the major source of uncertainty in the bulk-rock reconstructions stems from the estimates of the modal mineral abundances. For bimineralic kyanite-free samples, we assumed 49 vol.% for clinopyroxene and garnet each, and 1 vol.% for rutile and corundum each. For kyanite-bearing eclogite nodules, we assumed 44 vol.% for clinopyroxene and garnet each, 1 vol.% for rutile and corundum each, and 10 vol.% for kyanite. Bulk eclogite reconstruction results for the major elements (normalized to 100 wt.%) and the trace elements (in ppm) are given in Supp. Table 1. The results are plotted in Fig. 7a-c to classify the eclogites (see Discussion), which show low MgO, high Na₂O, and Eu/Eu* anomalies.

As shown in Figure 8a, the estimated modal abundance of rutile has a significant effect on the overall budget of Ti, which in combination with the concentration of V, provides an estimate for fO_2 (e.g., Aulbach and Stachel, 2022). Based on previous analyses of mantle eclogites and our own petrographic observations, we conclude that a modal rutile abundance between 0.5 and 1 vol.% is realistic. Regardless of the exact choice of rutile abundance, our KL2 eclogite samples fall just below FMQ-2 in Figure 8a. In terms of Lu/Gd versus Ce/Yb ratios, our reconstructed bulk eclogite compositions agree with mantle eclogites from cratons worldwide, partly overlapping with metasomatized eclogites (Fig. 8b). In terms of V/Sc versus MgO systematics, our reconstructed bulk eclogite compositions suggest a fO_2 value of around FMQ-2 (Fig. 8c), similar to the Ti-V based estimate (Fig. 8a).

4 Thermobarometer choice

Nimis (2022) presented an overview of geothermobarometers applicable to eclogites. Older thermometer calibrations based on garnet-clinopyroxene Fe-Mg exchange developed by Ellis and Green (1979) and Powell (1985) have been demonstrated to be unreliable because the effect of Ca was not considered (e.g., Brey and Köhler, 1990; Nimis and Grütter, 2010). Pressure estimates by Beyer et al. (2015) for eclogite xenoliths show systematically lower values (by ~10 kbar) when compared to pressures calculated using orthopyroxene-garnet assemblages at the same temperatures (Nimis, 2022). Furthermore, due to the high sensitivity of the equation to slight inaccuracies in clinopyroxene major element compositions, the Beyer et al. (2015) eclogite barometer is prone to produce significant errors.

In this study, temperatures were determined for six KL2 eclogite xenoliths (at an assumed pressure of 40 kbar) based on Fe-Mg exchange between garnet and clinopyroxene (Krogh, 1988;

 $T_{\rm K}$). Temperatures for KL2B eclogite could not be calculated due to the complete alteration of clinopyroxene. In addition, published data for 34 KL2 eclogite nodules were reprocessed with the same thermometer equation. The eclogite equilibrium pressures (P_{HC40}) were calculated by subsequently projecting the obtained temperatures onto the regional cratonic geotherm of 40 mW/m^2 (after Shaikh et al., 2020). Temperatures (T_K) obtained for an assumed pressure of 40 kbar range from 945 to 1180 °C. Pressures obtained after geotherm projection range from 38 to 50 kbar (Fig. 9a), equivalent to 120-160 km depths. Alternative temperature estimates calculated after Ellis and Green (T_{EG}; 1979), Powell (T_{Po}; 1985), Ai (T_{Ai}; 1994), Ganguly et al. (T_{Gn}; 1996), Krogh Ravna (T_{KR} ; 2000), and Nakamura (T_{Nk} ; 2009) yielded very similar results for the KL2 eclogite xenoliths (Supp. Table 1). Overall, our P-T results compare well to those obtained by Patel et al. (2009) and Dongre et al. (2015) (i.e., 36–54 kbar).

The equilibration pressures (P_{HC40}) for peridotitic garnet xenocrysts were calculated using Ni-in-garnet temperatures (970-1217 °C) (Canil, 1999) projected onto the regional 40 mW/m² geotherm (Fig. 9b). Pressure readings for two garnet xenocrysts (P9G51A, P9G52C) were corrected using a single-pyroxene thermobarometer applied to Cr-diopside inclusions (after Nimis and Taylor, 2000). The peridotitic garnet xenocrysts studied equilibrated at 40 to 53 kbar corresponding to ~125-170 km depth, similar to the depths of origin of the KL2 eclogite xenoliths.

48 314 Measured ferric iron contents in garnet, and calculated fO₂

Table 1 and Table 2 list the Fe³⁺/ Σ Fe ratios for garnets from KL2 eclogite xenoliths and peridotitic garnet xenocrysts, respectively. Fe³⁺/ Σ Fe ratios for our eclogitic garnets vary from 0.02 to 0.05 (± 0.01) , which is within the global range of mantle-derived eclogitic garnets (0.02–0.09; Stagno et al., 2015; Smart et al., 2017, 2021a, 2021b; Aulbach et al., 2019, 2022). The Fe³⁺/ Σ Fe ratios for

our peridotitic garnet xenocrysts range from 0.02 to 0.10 (±0.01), extending to higher values than
those measured for KL2 eclogites.

We calculated $\Delta \log fO_2$ values (relative to the FMQ buffer) for the KL2 eclogite xenoliths using the oxybarometer of Stagno et al. (2015), which was calibrated using coesite/quartz-bearing eclogites. By applying the calculated pressure-temperature values and measured chemical compositions for garnet and clinopyroxene, the KL2 eclogites yielded $\Delta \log O_2$ values between FMQ-2.5 and FMQ-0.9, with uncertainties of $\pm 0.6 \log$ units for corundum-free eclogites (Fig. 10). These redox compositions fall closer to the carbonate stability field compared with the fO_2 systematics of other xenolithic eclogite suites from cratons worldwide (Stagno et al., 2015; Burness et al., 2020; Smart et al., 2021a, b; Aulbach et al., 2022). The above fO₂ range for the KL2 eclogite xenoliths is in good agreement with our less-precise reconstructed bulk eclogite fO_2 estimate of $\langle FMQ-2 \rangle$ (see Fig. 8). As per Smart et al. (2021), a correction is required for the fO_2 calculations for corundum-bearing silica-undersaturated eclogites. The correction involves the incorporation of silica activity using corundum + SiO_2 (coesite) = kyanite equilibrium. Application of this correction to the KL2D and KL2F corundum-bearing silica-undersaturated eclogite xenoliths shifts their $\Delta \log fO_2$ values by 1.5 log units toward more reduced redox compositions (FMQ-3.9 and FMQ-3.6), more similar to eclogite xenoliths from the Kaapvaal craton root (Fig. 10). There is a weak correlation between $\Delta \log fO_2$ values and contents of MgO and CaO for garnet, with $\Delta \log fO_2$ increasing with decreasing MgO at increasing CaO (Fig. 11). No correlation is observed between the trace element concentrations of eclogitic garnets and their $\Delta \log fO_2$ values.

For the peridotitic garnet xenocrysts, we used the Stagno et al. (2013) equation, which requires input for pressure and temperature (see above), as well as the Fe# values for coexisting olivine and orthopyroxene. The Fe# values for orthopyroxene in equilibrium with garnet were

calculated using Equation 7 in Ryan et al. (1996) and Equation 11 in Harley (1984). The Fe# values for olivine in equilibrium with garnet were calculated by inverting the garnet-olivine thermometer of O'Neill and Wood (1979), as demonstrated by Gaul et al. (2000). The obtained Fe# values range from 0.06 to 0.08 for olivine and 0.03 to 0.04 for orthopyroxene (variance is <1%; Gaul et al., 2000; Ryan et al., 1996), corresponding to Mg# values of 0.92-0.94 and 0.96-0.97, respectively. These inferred values are similar to the Mg# values reported for olivine and orthopyroxene from rare peridotite xenoliths from the Dharwar craton root (0.90-0.94 for olivine; 0.90-0.95 for orthopyroxene; Ganguly and Bhattacharya, 1987; Nehru and Reddy, 1989; Pattnaik et al., 2020), and also overlap with the compositions of mantle-derived olivine xenocrysts from Wajrakarur kimberlites (0.90-0.94 Mg#; Shaikh et al., 2019, 2018). This complex procedure enabled us to calculate $\Delta \log fO_2$ values between FMQ-4.5 and FMQ-2.6 (±0.9) for the peridotitic garnet xenocrysts (Fig. 10).

Discussion

356 Nomenclature and origin of the KL2 eclogites

Mantle-derived eclogite xenoliths are generally thought to be formed by either (i) subduction of gabbroic or basaltic oceanic crust (e.g., Taylor and Neal, 1989; Barth et al., 2001; Jacob, 2004), (ii) emplacement of basaltic melt at depths within cratonic mantle lithosphere with high-pressure cumulate formation (e.g., Viljoen et al., 1996; Barth et al., 2002), or (iii) extraction of tonalitic melt from basaltic oceanic crust during subduction leaving behind an eclogitic residue (e.g., Jacob and Foley, 1999; Barth et al., 2002; Tappe et al., 2011a). Each of these genetic types of cratonic eclogites is supported by textural, mineralogical, geochemical, and isotopic evidence (Aulbach and Smart, 2023). Accordingly, mantle-derived eclogites have been classified into several types based

on major and trace element compositions of garnet and clinopyroxene (McCandless and Gurney, 1989; Taylor and Neal, 1989; Jacob et al., 2009) and reconstructed bulk compositions (e.g., Aulbach and Jacob, 2016; Smart et al., 2017).

The KL2 eclogites studied here contain clinopyroxene that is enriched in Na₂O (typically 4-7 wt.%) and has moderate MgO contents (typically 6-11 wt.%). These clinopyroxene compositions are characteristic for Type B and Type C eclogites of Taylor and Neal (1989; Fig. 5a, b), interpreted to originate from subducted ancient oceanic crust protoliths. The relatively low K_2O contents of clinopyroxene (<0.1 wt.%, or below detection limit), together with the low Na₂O (0.15 wt.%) and TiO₂ (0.5 wt.%) contents in garnets (Supp. Table 1), are similar to the mineral compositions of Group II non-metasomatized eclogite xenoliths described by McCandless and Gurney (1989). KL2 eclogites contain garnets with moderate Mg# values (typically between 0.4-0.7) and high Ca# values (0.2-0.6) (Fig. 5; 7), similar to the high-Ca eclogites as defined by Aulbach and Jacob (2016). KL2 eclogitic garnets and clinopyroxenes exhibit Eu anomalies in their REE patterns, which are also mirrored in the reconstructed bulk eclogite compositions (Fig. 5c, d; 7c). The reconstructed bulk eclogite compositions are relatively high in Na₂O (2-3.5 wt.%; Fig. 7a). These geochemical features indicate that the protolith of the KL2 eclogite xenoliths was a plagioclase-rich gabbroic lithology as part of ancient oceanic crust. This finding is consistent with the model by Dongre et al. (2015), who presented evidence for low-temperature alteration of the eclogite protolith near the Earth's surface based on significantly elevated δ^{18} O values for garnet (up to +7.8‰) from some KL2 eclogite xenoliths. However, features such as interlocking garnetclinopyroxene textures, rutile blebs in clinopyroxene and garnet, the presence of corundum and Na₂O-poor garnet and K₂O-poor clinopyroxene, and Eu-anomalies are more similar to Type II_L eclogite xenoliths from the Roberts Victor orangeite in South Africa (Hardman et al., 2021). These

eclogite nodules are considered to represent cumulates of magmas sourced from a depleted mantle reservoir that had undergone prior extraction of MORB-like melts (Hardman et al., 2021), but subducted oceanic crustal origins have also been put forward for the classic Roberts Victor eclogite xenolith suite (e.g., Jacob et al., 2005). For KL2 eclogites, Patel et al. (2009) proposed a high-pressure cumulate origin, which is supported by textural features such as graded layering and garnet necklaces as well as garnet-kyanite clusters, plus evidence from mineral compositions such as similar clinopyroxene geochemistry in the eclogite xenoliths and cumulate pyroxenites. In contrast, Griffin et al. (2009) suggested KL2 eclogite formation by solidification of mafic melts near the base of the SCLM. Clearly, there are 'eclogites and eclogites', and considering the scope of our study (mantle redox), we refrain from further speculations as to the nature and origin of the KL2 eclogite protolith.

