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4 **Sheared peridotite and megacryst formation beneath the**  
5 **Kaapvaal craton: a snapshot of tectonomagmatic processes**  
6 **across the lithosphere–asthenosphere transition**  
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50 **Running title:** Origins of sheared peridotite xenoliths  
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## ABSTRACT

The cratonic lithosphere–asthenosphere boundary is commonly invoked as the site of sheared peridotite and megacryst formation, a well-recognized petrologic assemblage whose genetic relationships – if any – remain poorly understood. We have undertaken a comprehensive petrology and Sr-Nd-Hf-Ca isotope study of sheared peridotite xenoliths and clinopyroxene megacrysts from the ca. 1150 Ma Premier kimberlite pipe on the central Kaapvaal craton in South Africa. New textural and mineral trace element evidence suggests that strong tectonic and magmatic overprinting affected the lower cratonic mantle over a vertical distance of  $\geq 50$  km from the lithosphere–asthenosphere boundary located at  $\sim 200$ – $225$  km depth. Although modification of the central Kaapvaal cratonic mantle is commonly linked to the ca. 2056 Ma Bushveld large igneous event, our thermobarometry, mantle redox, and Sr-Nd-Hf-Ca isotope data support a model in which volatile-rich low-volume melts and associated high-density fluids refertilized the lithosphere base shortly before or during asthenosphere-derived kimberlite and carbonatite magmatism at around 1150 Ma. This episode of lithospheric mantle enrichment was facilitated by exceptionally strong shear movements, as are recorded in the plastically deformed peridotites. We argue that stress-driven segregation of percolating carbonated melts contributed to megacryst formation along, or in close proximity to, shear zones within the cratonic mantle lithosphere.

Integration of our results from the Kaapvaal craton and modern petrologic concepts allows for the identification of a lithosphere–asthenosphere transition zone between  $\sim 150$ – $225$  km depth. This horizon is defined by intersections of the  $\sim 40$ – $42$  mW/m<sup>2</sup> Premier paleogeotherm with (1) CO<sub>2</sub>-H<sub>2</sub>O-present solidus curves for peridotite (upper bound), and (2) typical mantle adiabats with potential temperatures between 1315 and 1420°C (lower bound). At Premier, the most strongly deformed sheared peridotites occur mainly between  $\sim 160$ – $185$

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3 km depth, firmly within the lithosphere–asthenosphere transition zone. Contrary to many  
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5 previous models, we suggest that sheared peridotite formation occurs in localized deformation  
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7 zones spaced out across the entire width of the lithosphere–asthenosphere transition zone,  
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9 rather than being restricted to a single thin layer at the craton base where mantle flow causes  
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11 viscous drag. Hence, plate tectonic stresses acting on the lower cratonic lithosphere may be  
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13 accommodated by extensive networks of shear zones, which provide transient pathways and  
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15 sinks for percolating volatile-rich melts, linking the formation of megacrysts and sheared  
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17 peridotites.  
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22 **Keywords:** Bushveld igneous event; Cratonic lithosphere evolution; Deformation-assisted  
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24 melt segregation; Mantle redox; Sr-Nd-Hf-Ca isotopes; Thermobarometry  
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## INTRODUCTION

Plate tectonics drives the compositional and dynamic evolution of the solid Earth. It mainly involves the rigid lithosphere, tessellated into discrete ‘tectonic plates’, and the rheologically weaker asthenosphere, *or* convecting upper mantle, over which the lithospheric plates move at different speeds and directions. The lithosphere–asthenosphere boundary (LAB) can be envisaged as a series of large-scale detachment zones along which tectonic strain caused by differential movements of mantle materials is accommodated (Eaton *et al.*, 2009). Beneath Archean cratons, the present-day LAB is typically located at 150–250 km depth, much deeper than in other continental settings or within the ocean basins, which complicates an accurate delineation of the cratonic LAB by geophysical methods (Jordan, 1978; Artemieva & Mooney, 2001; Jones *et al.*, 2010; Priestley *et al.*, 2019). Lithospheric plates appear to be most strongly coupled to asthenospheric flow at the base of thick continental roots (Stoddard & Abbott, 1996; Conrad & Lithgow-Bertelloni, 2006; Mancinelli *et al.*, 2017), and this ‘basal drag’ provides an important driving force of plate tectonics in addition to slab pull and ridge push (Bokelmann, 2002; Bokelmann & Silver, 2002; Kennedy *et al.*, 2002; Eaton & Frederiksen, 2007; Kaban *et al.*, 2015). For example, kinematic reconstructions for the past 200 million years suggest that lithospheric plates with a high proportion of thick continental roots drifted more slowly than the much thinner oceanic plates or composite oceanic–continental plates that lack significant cratonic regions (Zahirovic *et al.*, 2015).