Redox state of KL2 eclogite xenoliths

Using compositions of eclogitic garnets from the southern region of the WKF (Kalyandurga kimberlite cluster), we calculated $\Delta \log fO_2$ values between FMQ-2.5 and FMQ-0.9 (±0.6), with two eclogite nodules (KL2C and KL2E) recording more reduced oxidation states (FMQ-3.9 and FMQ-3.6). On the basis of these $\Delta \log fO_2$ values and their depth distribution, the KL2 eclogites straddle the boundary between diamond and carbonate stability fields in terms of carbon speciation (Fig. 10). The peridotitic garnet xenocrysts from the P9 and P10 kimberlite bodies of the northern region of the WKF (Wajrakarur-Lattavaram kimberlite cluster) yielded $\Delta \log fO_2$ values ranging from FMQ-4.5 to FMQ-2.6 (± 0.9), at the low end of eclogitic $\Delta \log fO_2$ values determined here. This observation suggests that the majority of KL2 mantle eclogites have experienced more oxidizing conditions compared to cratonic peridotites at similar depths prior to entrainment into

that overlap with those of many kimberlite-borne eclogite nodules from cratons worldwide (FMQ-1 to FMQ-5; Stagno et al., 2015; Burness et al., 2020; Smart et al., 2021a, b; Aulbach et al., 2022). Depth related redox heterogeneity has been observed for eclogites from different intervals within cratonic SCLM worldwide (e.g., Burness et al., 2020). For example, Kaapvaal cratonic eclogites from mid-lithospheric depths show more reducing redox conditions (ΔlogfO₂ = FMQ-5.3 to FMQ-3.3) compared to eclogites from the lowermost lithosphere (FMQ-3.9 to FMQ-1.5) (Burness et al., 2020). However, this apparent relationship was not observed in the data produced for other localities on the Kaapvaal craton or elsewhere (Smart et al., 2017, 2021a, b; Aulbach et al., 2022). Based on our data for KL2 eclogites, we observe notable variations in their oxidation state over a relatively narrow depth interval, but large-scale redox variations with depths cannot be discerned. Further work is required to explore such a potential relationship. We can conclude, however, that the KL2 eclogites present a suitable diamond host rock given their P-T-fO₂ systematics (Fig. 10). This conclusion is supported by the diamondiferous nature of the KL2 kimberlite body and mineral inclusion data from the WKF (Ravi et al., 2013).

erupting kimberlite magmas at ca. 1.1 Ga. The KL2 eclogite xenoliths studied have $\Delta \log fO_2$ values

Redox state of mantle peridotites and metasomatism of the Dharwar craton root

Garnet xenocrysts of peridotitic affinity from the P9 and P10 kimberlites revealed a wide range of $\Delta \log fO_2$ values between FMQ-4.5 and FMQ-2.6 (±0.9 log units). Griffin et al. (2009) and Shaikh et al. (2020) presented extensive trace element data for garnet and clinopyroxene xenocrysts from Wajrakarur kimberlites, which allows us to explore any possible relationship between melt metasomatism of the SCLM and its redox composition. Shaikh et al. (2020) proposed the presence of a mid-lithospheric discontinuity between ~145 and 160 km depth.

The Dharwar craton root is strongly metasomatized at the very bottom, a trend known from many lithospheric mantle profiles beneath cratons worldwide (e.g., Griffin et al., 1999b; Kopylova et al., 1999; Kargin et al., 2016; Smart et al., 2017; Aulbach et al., 2007, 2013, 2017; Tappe et al., 2021). However, unlike the Kaapvaal craton, the Dharwar SCLM shows concomitant enrichment in Zr and Y in garnet (Shaikh et al., 2020). This feature has been interpreted to result from high-temperature mantle metasomatism by mixed silicate-carbonate melts (Shaikh et al., 2020) or by basaltic melts (Griffin et al., 2009). Although, the Ti/Eu versus Zr/Hf systematics of peridotitic garnets, suggest a kimberlitic to carbonatitic nature of the metasomatic agents (Shaikh et al., 2020). According to global datasets, the oxidation state of mantle peridotite varies laterally and vertically across the SCLM (e.g., Woodland and Koch, 2003; Creighton et al., 2009; Yaxley et al., 2017; Stagno et al., 2013; Tappe et al., 2021). For example, in the garnet stability field, the Kaapvaal SCLM shows a progressively more reducing trend with increasing depth ($\Delta \log fO_2$ FMQ-2 at 110 km to FMQ-4 at 210 km; Luth, 1990; Woodland and Koch, 2003; Creighton et al., 2009, 2010; Tappe et al., 2021). This crystal chemistry-controlled depth- fO_2 trend has been masked by oxidative metasomatism beneath the Kimberley area on the Kaapvaal craton, where many peridotite xenoliths are offset to higher-than-expected fO₂ values (Creighton et al., 2009; Hanger et al., 2015). This is also consistent with observed Fe³⁺ variations in metasomatic growth zones on mantle-derived garnet crystals (McCammon et al., 2001). The peridotitic garnet xenocrysts from the WKF studied here show variations in $\Delta \log fO_2$ and they fall within the Kaapvaal peridotite P- fO_2 array, with a few samples showing slightly higher $\Delta \log fO_2$ values approaching the carbonate stability field (Fig. 10). The Fe³⁺/ Σ Fe ratios measured for trace element enriched garnet xenocrysts with normal REE patterns show a positive correlation with incompatible trace element concentrations (Ti, Zr, Y, V; Fig. 12), possibly indicating a metasomatic control on cratonic mantle

redox (Creighton et al., 2009). These garnet xenocrysts were sampled from $\sim 125-160$ km depths, an SCLM interval that was identified as a modally metasomatized mid-lithospheric discontinuity by Shaikh et al. (2020). Overall, the link between carbonated silicate melt metasomatism and mantle redox in the Dharwar craton root may be responsible for local diamond destruction, as reported for other cratons worldwide (Creighton et al., 2009; Fedortchouk et al., 2019).

Linking mantle redox and the diversity of 1.1 Ga kimberlitic magmatism in India

The WKF comprises a diverse range of diamondiferous magma types, including archetypal kimberlites, lamproites, orangeites, and ultramafic lamprophyres (see compilation in Shaikh et al., 2017; Pandey and Chalapathi Rao, 2020). To date, 48 minor intrusions of exclusively Mesoproterozoic age (ca. 1.1 Ga) are known to occur within a relatively small area that is 80×70 km in size. The coeval emplacement of diverse volatile-rich ultramafic magma types reflects prominent mineralogical heterogeneity in the Dharwar craton root (e.g., Sarkar et al., 2021), as was also demonstrated for cratonic regions and their primitive potassic magmatism on either side of the North Atlantic (Tappe et al., 2008, 2011b; Dalton et al., 2019).

The correlation between metasomatic overprint and redox state of the Dharwar craton root, as demonstrated in this study, appears to have important implications for the petrogeneses of kimberlites, lamproites and closely related primitive potassic rocks. Progressive oxidative metasomatism along conduits in the SCLM was caused by infiltrating carbonated silicate melts that probably originated from low-degree partial melting of the underlying convecting mantle including its recycled crustal components (e.g., Nowell et al., 2004; Malkovets et al., 2007). The resultant fO_2 heterogeneities in the SCLM allowed for stabilization of distinctly different metasomatic assemblages that may be dominated by either amphiboles, micas, or carbonates

(Foley et al., 1986; Foley, 2011; Tappe et al., 2008; Yaxley et al., 2017). The highly variable nature of SCLM metasomes, and also local lack of metasomes, may explain the close spatiotemporal association of diverse types of primitive K-rich magmas (lamproites, orangeites, ultramafic lamprophyres) and kimberlites in the WKF.

Shaikh et al. (2017) suggested a model for the origin of various types of diamond-bearing magmas in the WKF, following concepts developed in Tappe et al. (2008, 2011b), where carbonated silicate melts from the asthenosphere interacted with variably metasomatized lithologies of the SCLM to produce the diversity of ca. 1.1 Ga old volatile-rich ultramafic rocks known from southern India. According to this model, metasomatic phlogopite-carbonate dominated veins were reactivated by infiltrating asthenosphere-derived melts (proto-kimberlitic melts), which gave rise to ultramafic lamprophyre, orangeite and lamproite magmas as a function of increasingly higher proportions of micas in the cratonic mantle source (Shaikh et al., 2017). Sarkar et al. (2021) compared the compositions of magmatic and xenocrystic olivine populations from Wajrakarur kimberlites and lamproites. These authors suggested that both magma types have a common asthenospheric mantle source, with lamproites revealing a larger extent of assimilation of Fe-rich lithospheric metasomes.

On the 'overabundance' of eclogite xenoliths in the KL2 kimberlite

Despite the general xenolith deficiency, the WKF minor intrusions reveal contrasting mantle sampling patterns. For example, the KL2 kimberlite is marked by an overabundance of eclogite xenoliths, whereas kimberlites and related rocks from the Wajrakarur-Lattavaram cluster contain more peridotite xenoliths (Rao et al., 2001; Patel et al., 2006). This apparent bias is also mirrored

by the diamond occurrences (Ravi et al., 2013) and distributions of eclogitic versus peridotitic garnet xenocrysts (Griffin et al., 2009; Shaikh et al., 2020).

- Although kimberlites and related rocks with an overabundance of eclogite xenoliths are very rare, they occur on most cratons worldwide such as the Kaapvaal craton (e.g., Roberts Victor, Bellsbank), the Slave craton (e.g., Jericho, Voyageur), the North Atlantic craton (e.g., Nunatak 1390) and the Siberian craton (e.g., Zagadochnaya) (Schulze, 1989; Jacob, 2004; Smart et al., 2009, 2017, 2021a; Tappe et al., 2011a; Kopylova et al., 2016; Hardman et al., 2021; Aulbach and Smart, 2023). Eclogite xenolith overabundance has been linked to localized enrichment of the peridotite-dominated cratonic mantle column with eclogite components (e.g., Nixon and Davies, 1987) or biased preservation of xenoliths during entrainment into kimberlitic magmas (e.g., Schulze, 1989). Kopylova et al. (2016) inferred lateral continuity of eclogites within the Slave craton SCLM, where an eclogite-rich layer may represent the remnant of an imbricated oceanic lithosphere slab. The abundance of eclogite components has great economic significance because they contribute up to 30% of lithospheric diamonds in global statistics (Stachel and Luth, 2015), which can be even higher on a regional scale. The overall abundance of eclogitic components in peridotite-dominated SCLM has been estimated at <1 vol.% (Schulze, 1989); however, higher abundances of up to 4-10 vol.% have been suggested for the Slave craton (Griffin et al., 1999; Kopylova et al., 2016). The combined petrological-geophysical approach by Garber et al. (2018) devised an even higher eclogite abundance within the SCLM beneath cratons (up to 20 vol.%). Regardless of the exact amount of eclogites within the cratonic mantle lithosphere, the KL2 occurrence on the Eastern Dharwar craton adds to the kimberlite localities that have a somewhat inexplicable abundance of xenolithic eclogites, which may influence the redox distribution within the local SCLM and, by inference, also its volatile element and diamond inventory.