The physicochemical properties of mantle materials in the vicinity of the cratonic LAB are only poorly understood (O'Reilly & Griffin, 2010). While some workers suggested that strongly deformed lherzolite xenoliths in kimberlites represent samples of upwelling asthenosphere near the cratonic LAB (Nixon & Boyd, 1973b; Green & Gueguen, 1974; Boyd & Nixon, 1975; Goetze, 1975; Mercier, 1979; Allegre *et al.*, 1982; Boyd *et al.*, 1985), more

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3 recent interpretations view such ‘porphyroclastic’ or ‘hot sheared’ lherzolites (hereafter  
4 ‘sheared peridotites’) as the product of plastic deformation and refertilization–rehydration of  
5 ancient depleted peridotite protoliths at the base of cratonic lithosphere (Pearson *et al.*, 1995;  
6 Kopylova *et al.*, 1999; Kennedy *et al.*, 2002; Baptiste *et al.*, 2012; Agashev *et al.*, 2013;  
7 Doucet *et al.*, 2013; van der Meer *et al.*, 2013; Kargin *et al.*, 2017; Kopylova *et al.*, 2019),  
8 although somewhat shallower cratonic mantle shear zones have also been invoked (Katayama  
9 *et al.*, 2009). As such, sheared peridotites provide rare snapshots of the tectonic and magmatic  
10 interactions between the lithosphere and the underlying asthenosphere, adding to our  
11 knowledge about the dynamic Earth. More specifically, studies of sheared peridotite xenolith  
12 suites allow for estimates on the positions of the LAB at the time of kimberlite eruptions  
13 (O'Reilly & Griffin, 2010). They also provide insights into whether the lithosphere–  
14 asthenosphere interface is diffuse and several 10s of kilometers thick (Mancinelli *et al.*,  
15 2017), or relatively sharp occurring over  $\leq 15$  km vertical distance (Eaton *et al.*, 2009;  
16 Tharimena *et al.*, 2017). The elemental and isotopic compositions of sheared peridotites and  
17 their constituent minerals provide information on the nature and origin of deep percolating  
18 melts (e.g., basaltic *versus* volatile-rich melts including diverse types of high-density fluids),  
19 with possible implications for diamond formation (Weiss *et al.*, 2011; Smith *et al.*, 2012;  
20 Jablon & Navon, 2016), kimberlite magma production and ascent (Moore & Belousova, 2005;  
21 Gregoire *et al.*, 2006; Arndt *et al.*, 2010), as well as high-resolution geophysical imaging of  
22 the elusive LAB beneath continental shields (Gaillard *et al.*, 2008; Massuyeau *et al.*, 2021).  
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This study evaluates a new comprehensive petrology (e.g., thermobarometry calculations, mantle redox measurements, mineral trace element analyses) and Sr-Nd-Hf-Ca isotope dataset for sheared peridotite xenoliths and clinopyroxene megacrysts from the ca. 1150 Ma Premier kimberlite pipe on the Kaapvaal craton in South Africa (Fig. 1). The results are integrated to further develop concepts of the nature and evolution of the cratonic LAB,

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3 including the tectonomagmatic processes that shape this important boundary layer. We also  
4 assess the possibility that our sheared and metasomatized peridotite samples may present  
5 vestiges of the viscous drag applied to the underside of the ‘Kalahari’ tectonic plate by strong  
6 asthenospheric flow during the 1220–1090 Ma Namaqua-Natal orogeny, which was caused by  
7 collisions between the Kaapvaal craton and numerous surrounding terranes (Fig. 1).  
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## 15 **GEOLOGICAL BACKGROUND AND SAMPLES**