Summary and Conclusions

The Fe³⁺/ Σ Fe compositions of garnets were determined by the EPMA flank method for six eclogite xenoliths from the KL2 kimberlite and fourteen peridotite-derived xenocrysts from the P9 and P10 kimberlites on the Eastern Dharwar craton in southern India. KL2 eclogites are characterized by low $\Delta \log fO_2$ values between FMQ-3.9 and FMQ-0.9 (±0.6), which suggests that they could have acted as diamond host rocks within the deeper Dharwar craton root. The peridotite-derived garnet xenocrysts reveal a wide range of $\Delta \log fO_2$ values between FMQ-4.5 and FMQ-2.6 (±0.9), firmly within the well-studied Kaapvaal lithospheric mantle array. At mid-lithospheric depths beneath the Dharwar craton, redox compositions of the garnet xenocrysts correlate with geochemical proxies for melt-rock interactions, which provides evidence for the operation of oxidative mantle metasomatism that was likely diamond-destructive. Strong redox variability at relatively confined SCLM intervals would promote stability of diverse volatile-rich metasomatic mineral assemblages (e.g., roles of carbonates versus phlogopite and amphibole), which helps to explain the spatiotemporal association of ca. 1.1 Ga kimberlites, lamproites, orangeites and ultramafic lamprophyres in southern India.

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Fig. 2. Scanned thin section images of KL2 eclogite nodules.

Fig. 3. BSE images of KL2 eclogite nodules showing mineral grain relations and the extensive alteration of primary and matrix minerals. (a) Highly fractured garnet in sample KL2A1 showing alteration to hydro-garnet along margins. (b) Sample KL2B1 shows garnet altered to carbonate and chlorite along fractures and no preservation of omphacite and kyanite. (c) Highly fractured and altered omphacite in sample KL2A1. (d) Kyanite blades in sample KL2C3, with inclusions of garnet and omphacite, partially altered to celsian. (e) Kyanite blade in sample KL2G1 with inclusions of garnet and omphacite. (f) Interlocked kyanite blades in sample KL2F2 with an omphacite inclusion.

Fig. 4. BSE images of KL2 eclogite nodules with corundum associated with kyanite (a) and a rutile bleb (b).

Fig. 5. Major and trace element (REE) plots for garnet (a, b, c) and clinopyroxene (b, d) from KL2 eclogites. The fields in figure (a) are after Aulbach and Jacob (2016) and in (b) after McCandless and Gurney (1989). The A, B, and C fields in the inset figure are after Taylor and Neal (1989).

Fig. 6. Cr₂O₃ versus CaO wt.% plot (a) and chondrite normalized REE plot (b; after (Sun and McDonough, 1989) for garnet xenocrysts from P9 and P10 intrusions of the WKF.

Fig. 7. Reconstructed bulk-rock composition diagrams for classifying KL2 eclogite nodules. Fields in figures (a) and (b) are after Aulbach and Jacob (2016), and primitive mantle normalization in

(c) is after (Sun and McDonough, 1989). Published KL2 eclogite data are from Dongre et al. (2015). Symbol sizes in (a) and (b) are larger than the 2σ uncertainties.

Fig. 8. Trace element variations diagrams based on reconstructed bulk-rock compositions for eclogite nodules. Note that reconstructed bulk-rock compositions indicate fO_2 values below FMQ-2. Figures (a) Ti vs. Vi/Ti*100, (b) Ce/Yb vs. Lu/Gd, and (c) MgO vs. V/Sc are after Wang et al. (2019) and Aulbach and Viljoen (2015). Symbol sizes are larger than the 2σ uncertainties.

Fig. 9. Projections of equilibration temperatures calculated for KL2 eclogite nodules (a; after Krogh, 1988) and P9-P10 garnet xenocrysts (b; after Canil, 1999). Published KL2 data from Patel et al. (2006) and Dongre et al. (2015). The conductive model geotherm (after Hasterok and Chapman, 2011) for the Wajrakarur lithosphere is from Shaikh et al. (2020). The diamond-graphite transition is after Day et al. (2012).

Fig. 10. $\Delta \log fO_2$ (FMQ) values calculated for KL2 eclogitic garnets (after Stagno et al., 2015) and P9 and P10 peridotitic garnet xenocrysts (after Stagno et al., 2013) plotted against the estimated pressure. The field for Kaapvaal peridotites is after Woodland and Koch (2003) and Creighton et al. (2009), enriched peridotites from Slave craton after Yaxley et al. (2017) and graphite-diamondcarbonate melt transition lines after Stagno et al. (2015). Updated Kaapvaal data (small open circles) are after Tappe et al. (2021).

Fig. 11. $\Delta \log fO_2$ (FMQ) versus MgO and CaO (in wt%) for KL2 eclogitic garnets from Wajrakarur kimberlites.

Fig. 12. Fe³⁺/ΣFe ratios plotted against TiO₂ (a), Y (b), Zr (c), and V (d) for peridotitic garnet
xenocrysts from the P9 and P10 kimberlite bodies at Wajrakarur. Kaapvaal craton xenolith data
are taken from Creighton et al. (2009).




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Figure 5

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00T∗!T/V



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Sample	Minerals	T _{KR88} (°C) ^{\$}	$P_{KR88} (GPa)^{\$\$}$	Grt $Fe^{3+}/\Sigma Fe^{\psi}$	$logO_2 (\Delta FMQ)^{\psi\psi}$
KL2A	grt, omp, rt	1100	4.7	0.02	-2.5
KL2C	grt, omp, ky	1160	5.0	0.05	-0.9
KL2D	grt, omp, ky, crn	1120	4.8	0.02	-3.9
KL2E	grt, omp, ky	1090	4.7	0.05	-1.0
KL2F	grt, omp, ky, crn	1130	4.8	0.02	-3.6
KL2G	grt, omp, ky	1125	4.8	0.02	-2.3

Table 1. Mineral assemblages, temperature, pressure, garnet $Fe^{3+}/\Sigma Fe$, and oxygen fugacity of the KL2 eclogites.

[§]Temperatures (T_{KR88}) were calculated iteratively using temperatures obtained from the Krogh (1988) thermometer with pressures (P_{HC40})^{§§} calculated from the 40 mW/m² conductive geotherm from Hasterok and Chapman (2011). The discrepancies for T_{KR88} are mostly within ±50 °C; which poses an uncertainty in iteratively calculated P_{HC40} around ±0.3 GPa.

 $^{\psi}$ Fe³⁺/ Σ Fe measured using the EPMA flank method after Höfer et al. (1994) and Höfer and Brey (2007) with an uncertainty of ± 0.01 .

 $^{\psi\psi}$ Oxygen fugacities were calculated using the oxybarometer of Stagno et al. (2015) with T_{KR88}–P_{HC40}. The uncertainties for fO_2 are mostly ~0.6 log units.

Sample	T _{C99} (°C) ^{\$}	P _{HC40} (GPa) ^{\$\$}	Grt $Fe^{3+}/\Sigma Fe^{\psi}$	Fe# Opx ^{ψψ}	Fe# Ol [⊕]	$\log O_2 \left(\Delta FMQ\right)^{\Phi\Phi}$
P9G47C	1025	4.3	0.03	0.040	0.081	-4.5
P9G51A	1037	5.1	0.10	0.039	0.077	-2.9
P9G52A	1023	4.3	0.08	0.038	0.076	-2.7
P9G52C	1102	4.7	0.08	0.038	0.076	-3.1
P10G3D	1030	4.1	0.04	0.040	0.081	-3.9
P10G10B	980	4.0	0.04	0.032	0.067	-3.5
P10G10D	1192	5.2	0.10	0.035	0.068	-3.0
P10G13A	983	4.0	0.07	0.035	0.074	-2.6
P10G13B	1013	4.2	0.04	0.041	0.082	-3.8
P10G13C	975	4.0	0.05	0.032	0.067	-3.1
P10G13D	1193	5.2	0.06	0.037	0.070	-3.8
P10G19C	1217	5.3	0.07	0.038	0.070	-3.6
P10G19D	970	4.0	0.03	0.033	0.069	-4.2
P10G28D	998	4.7	0.05	0.033	0.063	-3.7

Table 2. Temperature, pressure, and $Fe^{3+}/\Sigma Fe$ in garnet, $Fe\# Opx^{\psi} = Fe^{2+}/(Fe^{2+}+Mg)$ in orthopyroxene, $Fe\# Ol^{\psi\psi} = Fe^{2+}/(Fe^{2+}+Mg)$ in olivine and oxygen fugacity of the peridotitic garnet xenocrysts from Wajrakarur.

[§]Temperatures (T_{C99}) calculated iteratively using temperatures obtained from the Canil (1999) thermometer with pressures (P_{HC40})^{§§} calculated from the 40 mW/m² conductive geotherm from Hasterok and Chapman (2011). The discrepancies for T_{C99} are mostly within ± 50 °C; which poses an uncertainty in iteratively calculated P_{HC40} around ± 0.3 GPa.

 Ψ Fe³⁺/ Σ Fe measured using the EPMA flank method after Höfer et al. (1994) and Höfer and Brey (2007). The uncertainty is ±0.01.

^{ww}Fe# in orthopyroxene in equilibrium with garnet calculated using Equation 7 in Ryan et al. (1996) and Equation 11 in Harley (1984). The assumed uncertainty is below 5% (Gaul et al., 2000; Ryan et al., 1996).

⁶Olivine in equilibrium with garnet calculated from the garnet-olivine thermometer of O'Neill and Wood (1979) using the algorithm by Gaul et al. (2000). The assumed uncertainty is below 5% (Gaul et al., 2000; Ryan et al., 1996).

 $^{\Phi\Phi}$ Oxygen fugacities calculated using the oxybarometer of Stagno et al. (2013). The uncertainty is mostly ~0.9 log units.

Redox state of the Dharwar craton root as inferred from eclogite and peridotite sourced mantle cargo, with implications for kimberlite and lamproite magma formation

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Appendix 1

Details of the analytical techniques, (a) EPMA and (b) LA-ICP-MS

(a) EPMA calibration settings for major elements of olivine

Major elements were measured at an acceleration voltage of 20 kV and a beam current of 20 nA with a beam diameter of 1 μ m. Data was reduced by using the X-PHI method (Merlet, 1992, 1994). The following calibration was used.

Element	Crystal	Peak Time	Backgrou	Background	Standard	Standard
and line		(sec)	nd (-ve)	(+ve)		intensity
						(cps/nA)
Na K_{α}	TAP	40	-1200	660	Jadeite	34
Mg Ka	TAP	20	-1050	1000	Olivine	120
Al Ka	TAP	34	-750	1000	Almandine	287
Si Ka	TAP	26	-1760	2250	Diopside	287
K Ka	PET	24	-1100	1000	Orthoclase	39
Ca Ka	PET	20	-700	760	Wollastonite	99
Ti Ka	PET	16	-500	700	TiO ₂	124
Cr Ka	LLIF	18	-1000	900	CrO	164
Mn Ka	LLIF	16	-1200	1400	Rhodonite	127

Table 1. Calibration settings microprobe measurements

Fe Ka	LLIF	14	-880	800	Hematite	142
Ni Ka	LLIF	12	-1600	1000	NiO	130

(Pulse height analysis mode was integral. cps/nA = counts per second per nano ampere) Counting time for the background was half of the peak time.