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19 The 1153.3 ± 5.3 Ma Premier kimberlite diatreme on the central Kaapvaal craton in South  
20 Africa has a preserved surface area of ~32 hectares and thus presents one of the largest  
21 volcanic pipe structures in the world (Tappe *et al.*, 2018a). Premier pipe has been mined  
22 intermittently since 1903 and holds the record for the largest ever discovered gem-quality  
23 diamond – the 3106 carats ‘Cullinan’ stone. The deposit is renowned for the recovery of  
24 exceptionally large and pure high-value Type-II diamonds, and it ranks among the top five  
25 diamond mines by value with more than 20 billion US\$ in contained revenue (de Wit *et al.*,  
26 2016).  
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38 The Premier kimberlite and associated rare carbonatite dykes are part of a cluster of 11  
39 known pipes that intruded at the southern periphery of the Bushveld Complex some 50 km  
40 northeast of Johannesburg (Fig. 1). At the present erosion and mining levels, the kimberlite  
41 magmas and associated volcanoclastic materials erupted through sedimentary rocks of the  
42 2650 – 2200 Ma Transvaal Supergroup and mafic–ultramafic rocks of the ca. 2056 Ma  
43 Bushveld Complex (Field *et al.*, 2008). Although emplacement of the Premier kimberlite  
44 cluster has previously been linked to the formation of Mesoproterozoic alkaline and  
45 carbonatite complexes on the central Kaapvaal craton (Kramers & Smith, 1983; Harmer,  
46 1999; Verwoerd, 2006), improved age constraints established a >200 million years gap  
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3 between these events (Elburg & Cawthorn, 2017; Tappe *et al.*, 2018a). This new finding rules  
4 out direct petrogenetic links between these discrete mantle-derived magmatic episodes, and it  
5 underpins that the Kaapvaal lithospheric root had been affected repeatedly by carbonate-rich  
6 and alkaline magmatism during the Mesoproterozoic (Fig. 1).  
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13 The combined Nd-Hf-Os isotope systematics of kimberlite and carbonatite dykes from  
14 Premier pipe suggest ultimate melt origins from the convecting upper mantle to the transition  
15 zone, with variable degrees of melt–rock interaction upon encounter of the overriding cratonic  
16 mantle lithosphere (Tappe *et al.*, 2020b). These radiogenic isotope data are compatible with  
17 recent results for rare mineral inclusion-bearing Type-II diamonds from Premier pipe that  
18 suggest ultimate origins of these stones from below the cratonic lithosphere well within the  
19 convecting mantle (Smith *et al.*, 2016; Korolev *et al.*, 2018a; Nestola *et al.*, 2018; Smith *et*  
20 *al.*, 2018). However, an origin of ‘ultradeep’ Type-II diamonds from the cratonic mantle near  
21 the LAB, either as part of megacryst-forming events (Moore, 2009; Moore & Helmstaedt,  
22 2019) or as subcreted ‘fossil’ upwelling mantle (Haggerty, 2017; Tappe *et al.*, 2020a), is  
23 equally plausible.  
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38 The Kaapvaal craton is one of the best studied sizeable remnants of an Archean  
39 continent (de Wit *et al.*, 1992; Poujol *et al.*, 2003; Hofmann *et al.*, 2019), and the nature of its  
40 thick mantle root has profoundly influenced models of cratonic lithosphere formation (Boyd  
41 *et al.*, 1985; Pearson, 1999; O'Reilly *et al.*, 2001; Woodland & Koch, 2003; Carlson *et al.*,  
42 2005; Aulbach, 2012; Herzberg & Rudnick, 2012; Kamber & Tomlinson, 2019). The craton  
43 comprises two major Early Archean blocks of typical granite–greenstone lineage, which were  
44 brought together during Late Archean subduction and collision tectonics (Shirey *et al.*, 2002;  
45 Schmitz *et al.*, 2004; Smart *et al.*, 2016). Whereas the western cratonic domain, or so-called  
46 Kimberley block, exhibits one of the highest incidences of kimberlite magma eruptions during  
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3 the Mesozoic (Tappe *et al.*, 2018b) with fairly efficient sampling of the western Kaapvaal  
4 mantle lithosphere (Bell *et al.*, 2003; Grégoire *et al.*, 2003; Griffin *et al.*, 2003; Simon *et al.*,  
5 2007; Gibson *et al.*, 2008; Rehfeldt *et al.*, 2008; Creighton *et al.*, 2009; Katayama *et al.*, 2009;  
6 Lazarov *et al.*, 2009; Hanger *et al.*, 2015; Tomlinson *et al.*, 2018; Shu *et al.*, 2019; Smart *et*  
7 *al.*, 2021a), the central plus eastern domains (i.e., the Witwatersrand block – host to Premier  
8 kimberlite pipe) are less well studied in terms of their cratonic mantle compositions (Pearson  
9 *et al.*, 1995; Gregoire *et al.*, 2005; Viljoen *et al.*, 2009; Aulbach & Viljoen, 2015; Shu &  
10 Brey, 2015; Burness *et al.*, 2020; Smart *et al.*, 2021b). In general, the thermal state and  
11 chemical composition of the Kaapvaal mantle lithosphere are regionally highly heterogeneous  
12 (Bell *et al.*, 2003; Griffin *et al.*, 2003), which is also recognized in irregular present-day P-  
13 wave velocity patterns at cratonic mantle depth levels imaged in seismic tomography studies  
14 (James *et al.*, 2001; Fouch *et al.*, 2004). Regions with high seismic P-wave velocities reflect  
15 strongly depleted mantle lithosphere or cooler conductive geotherms. In contrast, low P-wave  
16 velocities reflect refertilisation–rehydration of the cratonic mantle root or warmer local  
17 geotherms, which may be causally linked to voluminous regional intraplate magmatism such  
18 as the ca. 2056 Ma Bushveld or the ca. 180 Ma Karoo large igneous events (Griffin *et al.*,  
19 2003; Fouch *et al.*, 2004; Shirey *et al.*, 2004; Rehfeldt *et al.*, 2007; Giuliani *et al.*, 2014). In  
20 this context it should be noted that the Premier kimberlite pipe at the southern margin of the  
21 Bushveld Complex is located in a region of the central Kaapvaal craton that is characterized  
22 by lower present-day seismic P-wave velocities (James *et al.*, 2001; Shirey *et al.*, 2002).  
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The paleogeotherm beneath the ca. 1150 Ma Premier kimberlite pipe has been constrained at ~39 – 45 mW/m<sup>2</sup> based on peridotite xenolith and single-grain clinopyroxene thermobarometry (Danchin, 1979; Finnerty & Boyd, 1987; Viljoen *et al.*, 2009; Nimis *et al.*, 2020). These heat flow estimates were obtained by application of a range of methods and models, which are often not directly comparable (see Kopylova *et al.*, 2019). Nevertheless, it



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3 is reasonable to conclude that the study area had a slightly elevated cratonic geotherm at 1150  
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5 Ma. This observation, together with ca. 2 Ga old lherzolitic mineral inclusions in some  
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7 diamonds (Richardson *et al.*, 1993) and ca. 2 Ga rhenium depletion model ages of many  
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9 peridotite xenoliths from Premier (Pearson *et al.*, 1995; Morel *et al.*, 2008), led several  
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11 authors to propose that the central Kaapvaal mantle lithosphere had been heavily overprinted  
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13 by the Bushveld magmatic event (Danchin, 1979; Griffin *et al.*, 2003; Gregoire *et al.*, 2005;  
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15 Richardson & Shirey, 2008; Viljoen *et al.*, 2009; Viljoen *et al.*, 2014; Korolev *et al.*, 2018b).  
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17 It should be noted, however, that the southern African cratonic and peri-cratonic lithosphere is  
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19 generally characterized by ‘warmer’ conductive geotherms over the past one billion years  
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21 approaching 42 mW/m<sup>2</sup> in places (Rudnick & Nyblade, 1999; Grütter, 2009; Ngwenya &  
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23 Tappe, 2021). Hence, the impact by heat advection, emanating from giant mantle-derived  
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25 magmatic events, on the thermal structure of dynamically evolving cratonic mantle  
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27 lithosphere may be overestimated (Bell *et al.*, 2003; Kobussen *et al.*, 2009).  
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33 In 2015 and 2016, we collected >50 new sheared peridotite xenoliths from the Premier  
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35 kimberlite pipe at Cullinan Diamond Mine. After an initial inspection of these nodules in  
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37 petrographic thin sections, 14 fresh sheared peridotite xenoliths were identified as suitable for  
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39 pressure–temperature estimations and geochemical analyses; i.e., they exhibit a ‘touching’  
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41 garnet, orthopyroxene, clinopyroxene and olivine phase assemblage (Figs. 2-4). For  
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43 comparative purposes we also included a single coarse-grained granular garnet peridotite  
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45 xenolith into the analytical program (Fig. 2D). In addition, 10 clinopyroxene megacrysts  
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47 recovered from the oversize tailings dumps are incorporated into our petrology and isotope  
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49 geochemistry study, because previous models suggest links between the formation of sheared  
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51 peridotites and megacrysts near the cratonic LAB (Nixon & Boyd, 1973a, b; Moore & Lock,  
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53 2001; Moore & Belousova, 2005; Bussweiler *et al.*, 2018).  
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## METHODS AND APPROACH