(b) LA-ICP-MS: trace elements in clinopyroxene and garnet

Trace elements of clinopyroxene, garnet and kyanite in eclogite nodules were measured at Spectrum, University of Johannesburg, using a 193 nm ArF RESOlution SE excimer laser system (Australian Scientific Instruments, Fyshwick) enabled with a Thermo Scientific iCAP RQ ICPMS. Laser sampling was done with the SE155 dual-volume ablation cell (Laurin Technic, Canberra, Australia) that delivers the ablated material in a He-Ar gas mixture. The gas settings for ICP-MS were 14 L/min cooling gas, 0.80 L/min auxiliary gas and 1.2 L/min nebulizer gas with a flow rate of He laser carrier gas at 0.35 L/min, and a minor amount of N₂ additional gas at a flow rate of 0.001 L/min. Laser spots were of 80 µm diameter with 6 mJ laser energy at 25% attenuation to obtain an on-sample fluence of 2 J/cm². The repetition rate was 10 Hz. The grain surface before each spot analysis by two laser pulses. The background signal was measured for 15 seconds at the beginning of each measurement and ablation signal was measured for 50 to 60 seconds. Tuning of the LA-ICPMS system for sensitivity and interferences was done using line scans on NIST 612 glass for ⁶Li, ⁵⁹Co and ²⁰⁸Pb. Samples were measured using standard bracketing, with ²⁹Si in NIST612 used as a calibration standard. The primary standard (NIST612) was analyzed twice at the start and twice at the end of each experiment or sequence, as well as once after every 25 sample measurements. Two to four secondary standard measurements were also included at these 25 sample measurement intervals for quality control purpose. Raw data were processed using the iCAP Qtegra software version 2.10.3324.62. The following masses were measured: 7Li, 23Na, 24Mg, 25Mg, 26Mg, 27Al, 29Si, 31P, 43Ca, 45Sc, 47Ti, 48Ca, 49Ti, 51V, 53Cr, 55Mn, 57Fe, 59Co, 60Ni, 63Cu, 65Cu, 66Zn, 68Zn, 69Ga, 71Ga, 75As, 85Rb, 88Sr, 89Y, 90Zr, 93Nb, 96Mo, 118Sn, 133Cs, 137Ba, 139La, 140Ce, 141Pr, 146Nd, 147Sm, 153Eu, 157Gd, 159Tb, 163Dy, 165Ho, 166Er, 169Tm, 172Yb, 175Lu, 178Hf, 181Ta, 182W, 208Pb, 232Th, 238U. Isotope ²⁹Si was used for internal standardization. The dwell times of all isotopes are provided in Table 2 given below. The primary standard used for calibration was NIST 612 (Jochum et al., 2011) and secondary standards used to evaluate the accuracy and precision were USGS natural glasses BHVO2G, BCR-2G and GHR1 (Gao et al., 2002) and MongOl-sh11-2 (Batanova et al., 2019).

Most of the trace element measurements were comparable within 10%, while some were within 15%, compared to accepted values (Supp. Table 1). Data reduction was performed using the Glitter software version 4.4.4 (<u>http://www.glitter-gemoc.com/</u>).

Identifier	Dwell time (s)	Channels	Spacing (u)	Resolution
7Li	0.03	1	0.1	Normal
23Na	0.01	1	0.1	Normal
24Mg	0.01	1	0.1	Normal
25Mg	0.01	1	0.1	Normal
26Mg	0.01	1	0.1	Normal
27A1	0.01	1	0.1	Normal
29Si	0.01	1	0.1	Normal
31P	0.01	1	0.1	Normal
43Ca	0.01	1	0.1	Normal
45Sc	0.03	1	0.1	Normal
47Ti	0.01	1	0.1	Normal
48Ca	0.01	1	0.1	Normal
49Ti	0.01	1	0.1	Normal
51V	0.01	1	0.1	Normal
53Cr	0.01	1	0.1	Normal
55Mn	0.01	1	0.1	Normal
57Fe	0.01	1	0.1	Normal
59Co	0.01	1	0.1	Normal
60Ni	0.03	1	0.1	Normal
63Cu	0.01	1	0.1	Normal
65Cu	0.03	1	0.1	Normal
66Zn	0.03	1	0.1	Normal
68Zn	0.03	1	0.1	Normal
69Ga	0.03	1	0.1	Normal
71Ga	0.01	1	0.1	Normal
75As	0.01	1	0.1	Normal
85Rb	0.01	1	0.1	Normal
88Sr	0.03	1	0.1	Normal
89Y	0.03	1	0.1	Normal
90Zr	0.03	1	0.1	Normal
93Nb	0.03	1	0.1	Normal
96Mo	0.01	1	0.1	Normal
118Sn	0.01	1	0.1	Normal

Table 2. Dwell times of isotopes measured during the LA-ICPMS analysis.

133Cs	0.02	1	0.1	Normal
137Ba	0.01	1	0.1	Normal
139La	0.03	1	0.1	Normal
140Ce	0.03	1	0.1	Normal
141Pr	0.03	1	0.1	Normal
146Nd	0.01	1	0.1	Normal
147Sm	0.02	1	0.1	Normal
153Eu	0.03	1	0.1	Normal
157Gd	0.03	1	0.1	Normal
159Tb	0.03	1	0.1	Normal
163Dy	0.03	1	0.1	Normal
165Ho	0.03	1	0.1	Normal
166Er	0.03	1	0.1	Normal
169Tm	0.03	1	0.1	Normal
172Yb	0.03	1	0.1	Normal
175Lu	0.03	1	0.1	Normal
178Hf	0.02	1	0.1	Normal
181Ta	0.03	1	0.1	Normal
182W	0.01	1	0.1	Normal
208Pb	0.01	1	0.1	Normal
232Th	0.03	1	0.1	Normal
238U	0.03	1	0.1	Normal

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1	Redox state of the Dharwar craton root as inferred from mantle eclogite
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3	with implications for kimberlite and lamproite magma formation
5	with implications for kimber net and fampforte magina for mation
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5	Azhar M. Shaikh ^{1, 2, 3} *, Yannick Bussweiler ⁴ , Fanus Viljoen ² , Robert Bolhar ³ , S. Ravi ⁵ , Dominik
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21	ADJIKAUI
22	Despite over 400 occurrences of kimberlites and related rocks in India, mantle-derived xenoliths
23	are known only from a few occurrences. This paucity of mantle-derived xenoliths in Indian

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kimberlites has hampered investigations of the subcontinental lithospheric mantle (SCLM). Using a valuable selection of the rare xenolith inventory, we here report $Fe^{3+}/\Sigma Fe$ measurements for garnets using the electron microprobe (EPMA) flank method, targeting six mantle eclogite xenoliths (KL2 pipe) and fourteen peridotitic garnet xenocrysts (P9 and P10 hypabyssal intrusions) from the Wajrakarur kimberlite field (WKF) on the Eastern Dharwar craton (EDC). These data provide some of the first direct constraints on the oxygen fugacity (fO_2) of the lithospheric mantle beneath the Indian subcontinent.

The measured Fe³⁺/ Σ Fe ratios vary between 0.02 and 0.05 (±0.01) for the eclogite xenoliths and between 0.02 and 0.10 (±0.01) for the peridotitic garnets. Calculated $\Delta \log fO_2$ values for the KL2 eclogites show a wide range from FMQ-3.9 to FMQ-0.9 (±0.6), straddling the boundary between the diamond and carbonate stability fields. In terms of redox compositions, it appears that the KL2 eclogites are able to host diamond, which is consistent with the diamondiferous nature of this particular WKF locality and the presence of eclogitic garnet inclusions in diamonds from the nearby TK4 kimberlite body.

The peridotitic garnet xenocrysts from the P9 and P10 kimberlite bodies, which were 38 39 entrained between ~125 and 170 km depth, reveal $\Delta \log fO_2$ values between FMQ-4.5 and FMQ-2.6 (± 0.9). Garnet xenocrysts with 'normal' REE patterns exhibit higher Fe³⁺/ Σ Fe ratios compared 40 to garnets with 'sinusoidal' REE patterns. Importantly, the Fe³⁺/2Fe ratios of garnet xenocrysts 41 42 with 'normal' REE patterns (~125-160 km depth) correlate with metasomatic Ti-Y-Zr-V 43 enrichment, which suggests metasomatism-driven oxidation of the cratonic mantle at midlithospheric depths. Such melt-related mantle metasomatism was probably diamond-destructive 44 45 within the otherwise diamond-fertile lithospheric keel.

2

46 The observed wide range of $\Delta \log fO_2$ values for the Dharwar cratonic mantle lithosphere 47 allows for stabilization of various metasomatic phases (e.g., amphiboles, micas, carbonates) that 48 may have formed (or concentrated in) distinctly different metasome assemblages within the 49 continental root that underpins Peninsular India. Changing the relative contributions from such 50 highly diverse volatile-rich metasomes may explain the close close spatiotemporal association of 51 kimberlites and various diamond-bearing potassic magma types such as orangeites, ultramafic 52 lamprophyres and lamproites, a scenario that is influenced by the redox composition of the 53 Dharwar craton root.

54

Keywords: Continental lithospheric mantle, Eclogite, Garnet, Oxygen fugacity, Mantle redox,
Wajrakarur kimberlites, Southern India

57

58 Introduction

59 Oxygen fugacity (fO_2) has significant implications for the speciation of CHONS-volatiles and the 60 stability of diamond/graphite in the Earth's mantle (Stagno, 2019). Oxygen fugacity also 61 influences (i) melting and metasomatism of mantle rocks, thereby influencing fluxes of various 62 magma types, and (ii) recycling of volatiles, thereby affecting global volatile cycles (Yaxley et al., 63 2017). Therefore, understanding fO_2 in Earth's mantle is crucial for comprehending terrestrial 64 magmatism and volatile cycles (Foley, 2011; Tappe et al., 2018). The fO₂ compositions of the 65 subcontinental lithospheric mantle (SCLM) have been shown to vary with depth and time (Woodland and Koch, 2003; Creighton et al., 2009, 2010; Yaxley et al., 2017; Tappe et al., 2021; 66 67 Aulbach et al., 2022), which has the potential to shift melting regimes over geological timescales 68 (Foley, 2011). The fO₂ values can be estimated for mantle-derived peridotite (Ballhaus et al., 1991; 59 Stagno et al., 2013) and eclogite (Stagno et al., 2015) xenoliths by measuring the Fe³⁺/ Σ Fe ratios 50 of their constituting garnet fractions.

71 This contribution presents the first measurements of $Fe^{3+}/\Sigma Fe$ in garnets from six eclogite xenoliths from the KL2 hypabyssal intrusion and fourteen peridotitic garnet xenocrysts from the 72 73 P9 and P10 intrusions of the Wajrakarur Kimberlite Field (WKF) located on the Eastern Dharwar 74 craton (EDC) in southern India. We aim to constrain the redox state of the regional SCLM that 75 underpinned Peninsular India at ca. 1.1 Ga. The KL2 intrusion forms part of the Kalyandurga 76 cluster, which consists of seven minor kimberlite intrusions (KL1 to KL7) and is located in the 77 southern part of the WKF (Fig. 1). The P9 and P10 minor intrusions form part of the Wajrakarur-78 Lattavaram cluster, situated in the north. Shaikh et al. (2020) constrained the architecture and 79 thermal state of the regional SCLM using major and trace element compositions of garnets from P9 and P10, some of which were re-analyzed here for their Fe³⁺/ Σ Fe compositions. According to 80 81 Shaikh et al. (2020), lithospheric thinning beneath the Dharwar craton, from ~190 km depth at 1.1 82 Ga to ~120 km depth at present, was aided by extensive melt metasomatism at the bottom of the 83 SCLM.