### Electron microprobe analysis

Quantitative major and minor element compositions of silicates were determined on polished thin sections and epoxy grain mounts with a Cameca SX100 electron microprobe at the University of Johannesburg in South Africa. The instrument was operated at 20 kV acceleration voltage and a beam current of 20 nA was applied. The electron beam was adjusted for 1  $\mu\text{m}$  spot analyses. Signal counting times varied between 40 and 60 seconds on peak, depending on the chemical element of interest. The electron microprobe was calibrated using the following natural and synthetic reference materials: diopside (Si), almandine garnet (Al), hematite (Fe), wollastonite (Ca), olivine (Mg), rhodonite (Mn), orthoclase (K), jadeite (Na), as well as pure  $\text{TiO}_2$ ,  $\text{Cr}_2\text{O}_3$  and NiO. The elements were measured on their X-ray  $K\alpha$  lines employing all four wavelength dispersive spectrometers. Raw data reduction and matrix corrections were done with the 'X-PHI' method, which is a  $\phi(\rho z)$ -type off-line analytical protocol. Emphasis was laid on the detection of potentially occurring major and minor element zoning within individual mineral grains; however, the observed intra-grain compositional variability – if any – is within the reproducibility of our analytical technique (Ngwenya & Tappe, 2021). Backscatter electron imagery also demonstrates compositional homogeneity of all present silicate phases within any given peridotite xenolith sample (except for porous/spongy clinopyroxene rims due to partial melting) (Figs. 3, 4). This confirms attainment of full mineral major element equilibrium (Finnerty & Boyd, 1984; Nimis & Grütter, 2010), which is a prerequisite to the application of meaningful thermobarometry. To this end, we also generated backscatter electron maps for selected thin sections at full scale (Fig. 3A) using a FEI Quanta 600F Mineral Liberation Analyser system at the University of

Johannesburg. The instrument is equipped with two Bruker 4010 Xflash X-ray detectors, and operating conditions are given in Ngwenya & Tappe (2021).

### **Laser ablation ICP-MS analysis**

Mineral trace element concentrations were determined in situ by laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) at Stellenbosch University in South Africa and at the University of Münster in Germany. Eight representative peridotite xenoliths and all ten clinopyroxene megacrysts from the Premier kimberlite pipe were crushed in an agate mortar and visibly clear crystal fragments of the 0.5 – 1.2 mm size fraction were mounted in epoxy resin at the University of Johannesburg. Orthopyroxene and clinopyroxene grain mounts were analysed at Stellenbosch University and the garnet grain mounts were measured at the University of Münster. Several garnet grains were analysed as replicates in Stellenbosch, and the trace element results from the two laboratories agree within 20% in all cases and within 10% in most cases. However, only the garnet dataset from Münster is reported and discussed in the main manuscript.

The LA-ICP-MS setup at Stellenbosch University consists of an excimer laser system emitting at 193 nm (Resonetics Resolution M50) coupled to an Agilent 7700 quadrupole ICP-MS instrument. Ablations were performed in a double helix Laurin Technic cell in helium carrier gas that was mixed into the argon sample gas, with nitrogen added to the gas mixture at a rate of 5 ml per minute prior to introduction into the mass spectrometer. Each analysis included 35 seconds of sample ablation plus background readings for 15 seconds prior to ablation. NIST SRM 610 glass was analysed as the calibration standard every 15 unknowns, which included analyses of the USGS basalt glasses BCR and BHVO as secondary reference materials for evaluations of accuracy and precision. Trace element concentrations were obtained by spot analysis using a beam diameter of 100  $\mu\text{m}$ . Up to five spot analyses were