84 The ca. 1.1 Ga Kalyandurga kimberlites are special in several ways. For instance, (i) the 85 diamondiferous KL2 minor intrusion is distinguished by an overabundance of eclogite xenoliths (>95% of the mantle xenolith population are eclogite nodules: Rao et al., 2001; Neelakantam, 86 87 2001; Patel et al., 2006); (ii) the KL4 minor intrusion exhibits an exceptional abundance of mantle-88 derived ilmenite macrocrysts (Sastry et al., 2005); (iii) the Kalyandurga kimberlites intruded the 2.6–2.5 Ga Closepet granite in close proximity to the Chitradurga Boundary Fault, which presents 89 90 an important structure for the understanding of the evolution of the Dharwar craton (e.g., Chadwick et al., 2000). KL2 eclogite nodules have previously been investigated for petrography, in-situ 91

92	mineral major and trace element contents, and oxygen isotopic compositions to constrain their
93	origins (Ganguly and Bhattacharya, 1987; Rao et al., 2001; Patel et al., 2006, 2009; Griffin et al.,
94	2009; Dongre et al., 2015). Four eclogite xenoliths were examined by Ganguly and Bhattacharya
95	(1987), while two eclogite nodules were examined by Rao et al. (2001) to determine mineral major
96	element compositions. The first detailed petrographic observations and mineral major element data
97	for a suite of eleven KL2 eclogite xenoliths were provided by Patel et al. (2006). In order to review
98	the P-T conditions and origins of eclogites and pyroxenites from the WKF, Patel et al. (2009)
99	combined published xenolith data with nine newly analyzed xenoliths and argued against the
100	involvement of subducted ancient oceanic crust in the formation of KL2 eclogites. These authors
101	used textural and mineral compositional evidence to support a magmatic cumulate origin for the
102	KL2 eclogites. Babu et al. (2008) presented a much larger eclogite dataset (35 xenoliths) at the 9 th
103	International Kimberlite Conference, and Griffin et al. (2009) utilized these results to argue that
104	this eclogite xenolith suite originated from mafic melts that ponded and crystallized at lithospheric
105	mantle depths. Mineral major and trace element compositions, as well as the lateral distribution of
106	mantle-derived xenoliths and xenocrysts, were used to support this interpretation (Griffin et al.,
107	2009). In opposition to this notion, Dongre et al. (2015) examined 28 KL2 eclogite xenoliths and
108	reported the first δ^{18} O data (+5.3 to +7.8‰), which are best explained by a subduction-recycling
109	model. Recently, a few more KL2 eclogite nodules were studied by Chatterjee et al. (2023)
110	supporting the subduction model. However, no attempt has been made so far to use the eclogites
111	and other mantle-derived materials to determine the oxidation state of the SCLM beneath southern
112	India.

Herein, we apply the calibration of Stagno et al. (2015), which estimates eclogite fO_2 using the major element compositions of garnet and clinopyroxene, together with Fe³⁺/ Σ Fe ratio 115 measurements for garnet, an approach taken in several previous eclogite xenolith studies from 116 cratons worldwide (e.g., Smart et al., 2017, 2021a; Aulbach et al., 2019, 2022; Burness et al., 2020; 117 Mikhailenko et al., 2020). Our garnet $Fe^{3+}/\Sigma Fe$ measurements are complemented by in-situ major 118 and trace element data for garnet crystals from the KL2 eclogites. For peridotitic garnet xenocrysts, we used the calibration of Stagno et al. (2013) and combined garnet Fe³⁺/ Σ Fe data with their 119 modeled Fe²⁺/(Fe²⁺+Mg) compositions, assuming equilibrium with olivine and orthopyroxene in 120 121 the original peridotite host rock (O'Neil and Wood, 1979; Harley, 1984; Ryan et al., 1996; Gaul 122 et al., 2000) at estimated equilibrium P-T conditions (Canil et al., 1999). Our results provide the 123 first fO₂ estimates for eclogites and peridotitic garnets from southern India entrained by ca. 1.1 Ga old Mesoproterozoic kimberlites that represent one of the earliest global emplacement events of 124 125 diamond-bearing deep-sourced magmas (Tappe et al., 2018).

126

127 Analytical techniques

Seven eclogite xenoliths (KL2A to KL2G), measuring 3 to 5 cm in diameter, were collected from the KL2 kimberlite body by S. Ravi. They were cut and polished into a total of 18 standard petrographic thin sections for this study. The eclogite xenoliths are commonly rounded to subrounded (oval) nodules with medium-grained textures. Honey brown pyrope garnet crystals are embedded in a light green to white matrix formed mainly by altered omphacitic clinopyroxene.

133 A CAMECA SX100 electron microprobe housed in the Spectrum lab at the University of 134 Johannesburg was used for quantitative in-situ mineral analyses of major and minor elements. The 135 analyses were conducted with a nominal beam size of 1 μ m, an accelerating voltage of 20 kV, and 136 a beam current of 20 nA. Trace elements were measured for garnet, clinopyroxene and kyanite in 137 representative eclogite thin sections at the University of Johannesburg using a Thermo Scientific 138 iCAP RQ inductively coupled plasma mass spectrometer (ICP-MS) coupled to a 193 nm ArF 139 RESOlution SE155 excimer laser. NIST SRM 612 was used as the calibration reference material, 140 and ²⁹Si was used for internal standardization. To verify accuracy of the data, various USGS 141 basaltic glasses (BCR-2G, BHVO-2, BIR-1G) and the in-house GHR1 megacrystic garnet from 142 the Monastery kimberlite were analyzed as secondary standards, similar to the setup reported in 143 Tappe et al. (2023). Data quality is documented in Supp. Table 1. When compared to 144 recommended values for the USGS glasses and the GHR1 garnet (e.g., Jochum et al., 2016; Tappe 145 et al., 2021), the majority of trace elements analyzed reproduced with a variance of less than 10 to 146 15%. To avoid any potential contamination from the host kimberlite during laser ablation, we 147 rejected ICP-MS trace element measurements with >1 ppm Ba (see Shaikh et al., 2020). The 148 analytical techniques and data are described in more detail in the Appendix 1 and Supp. Table 1, 149 respectively. The analytical techniques applied to measure the major and trace element 150 concentrations for fourteen peridotitic garnet xenocrysts from Wajrakarur kimberlites P9 and P10 151 are given in Shaikh et al. (2020).

The atomic $Fe^{3+}/\Sigma Fe$ proportions in garnet crystals from six eclogite nodules and fourteen 152 153 xenocrysts of peridotitic affinity were determined with the flank method as developed by Höfer et 154 al. (1994) and further refined by Höfer and Brey (2007). Measurements were conducted with a 155 JEOL JXA-8530F Plus electron microprobe at Goethe University Frankfurt, Germany. The flank 156 method and the quantitative elemental analyses were conducted simultaneously using WDS at 15 157 kV and 120 nA, with a beam diameter of 1 µm. Two spectrometers with TAPL crystals for high 158 intensities and the smallest detector slit (300 μ m) were used, with 100 s counting time for FeL_a and FeL_{β}. The Fe³⁺/ Σ Fe ratios for garnets were determined by applying the correction for self-159 absorption using natural and synthetic garnet crystals with variable total Fe and Fe³⁺/ Σ Fe, as 160

161	independently determined by Mössbauer spectroscopy (Höfer and Brey, 2007). We used
162	Damknolle garnet megacrysts from Nigeria (Rankenburg et al., 2004) and the McGuire almandine
163	from the U.S.A. (McGuire et al., 1992) along with three in-house Cr-pyrope standards (UA5,
164	UA10, UA17) as reference materials. The Cr-pyrope crystals have a wide range of total Fe, which
165	is ideal for calibration (Supp. Table 1) to obtain fit parameters derived exclusively from these three
166	natural garnets. The other three spectrometers, not involved in flank method measurements, carried
167	out elemental analysis of Si, Ti, Al, Cr, Fe, Mn, Ni, Mg, Ca, Na, K and P during the same
168	measurement cycle. Appropriate silicate minerals [pyrope (Mg, Al, Si), albite (Na), CaSiO ₃ (Ca)]
169	and a phosphate mineral [KTiOPO4(Ti,K,P)], as well as metals or metal oxides [iron metal (Fe),
170	NiO (Ni), MnTiO ₃ (Mn), Cr ₂ O ₃ (Cr)] were used as calibration standards. A PRZ routine was used
171	for matrix correction. The analytical uncertainty in $Fe^{3+}\!/\!\Sigma Fe$ ratio measurements by the EPMA
172	flank method is approximately \pm 0.01 (1s), and it is noted that in general the uncertainty is
173	inversely correlated with garnet FeO content. A recent discussion about the applicability of the
174	flank method for determining ferric-ferrous iron ratios can be found in Rzehak et al. (2020).

175

176 **Results**

177 Petrography

178 KL2 Eclogites

Secondary alteration is prominent in the KL2 eclogite nodules, with a general decrease in the level of alteration from omphacite to kyanite to garnet (Fig. 2, 3), which was also observed previously (Sastry et al., 2005; Patel et al., 2006; Dongre et al., 2015). Garnet crystals are least altered but show minor secondary carbonate and hydro-garnet replacement along grain boundaries and fractures. Despite the alteration of several of our KL2 eclogite samples, the secondary minerals preserve the original rock textures, such as elongation texture with substantial micro-fracturing in garnet and clinopyroxene relics (e.g., Fig. 2, 3). Among the examined eclogite nodules, substantial clinopyroxene and kyanite alteration was observed in five samples (KL2C, KL2D, KL2E, KL2F, KL2G), whereas these phases were completely altered in sample KL2B. The freshest sample KL2A does not contain kyanite.

189 Subhedral to spherical garnet grains are interlocked with anhedral clinopyroxene and 190 bladed kyanite in relatively fresh samples with minor alteration along grain boundaries. Inclusions 191 of garnet and clinopyroxene are frequent in kyanite (Fig. 3 d, e, f). A few eclogite nodules (KL2D, 192 KL2F) contain needles of corundum, which are generally spatially associated with kyanite (Fig. 193 4a). Rutile is preserved as an inclusion in kyanite within the KL2A eclogite nodule (e.g., Fig. 4b). 194 In the majority of samples analyzed, the relative modal proportions of garnet, omphacite and 195 kyanite are ~45:45:10, and ~50:50 in bimineralic kyanite-free eclogite nodules with only very 196 minor kyanite and corundum in a few samples. Carbonate and chlorite are the most prevalent 197 alteration products of all primary minerals. Omphacite is typically replaced by hornblende and 198 diopside, kyanite by celsian feldspar (along grain boundaries and fractures), and garnet by chlorite, 199 epidote, celsian and grossular. The matrix of the eclogites contains rare sulfides (NiFeS) and pure 200 nickel oxide (NiO). Mukherjee et al. (2021) reported native gold particles and Au-Pt alloys from 201 KL2 eclogite xenoliths. A very detailed petrographic report for KL2 eclogites is given by Patel et 202 al. (2006).

203

204 *Peridotitic garnet xenocrysts*

Fourteen peridotitic garnet xenocrysts (3-7 mm in size) from the P9 and P10 minor intrusions were
 mounted on thin sections and polished for in-situ mineral chemical examination of major and trace

elements. Garnet grains exhibit micro-fracturing and host rare inclusions of Cr-diopside consistentwith their peridotitic mantle origins.

209

210 Major and trace element compositions of minerals

211 KL2 eclogites

The five primary minerals in KL2 eclogites (garnet, clinopyroxene, kyanite, corundum, rutile) were analyzed for major and trace elements. Secondary phases such as celsian, andradite, grossular, serpentine and carbonates were also analyzed for major elements. Supp. Table 1 lists representative major, minor and trace element compositions of primary phases essential for this work, as well as the compositions of secondary phases for documentation purposes. We only address the compositions of primary phases here, and readers are referred to Patel et al. (2006) for a detailed description of secondary phases in the KL2 eclogite xenoliths.

219 Despite the secondary alteration along grain boundaries and fractures, garnets in KL2 220 eclogites show relatively uniform compositions (Supp. Table 1). The eclogitic garnet compositions 221 determined here vary between Alm₂₄₋₂₇Grs₂₅₋₅₈Pyp₁₈₋₄₈ falling within the range of published data 222 for KL2 (e.g., Patel et al., 2006; Dongre et al., 2015). Garnets have low Na₂O and TiO₂ contents 223 (<0.3 wt.%; Supp. Table 1), which is typical for low-pressure eclogites (Gurney and Moore, 1993). 224 Chondrite-normalized (after Sun and McDonough, 1989) REE patterns of garnets from KL2 225 eclogites show LREE-depletion, a positive Eu anomaly, and flat MREE-HREE distributions (Fig. 226 5c), which is characteristic for garnets from cratonic eclogites worldwide (e.g., Jacob, 2004; 227 Aulbach and Jacob, 2016; Aulbach and Smart, 2023). Garnets show variable trace element 228 concentrations: Sc (29-35 ppm), Ti (828-1135 ppm), V (48-71 ppm), Cr (109-518 ppm), Ni (1584 ppm), Zr (2–6 ppm), and Y (5–8 ppm). They have low Sr contents (4–10 ppm) and U-Th
concentrations at sub-ppm levels.

Clinopyroxenes are omphacitic in nature (Ae₇₋₁₀Jd₃₁₋₄₃Di₅₀₋₆₁) similar to the known compositions of KL2 eclogitic clinopyroxene (Ae₃₋₁₇Jd₁₃₋₅₂Di₄₁₋₇₉). Contents of Na₂O and Al₂O₃ range from 2.9-8.2 wt.% and 5.8-16.2 wt.%, respectively. Cr₂O₃ content is low at 0.1 wt.%, rarely approaching 1.6 wt.%. Clinopyroxene shows a clear negative correlation between MgO and Na₂O (inset in Fig. 5b), typical for kimberlite-borne eclogites. Their chondrite-normalized REE patterns exhibit high LREE/HREE with a notable positive Eu anomaly (Fig. 5d).

237Kyanite in KL2 eclogite xenoliths contains trace amounts of FeO (<0.5 wt.%) and Cr_2O_3 238(<0.9 wt.%) (Supp. Table 1). Most trace elements have concentrations below the EPMA detection</td>239limit (<30 ppm), except for Ti (106-294 ppm), V (35-70 ppm), Cr (157-604 ppm) and Ga (7-10</td>240ppm). Corundum is a pure aluminous phase with a mean Al₂O₃ content of ~99 wt.% and minor241amounts of FeO (<0.5 wt.%) and Cr_2O_3 (<0.16 wt.%). Rutile is a pure Ti-oxide phase (~99 wt.%</td>242TiO₂) with minor amounts of FeO (<0.12 wt.%) and Cr_2O_3 (<0.18 wt.%).</td>

243

244 Peridotitic garnet xenocrysts

Major and trace element compositions of peridotitic garnet xenocrysts were discussed in detail by Shaikh et al. (2020). For this redox study, we included 11 lherzolitic G9 grains, two harzburgitic G10 grains, and a single Ti-metasomatic G11 grain (garnet classification of Grütter et al., 2004; Fig. 6a). Overall, these garnet crystals show a wide range of Cr_2O_3 (1.8–11.8 wt.%), moderate CaO (4.3–5.9 wt.%) and very low TiO₂ contents (<0.35 wt.%). Most of these garnets show 'normal' chondrite-normalized REE patterns, however, xenocrysts with >5 wt.% Cr_2O_3 are characterized by 'sinusoidal' REE patterns (Fig. 6b). Clinopyroxene inclusions in the garnet xenocrysts are Crdiopsides (1.1–2.1 wt.% Cr_2O_3 ; $En_{50-55}Wo_{41-46}Fs_4$) with consistently high Mg# values (0.92–0.94).

253

254 Bulk eclogite reconstruction and fO₂ estimates based on redox-sensitive elements

255 Due to pervasive alteration and kimberlite melt infiltration of the KL2 eclogite nodules (see Fig. 256 3), bulk-rock compositions were reconstructed from the measured chemical compositions of the 257 primary minerals (clinopyroxene, garnet, corundum, rutile, ± kyanite) in combination with their 258 estimated modal mineral abundances. This method of bulk-rock reconstruction has been widely 259 used in the past and continues to be a valuable tool in the study of xenolithic eclogites (e.g., Jacob, 260 2004; Tappe et al., 2011; Smart et al., 2016; Aulbach and Smart, 2023). EPMA data were used for 261 the major elements and LA-ICP-MS data for the trace elements. The calculations were performed 262 taking into account the specific gravities (Deer et al., 2013) of the primary minerals (3.3 g/cm³ for 263 clinopyroxene, 3.6 g/cm³ for garnet, 3.3 g/cm³ for kyanite, 4.04 g/cm³ for rutile, 4.02 g/cm³ for 264 corundum) and for bulk eclogite (3.45 g/cm³). The specific gravity was utilized to calculate the 265 relative weight proportions. Given that the variability of the average chemical compositions of the 266 samples studied is low (generally <10% RSD), the major source of uncertainty in the bulk-rock 267 reconstructions stems from the estimates of the modal mineral abundances. For bimineralic 268 kyanite-free samples, we assumed 49 vol.% for clinopyroxene and garnet each, and 1 vol.% for 269 rutile and corundum each. For kyanite-bearing eclogite nodules, we assumed 44 vol.% for 270 clinopyroxene and garnet each, 1 vol.% for rutile and corundum each, and 10 vol.% for kyanite. 271 Bulk eclogite reconstruction results for the major elements (normalized to 100 wt.%) and the trace 272 elements (in ppm) are given in Supp. Table 1. The results are plotted in Fig. 7a-c to classify the 273 eclogites (see Discussion), which show low MgO, high Na2O, and Eu/Eu* anomalies.

274 As shown in Figure 8a, the estimated modal abundance of rutile has a significant effect on the overall budget of Ti, which in combination with the concentration of V, provides an estimate 275 276 for fO₂ (e.g., Aulbach and Stachel, 2022). Based on previous analyses of mantle eclogites and our 277 own petrographic observations, we conclude that a modal rutile abundance between 0.5 and 1 278 vol.% is realistic. Regardless of the exact choice of rutile abundance, our KL2 eclogite samples 279 fall just below FMQ-2 in Figure 8a. In terms of Lu/Gd versus Ce/Yb ratios, our reconstructed bulk 280 eclogite compositions agree with mantle eclogites from cratons worldwide, partly overlapping 281 with metasomatized eclogites (Fig. 8b). In terms of V/Sc versus MgO systematics, our 282 reconstructed bulk eclogite compositions suggest a fO_2 value of around FMQ-2 (Fig. 8c), similar 283 to the Ti-V based estimate (Fig. 8a).

284

285 Thermobarometer choice

286 Nimis (2022) presented an overview of geothermobarometers applicable to eclogites. Older 287 thermometer calibrations based on garnet-clinopyroxene Fe-Mg exchange developed by Ellis and 288 Green (1979) and Powell (1985) have been demonstrated to be unreliable because the effect of Ca 289 was not considered (e.g., Brey and Köhler, 1990; Nimis and Grütter, 2010). Pressure estimates by 290 Beyer et al. (2015) for eclogite xenoliths show systematically lower values (by ~10 kbar) when compared to pressures calculated using orthopyroxene-garnet assemblages at the same 291 292 temperatures (Nimis, 2022). Furthermore, due to the high sensitivity of the equation to slight 293 inaccuracies in clinopyroxene major element compositions, the Beyer et al. (2015) eclogite 294 barometer is prone to produce significant errors.

In this study, temperatures were determined for six KL2 eclogite xenoliths (at an assumed pressure of 40 kbar) based on Fe-Mg exchange between garnet and clinopyroxene (Krogh, 1988;

297	T_K). Temperatures for KL2B eclogite could not be calculated due to the complete alteration of
298	clinopyroxene. In addition, published data for 34 KL2 eclogite nodules were reprocessed with the
299	same thermometer equation. The eclogite equilibrium pressures $\left(P_{HC40}\right)$ were calculated by
300	subsequently projecting the obtained temperatures onto the regional cratonic geotherm of 40
301	mW/m^2 (after Shaikh et al., 2020). Temperatures $(T_{\rm K})$ obtained for an assumed pressure of 40 kbar
302	range from 945 to 1180 °C. Pressures obtained after geotherm projection range from 38 to 50 kbar
303	(Fig. 9a), equivalent to 120-160 km depths. Alternative temperature estimates calculated after Ellis
304	and Green (T_{EG} ; 1979), Powell (T_{Po} ; 1985), Ai (T_{Ai} ; 1994), Ganguly et al. (T_{Gn} ; 1996), Krogh
305	Ravna (T_{KR} ; 2000), and Nakamura (T_{Nk} ; 2009) yielded very similar results for the KL2 eclogite
306	xenoliths (Supp. Table 1). Overall, our P-T results compare well to those obtained by Patel et al.
307	(2009) and Dongre et al. (2015) (i.e., 36–54 kbar).

The equilibration pressures (P_{HC40}) for peridotitic garnet xenocrysts were calculated using Ni-in-garnet temperatures (970–1217 °C) (Canil, 1999) projected onto the regional 40 mW/m² geotherm (Fig. 9b). Pressure readings for two garnet xenocrysts (P9G51A, P9G52C) were corrected using a single-pyroxene thermobarometer applied to Cr-diopside inclusions (<u>after Nimis</u> <u>and Taylor, 2000</u>). The peridotitic garnet xenocrysts studied equilibrated at 40 to 53 kbar corresponding to ~125-170 km depth, similar to the depths of origin of the KL2 eclogite xenoliths.

315 Measured ferric iron contents in garnet, and calculated fO_2

Table 1 and Table 2 list the Fe³⁺/ Σ Fe ratios for garnets from KL2 eclogite xenoliths and peridotitic garnet xenocrysts, respectively. Fe³⁺/ Σ Fe ratios for our eclogitic garnets vary from 0.02 to 0.05 (±0.01), which is within the global range of mantle-derived eclogitic garnets (0.02–0.09; Stagno et al., 2015; Smart et al., 2017, 2021a, 2021b; Aulbach et al., 2019, 2022). The Fe³⁺/ Σ Fe ratios for our peridotitic garnet xenocrysts range from 0.02 to 0.10 (±0.01), extending to higher values than
those measured for KL2 eclogites.

322 We calculated $\Delta \log fO_2$ values (relative to the FMQ buffer) for the KL2 eclogite xenoliths using the oxybarometer of Stagno et al. (2015), which was calibrated using coesite/quartz-bearing 323 324 eclogites. By applying the calculated pressure-temperature values and measured chemical 325 compositions for garnet and clinopyroxene, the KL2 eclogites yielded $\Delta \log fO_2$ values between 326 FMQ-2.5 and FMQ-0.9, with uncertainties of ±0.6 log units for corundum-free eclogites (Fig. 10). 327 These redox compositions fall closer to the carbonate stability field compared with the fO_2 328 systematics of other xenolithic eclogite suites from cratons worldwide (Stagno et al., 2015; 329 Burness et al., 2020; Smart et al., 2021a, b; Aulbach et al., 2022). The above fO₂ range for the KL2 330 eclogite xenoliths is in good agreement with our less-precise reconstructed bulk eclogite fO_2 331 estimate of \langle FMQ-2 (see Fig. 8). As per Smart et al. (2021), a correction is required for the fO_2 332 calculations for corundum-bearing silica-undersaturated eclogites. The correction involves the 333 incorporation of silica activity using corundum + SiO₂ (coesite) = kyanite equilibrium. Application 334 of this correction to the KL2D and KL2F corundum-bearing silica-undersaturated eclogite 335 xenoliths shifts their $\Delta \log fO_2$ values by 1.5 log units toward more reduced redox compositions 336 (FMQ-3.9 and FMQ-3.6), more similar to eclogite xenoliths from the Kaapvaal craton root (Fig. 337 10). There is a weak correlation between $\Delta \log fO_2$ values and contents of MgO and CaO for garnet, 338 with $\Delta \log fO_2$ increasing with decreasing MgO at increasing CaO (Fig. 11). No correlation is 339 observed between the trace element concentrations of eclogitic garnets and their $\Delta \log fO_2$ values. 340 For the peridotitic garnet xenocrysts, we used the Stagno et al. (2013) equation, which 341 requires input for pressure and temperature (see above), as well as the Fe# values for coexisting 342 olivine and orthopyroxene. The Fe# values for orthopyroxene in equilibrium with garnet were

343 calculated using Equation 7 in Ryan et al. (1996) and Equation 11 in Harley (1984). The Fe# values 344 for olivine in equilibrium with garnet were calculated by inverting the garnet-olivine thermometer 345 of O'Neill and Wood (1979), as demonstrated by Gaul et al. (2000). The obtained Fe# values range 346 from 0.06 to 0.08 for olivine and 0.03 to 0.04 for orthopyroxene (variance is <1%; Gaul et al., 347 2000; Ryan et al., 1996), corresponding to Mg# values of 0.92-0.94 and 0.96-0.97, respectively. 348 These inferred values are similar to the Mg# values reported for olivine and orthopyroxene from rare peridotite xenoliths from the Dharwar craton root (0.90-0.94 for olivine; 0.90-0.95 for 349 350 orthopyroxene; Ganguly and Bhattacharya, 1987; Nehru and Reddy, 1989; Pattnaik et al., 2020), 351 and also overlap with the compositions of mantle-derived olivine xenocrysts from Wajrakarur 352 kimberlites (0.90-0.94 Mg#; Shaikh et al., 2019, 2018). This complex procedure enabled us to 353 calculate $\Delta \log fO_2$ values between FMQ-4.5 and FMQ-2.6 (± 0.9) for the peridotitic garnet 354 xenocrysts (Fig. 10).

355

356 Discussion

357 Nomenclature and origin of the KL2 eclogites

358 Mantle-derived eclogite xenoliths are generally thought to be formed by either (i) subduction of 359 gabbroic or basaltic oceanic crust (e.g., Taylor and Neal, 1989; Barth et al., 2001; Jacob, 2004), 360 (ii) emplacement of basaltic melt at depths within cratonic mantle lithosphere with high-pressure 361 cumulate formation (e.g., Viljoen et al., 1996; Barth et al., 2002), or (iii) extraction of tonalitic 362 melt from basaltic oceanic crust during subduction leaving behind an eclogitic residue (e.g., Jacob 363 and Foley, 1999; Barth et al., 2002; Tappe et al., 2011a). Each of these genetic types of cratonic 364 eclogites is supported by textural, mineralogical, geochemical, and isotopic evidence (Aulbach and 365 Smart, 2023). Accordingly, mantle-derived eclogites have been classified into several types based on major and trace element compositions of garnet and clinopyroxene (McCandless and Gurney,
1989; Taylor and Neal, 1989; Jacob et al., 2009) and reconstructed bulk compositions (e.g.,
Aulbach and Jacob, 2016; Smart et al., 2017).

369 The KL2 eclogites studied here contain clinopyroxene that is enriched in Na₂O (typically 370 4-7 wt.%) and has moderate MgO contents (typically 6-11 wt.%). These clinopyroxene 371 compositions are characteristic for Type B and Type C eclogites of Taylor and Neal (1989; Fig. 5a, b), interpreted to originate from subducted ancient oceanic crust protoliths. The relatively low 372 373 K₂O contents of clinopyroxene (<0.1 wt.%, or below detection limit), together with the low Na₂O 374 (0.15 wt.%) and TiO₂ (0.5 wt.%) contents in garnets (Supp. Table 1), are similar to the mineral 375 compositions of Group II non-metasomatized eclogite xenoliths described by McCandless and 376 Gurney (1989). KL2 eclogites contain garnets with moderate Mg# values (typically between 0.4-377 0.7) and high Ca# values (0.2-0.6) (Fig. 5; 7), similar to the high-Ca eclogites as defined by 378 Aulbach and Jacob (2016). KL2 eclogitic garnets and clinopyroxenes exhibit Eu anomalies in their 379 REE patterns, which are also mirrored in the reconstructed bulk eclogite compositions (Fig. 5c, d; 380 7c). The reconstructed bulk eclogite compositions are relatively high in Na₂O (2-3.5 wt.%; Fig. 381 7a). These geochemical features indicate that the protolith of the KL2 eclogite xenoliths was a 382 plagioclase-rich gabbroic lithology as part of ancient oceanic crust. This finding is consistent with 383 the model by Dongre et al. (2015), who presented evidence for low-temperature alteration of the 384 eclogite protolith near the Earth's surface based on significantly elevated δ^{18} O values for garnet 385 (up to +7.8%) from some KL2 eclogite xenoliths. However, features such as interlocking garnet-386 clinopyroxene textures, rutile blebs in clinopyroxene and garnet, the presence of corundum and 387 Na₂O-poor garnet and K₂O-poor clinopyroxene, and Eu-anomalies are more similar to Type II_L 388 eclogite xenoliths from the Roberts Victor orangeite in South Africa (Hardman et al., 2021). These

389 eclogite nodules are considered to represent cumulates of magmas sourced from a depleted mantle 390 reservoir that had undergone prior extraction of MORB-like melts (Hardman et al., 2021), but 391 subducted oceanic crustal origins have also been put forward for the classic Roberts Victor eclogite 392 xenolith suite (e.g., Jacob et al., 2005). For KL2 eclogites, Patel et al. (2009) proposed a high-393 pressure cumulate origin, which is supported by textural features such as graded layering and 394 garnet necklaces as well as garnet-kyanite clusters, plus evidence from mineral compositions such 395 as similar clinopyroxene geochemistry in the eclogite xenoliths and cumulate pyroxenites. In 396 contrast, Griffin et al. (2009) suggested KL2 eclogite formation by solidification of mafic melts 397 near the base of the SCLM. Clearly, there are 'eclogites and eclogites', and considering the scope 398 of our study (mantle redox), we refrain from further speculations as to the nature and origin of the 399 KL2 eclogite protolith.

400

401 Redox state of KL2 eclogite xenoliths

402 Using compositions of eclogitic garnets from the southern region of the WKF (Kalyandurga 403 kimberlite cluster), we calculated $\Delta \log fO_2$ values between FMQ-2.5 and FMQ-0.9 (±0.6), with 404 two eclogite nodules (KL2C and KL2E) recording more reduced oxidation states (FMQ-3.9 and 405 FMQ-3.6). On the basis of these $\Delta \log fO_2$ values and their depth distribution, the KL2 eclogites 406 straddle the boundary between diamond and carbonate stability fields in terms of carbon speciation 407 (Fig. 10). The peridotitic garnet xenocrysts from the P9 and P10 kimberlite bodies of the northern 408 region of the WKF (Wajrakarur-Lattavaram kimberlite cluster) yielded $\Delta \log fO_2$ values ranging 409 from FMQ-4.5 to FMQ-2.6(± 0.9), at the low end of eclogitic $\Delta \log fO_2$ values determined here. This 410 observation suggests that the majority of KL2 mantle eclogites have experienced more oxidizing 411 conditions compared to cratonic peridotites at similar depths prior to entrainment into erupting
412	kimberlite magmas at ca. 1.1 Ga. The KL2 eclogite xenoliths studied have $\Delta log fO_2$ values that
413	overlap with those of many kimberlite-borne eclogite nodules from cratons worldwide (FMQ-1 to
414	FMQ-5; Stagno et al., 2015; Burness et al., 2020; Smart et al., 2021a, b; Aulbach et al., 2022).
415	Depth related redox heterogeneity has been observed for eclogites from different intervals
416	within cratonic SCLM worldwide (e.g., Burness et al., 2020). For example, Kaapvaal cratonic
417	eclogites from mid-lithospheric depths show more reducing redox conditions ($\Delta log fO_2 = FMQ$ -
418	5.3 to FMQ-3.3) compared to eclogites from the lowermost lithosphere (FMQ-3.9 to FMQ-1.5)
419	(Burness et al., 2020). However, this apparent relationship was not observed in the data produced
420	for other localities on the Kaapvaal craton or elsewhere (Smart et al., 2017, 2021a, b; Aulbach et
421	al., 2022). Based on our data for KL2 eclogites, we observe notable variations in their oxidation
422	state over a relatively narrow depth interval, but large-scale redox variations with depths cannot
423	be discerned. Further work is required to explore such a potential relationship. We can conclude,
424	however, that the KL2 eclogites present a suitable diamond host rock given their $P-T-fO_2$
425	systematics (Fig. 10). This conclusion is supported by the diamondiferous nature of the KL2
426	kimberlite body and mineral inclusion data from the WKF (Ravi et al., 2013).
427	
428	Redox state of mantle peridotites and metasomatism of the Dharwar craton root
429	Garnet xenocrysts of peridotitic affinity from the P9 and P10 kimberlites revealed a wide range of
430	$\Delta \log fO_2$ values between FMQ-4.5 and FMQ-2.6 (±0.9 log units). Griffin et al. (2009) and Shaikh
431	et al. (2020) presented extensive trace element data for garnet and clinopyroxene xenocrysts from
432	Wajrakarur kimberlites, which allows us to explore any possible relationship between melt

metasomatism of the SCLM and its redox composition. Shaikh et al. (2020) proposed the presence 433 434 of a mid-lithospheric discontinuity between ~145 and 160 km depth.

435	The Dharwar craton root is strongly metasomatized at the very bottom, a trend known from
436	many lithospheric mantle profiles beneath cratons worldwide (e.g., Griffin et al., 1999b; Kopylova
437	et al., 1999; Kargin et al., 2016; Smart et al., 2017; Aulbach et al., 2007, 2013, 2017; Tappe et al.,
438	2021). However, unlike the Kaapvaal craton, the Dharwar SCLM shows concomitant enrichment
439	in Zr and Y in garnet (Shaikh et al., 2020). This feature has been interpreted to result from high-
440	temperature mantle metasomatism by mixed silicate-carbonate melts (Shaikh et al., 2020) or by
441	basaltic melts (Griffin et al., 2009). Although, the Ti/Eu versus Zr/Hf systematics of peridotitic
442	garnets, suggest a kimberlitic to carbonatitic nature of the metasomatic agents (Shaikh et al., 2020).
443	According to global datasets, the oxidation state of mantle peridotite varies laterally and
444	vertically across the SCLM (e.g., Woodland and Koch, 2003; Creighton et al., 2009; Yaxley et al.,
445	2017; Stagno et al., 2013; Tappe et al., 2021). For example, in the garnet stability field, the
446	Kaapvaal SCLM shows a progressively more reducing trend with increasing depth ($\Delta log fO_2$ FMQ-
447	2 at 110 km to FMQ-4 at 210 km; Luth, 1990; Woodland and Koch, 2003; Creighton et al., 2009,
448	2010; Tappe et al., 2021). This crystal chemistry-controlled depth-fO2 trend has been masked by
449	oxidative metasomatism beneath the Kimberley area on the Kaapvaal craton, where many
450	peridotite xenoliths are offset to higher-than-expected fO2 values (Creighton et al., 2009; Hanger
451	et al., 2015). This is also consistent with observed Fe^{3+} variations in metasomatic growth zones on
452	mantle-derived garnet crystals (McCammon et al., 2001). The peridotitic garnet xenocrysts from
453	the WKF studied here show variations in $\Delta log fO_2$ and they fall within the Kaapvaal peridotite P-
454	fO_2 array, with a few samples showing slightly higher $\Delta log fO_2$ values approaching the carbonate
455	stability field (Fig. 10). The Fe ³⁺ / Σ Fe ratios measured for trace element enriched garnet xenocrysts
456	with normal REE patterns show a <i>remarkable</i> positive correlation with incompatible trace element
457	concentrations (Ti, Zr, Y, V; Fig. 12), possibly indicating a metasomatic control on cratonic mantle

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redox (Creighton et al., 2009). These garnet xenocrysts were sampled from ~125–160 km depths, an SCLM interval that was identified as a modally metasomatized mid-lithospheric discontinuity by Shaikh et al. (2020). Overall, the link between carbonated silicate melt metasomatism and mantle redox in the Dharwar craton root may be responsible for local diamond destruction, as reported for other cratons worldwide (Creighton et al., 2009; Fedortchouk et al., 2019).

463

464 Linking mantle redox and the diversity of 1.1 Ga kimberlitic magmatism in southern India 465 The WKF comprises a diverse range of diamondiferous magma types, including archetypal 466 kimberlites, lamproites, orangeites, and ultramafic lamprophyres (see compilation in Shaikh et al., 467 2017; Pandey and Chalapathi Rao, 2020). To date, 48 minor intrusions of exclusively 468 Mesoproterozoic age (ca. 1.1 Ga) are known to occur within a relatively small area that is 80×70 469 km in size. The coeval emplacement of diverse volatile-rich ultramafic magma types reflects 470 prominent mineralogical heterogeneity in the Dharwar craton root (e.g., Sarkar et al., 2021), as 471 was also demonstrated for cratonic regions and their primitive potassic magmatism on either side 472 of the North Atlantic (Tappe et al., 2008, 2011b; Dalton et al., 2019).

473 The correlation between metasomatic overprint and redox state of the Dharwar craton root, 474 as demonstrated in this study, appears to have important implications for the petrogeneses of 475 kimberlites, lamproites and closely related primitive potassic rocks. Progressive oxidative 476 metasomatism along conduits in the SCLM was caused by infiltrating carbonated silicate melts 477 that probably originated from low-degree partial melting of the underlying convecting mantle 478 including its recycled crustal components (e.g., Nowell et al., 2004; Malkovets et al., 2007). The 479 resultant fO_2 heterogeneities in the SCLM allowed for stabilization of distinctly different 480 metasomatic assemblages that may be dominated by either amphiboles, micas, or carbonates (Foley et al., 1986; Foley, 2011; Tappe et al., 2008; Yaxley et al., 2017). The highly variable nature of SCLM metasomes, and also local lack of metasomes, may explain the close spatiotemporal association of diverse types of primitive K-rich magmas (lamproites, orangeites, ultramafic lamprophyres) and kimberlites in the WKF.

485 Shaikh et al. (2017) suggested a model for the origin of various types of diamond-bearing 486 magmas in the WKF, following concepts developed in Tappe et al. (2008, 2011b), where 487 carbonated silicate melts from the asthenosphere interacted with variably metasomatized 488 lithologies of the SCLM to produce the diversity of ca. 1.1 Ga old volatile-rich ultramafic rocks 489 known from southern India. According to this model, metasomatic phlogopite-carbonate 490 dominated veins were reactivated by infiltrating asthenosphere-derived melts (proto-kimberlitic 491 melts), which gave rise to ultramafic lamprophyre, orangeite and lamproite magmas as a function 492 of increasingly higher proportions of micas in the cratonic mantle source (Shaikh et al., 2017). 493 Sarkar et al. (2021) compared the compositions of magmatic and xenocrystic olivine populations 494 from Wajrakarur kimberlites and lamproites. These authors suggested that both magma types have 495 a common asthenospheric mantle source, with lamproites revealing a larger extent of assimilation 496 of Fe-rich lithospheric metasomes.

497

498 On the 'overabundance' of eclogite xenoliths in the KL2 kimberlite

Despite the general xenolith deficiency, the WKF minor intrusions reveal contrasting mantle sampling patterns. For example, the KL2 kimberlite is marked by an overabundance of eclogite xenoliths, whereas kimberlites and related rocks from the Wajrakarur-Lattavaram cluster contain more peridotite xenoliths (Rao et al., 2001; Patel et al., 2006). This apparent bias is also mirrored by the diamond occurrences (Ravi et al., 2013) and distributions of eclogitic versus peridotitic
garnet xenocrysts (Griffin et al., 2009; Shaikh et al., 2020).

505 Although kimberlites and related rocks with an overabundance of eclogite xenoliths are 506 very rare, they occur on most cratons worldwide such as the Kaapvaal craton (e.g., Roberts Victor, 507 Bellsbank), the Slave craton (e.g., Jericho, Voyageur), the North Atlantic craton (e.g., Nunatak 508 1390) and the Siberian craton (e.g., Zagadochnaya) (Schulze, 1989; Jacob, 2004; Smart et al., 509 2009, 2017, 2021a; Tappe et al., 2011a; Kopylova et al., 2016; Hardman et al., 2021; Aulbach and 510 Smart, 2023). Eclogite xenolith overabundance has been linked to localized enrichment of the 511 peridotite-dominated cratonic mantle column with eclogite components (e.g., Nixon and Davies, 512 1987) or biased preservation of xenoliths during entrainment into kimberlitic magmas (e.g., 513 Schulze, 1989). Kopylova et al. (2016) inferred lateral continuity of eclogites within the Slave 514 craton SCLM, where an eclogite-rich layer may represent the remnant of an imbricated oceanic 515 lithosphere slab. The abundance of eclogite components has great economic significance because 516 they contribute up to 30% of lithospheric diamonds in global statistics (Stachel and Luth, 2015), 517 which can be even higher on a regional scale. The overall abundance of eclogitic components in 518 peridotite-dominated SCLM has been estimated at <1 vol.% (Schulze, 1989); however, higher 519 abundances of up to 4-10 vol.% have been suggested for the Slave craton (Griffin et al., 1999; 520 Kopylova et al., 2016). The combined petrological-geophysical approach by Garber et al. (2018) 521 devised an even higher eclogite abundance within the SCLM beneath cratons (up to 20 vol.%). 522 Regardless of the exact precise amount of eclogites within the cratonic mantle lithosphere, the 523 KL2 occurrence on the Eastern Dharwar craton adds to the kimberlite localities that have a 524 somewhat inexplicable abundance of xenolithic eclogites, which may influence the redox

distribution within the local SCLM and, by inference, also its volatile element and diamondinventory.

527

528 Summary and Conclusions

529 The $Fe^{3+}\Sigma Fe$ compositions of garnets were determined by the EPMA flank method for six eclogite 530 xenoliths from the KL2 kimberlite and fourteen peridotite-derived xenocrysts from the P9 and P10 531 kimberlites on the Eastern Dharwar craton in southern India. KL2 eclogites are characterized by 532 low $\Delta log/O_2$ values between FMQ-3.9 and FMQ-0.9 (±0.6), which suggests that they could have 533 acted as diamond host rocks within the deeper Dharwar craton root. The peridotite-derived garnet 534 xenocrysts reveal a wide range of $\Delta \log fO_2$ values between FMQ-4.5 and FMQ-2.6 (± 0.9), firmly 535 within the well-studied Kaapvaal lithospheric mantle array. At mid-lithospheric depths beneath 536 the Dharwar craton, redox compositions of the garnet xenocrysts correlate with geochemical 537 proxies for melt-rock interactions, which provides evidence for the operation of oxidative mantle 538 metasomatism that was likely diamond-destructive. Strong redox variability at relatively confined SCLM intervals would promote stability of diverse volatile-rich metasomatic mineral assemblages 539 (e.g., roles of carbonates versus phlogopite and amphibole), which helps to explain the 540 541 spatiotemporal association of ca. 1.1 Ga kimberlites, lamproites, orangeites and ultramafic 542 lamprophyres in southern India.

543

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- 843 Figure Captions
- **Fig. 1.** Geological map of Wajrakarur Kimberlite Field (WKF) modified after Shaikh et al. (2017).
- 845 This study deals with samples from the KL2, P9, and P10 kimberlite intrusions (highlighted by
- 846 yellow-filled circles).

Fig. 2. Scanned thin section images of KL2 eclogite nodules.

850	Fig. 3. BSE images of KL2 eclogite nodules showing mineral grain relations and the extensive
851	alteration of primary and matrix minerals. (a) Highly fractured garnet in sample KL2A1 showing
852	alteration to hydro-garnet along margins. (b) Sample KL2B1 shows garnet altered to carbonate
853	and chlorite along fractures and no preservation of omphacite and kyanite. (c) Highly fractured
854	and altered omphacite in sample KL2A1. (d) Kyanite blades in sample KL2C3, with inclusions of
855	garnet and omphacite, partially altered to celsian. (e) Kyanite blade in sample KL2G1 with
856	inclusions of garnet and omphacite. (f) Interlocked kyanite blades in sample KL2F2 with an
857	omphacite inclusion.
858	
859	Fig. 4. BSE images of KL2 eclogite nodules with corundum associated with kyanite (a) and a rutile
860	bleb (b).
861	
862	Fig. 5. Major and trace element (REE) plots for garnet (a, b, c) and clinopyroxene (b, d) from KL2
863	eclogites. The fields in figure (a) are after Aulbach and Jacob (2016) and in (b) after McCandless
864	and Gurney (1989). The A, B, and C fields in the inset figure are after Taylor and Neal (1989).
865	
866	Fig. 6. Cr ₂ O ₃ versus CaO wt.% plot (a) and chondrite normalized REE plot (b; after (Sun and
867	McDonough, 1989) for garnet xenocrysts from P9 and P10 intrusions of the WKF.
868	

870	in figures (a) and (b) are after Aulbach and Jacob (2016), and primitive mantle normalization in
871	(c) is after (Sun and McDonough, 1989). Published KL2 eclogite data are from Dongre et al.
872	(2015). Symbol sizes in (a) and (b) are larger than the 2σ uncertainties.
873	
874	Fig. 8. Trace element variations diagrams based on reconstructed bulk-rock compositions for
875	eclogite nodules. Note that reconstructed bulk-rock compositions indicate fO2 values below FMQ-
876	2. Figures (a) Ti vs. Vi/Ti*100, (b) Ce/Yb vs. Lu/Gd, and (c) MgO vs. V/Sc are after Wang et al.
877	(2019) and Aulbach and Viljoen (2015). Symbol sizes in (a) and (b) are larger than the 2σ
878	uncertainties.
879	
880	Fig. 9. Projections of equilibration temperatures calculated for KL2 eclogite nodules (a; after
881	Krogh, 1988) and P9-P10 garnet xenocrysts (b; after Canil, 1999). Published KL2 data from Patel
882	et al. (2006) and Dongre et al. (2015). The conductive model geotherm (after Hasterok and
883	Chapman, 2011) for the Wajrakarur lithosphere is from Shaikh et al. (2020). The diamond-graphite
884	transition is after Day et al. (2012).
885	
886	Fig. 10. Δlog/O ₂ (FMQ) values calculated for KL2 eclogitic garnets (after Stagno et al., 2015) and
887	P9 and P10 peridotitic garnet xenocrysts (after Stagno et al., 2013) plotted against the estimated
888	pressure. The field for Kaapvaal peridotites is after Woodland and Koch (2003) and Creighton et
889	al. (2009), enriched peridotites from Slave craton after Yaxley et al. (2017) and graphite-diamond-
890	carbonate melt transition lines after Stagno et al. (2015). Updated Kaapvaal data (small open

Fig. 7. Reconstructed bulk-rock composition diagrams for classifying KL2 eclogite nodules. Fields

869

891

circles) are after Tappe et al. (2021).

35

8	893	Fig. 11. $\Delta log fO_2$ (FMQ) versus MgO and CaO (in wt%) for KL2 eclogitic garnets from Wajrakarur
8	894	kimberlites.
8	895	
٤	896	Fig. 12. $Fe^{3+}/\Sigma Fe$ ratios plotted against respective $TiO_2(a)$, Y (b), Zr (c), and V (d) for in-peridotitic
8	897	garnet xenocrysts from the P9 and P10 kimberlite bodies at intrusions of the Wajrakarur. Kaapvaal

898 <u>craton xenolith data are taken from Creighton et al. (2009).</u>

Supp. Table 1

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