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3 carried out per mounted grain and average trace element compositions for each grain studied  
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5 were calculated. The reduction of time-resolved raw data and calculations of trace element  
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7 concentrations were performed with Iolite 3.3, using  $^{29}\text{Si}$  as internal standard matched against  
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9 the electron microprobe data. The results for the BCR and BHVO glasses are within the 2-  
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11 sigma uncertainty margins of the recommended average element concentrations and typically  
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13 reproduce better than 6% (<http://georem.mpch-mainz.gwdg.de>). Detection limits for most  
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15 analysed trace elements are in the low ppb range or better. Further details about the  
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17 Stellenbosch LA-ICP-MS setup are provided in Smart et al. (2017).  
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22 In Münster, the LA-ICP-MS system consists of a 193 nm Photon Machine excimer  
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24 laser coupled to a ThermoFisher Scientific Element-2 SF-ICP-MS instrument (see the detailed  
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26 method description in Beyer et al., 2013). Ablations were performed in a Helex cell in helium  
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28 carrier gas that was mixed into the argon sample gas, with nitrogen added to the gas mixture  
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30 prior to entering the mass spectrometer. Each analysis included 40 seconds of sample ablation  
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32 plus background readings for 20 seconds before ablation. The NIST SRM 612 glass was  
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34 analysed as the calibration standard every 30 unknowns, which included analyses of the basalt  
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36 glasses BIR-1, BCR-2 and BHVO-2 as secondary reference materials for data quality control  
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38 (Supplementary Data Appendix A). Trace element concentrations were determined by spot  
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40 analysis applying a 85  $\mu\text{m}$  beam diameter. Five spot analyses were carried out per mounted  
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42 garnet grain and average trace element compositions for each grain studied were calculated.  
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44 Data reduction and the calculation of trace element concentrations were performed with  
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46 GLITTER 4.4, using  $^{29}\text{Si}$  as internal standard. Results for the basaltic glasses are within the 2-  
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48 sigma uncertainty margins of the recommended average element concentrations and  
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50 reproduce better than 5% (<http://georem.mpch-mainz.gwdg.de>). We also analysed mounted  
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52 GHR-1 garnet and KBY17-4 clinopyroxene as in-house reference materials (Supplementary  
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54 Data Appendix A), and the results for petrogenetically important trace elements such as Ni  
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3 and the REE are within the 2-sigma uncertainty margins of the average concentrations  
4 recommended in Smart et al. (2021). Detection limits for all analysed trace elements of  
5 interest are in the low ppb range or better.  
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### 10 **Mössbauer spectroscopy**

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12 The Fe<sup>3+</sup> contents of garnet were determined by Mössbauer spectroscopy at Frankfurt  
13 University in Germany, and they are reported in Table 1 as Fe<sup>3+</sup>/ΣFe. Spectra were obtained  
14 from visually clean hand-selected garnet grains that had been separated from five suitable  
15 sheared peridotite xenoliths from the Premier kimberlite pipe. The cleaned garnet separates  
16 were milled into powder and subsequently mixed with a small amount of sugar crystals prior  
17 to placement into a Pb sample holder. The amount of sample (10-20 mg) and sample diameter  
18 were adjusted to create sample thicknesses of <5 mg/cm<sup>2</sup> to mitigate saturation effects.  
19 Measurements were made at constant acceleration with a velocity ramp of ±5 mm/s and a 50  
20 mCi <sup>57</sup>Co in Rh source. Mirror-image spectra, calibrated against α-Fe metal foil, were  
21 collected over 512 channels until 2×10<sup>6</sup> counts per channel were reached. Fitting of the garnet  
22 spectra followed the approach of Woodland & Koch (2003), which included a correction for  
23 different recoil free fractions of Fe<sup>2+</sup> and Fe<sup>3+</sup> in garnet.  
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42 Corrected Fe<sup>3+</sup>/ΣFe ratios for the peridotite-derived garnet fractions are reported in  
43 Table 1, and they have absolute uncertainties of ±0.01 (Woodland & Koch, 2003). Peridotite  
44 *f*O<sub>2</sub> values are reported relative to the fayalite-magnetite-quartz (FMQ) reference buffer and  
45 they were calculated using the oxybarometer formulation of Stagno et al. (2013). This  
46 oxybarometer requires as input: (1) pressure-temperature estimates, (2) garnet major element  
47 compositions including the corrected Fe<sup>3+</sup>/ΣFe ratios, and (3) Fe<sup>2+</sup>/Mg ratios of coexisting  
48 olivine and orthopyroxene. All reported peridotite *f*O<sub>2</sub> literature data (Supplementary Data  
49 Appendix B), as determined by garnet Fe<sup>3+</sup> analyses, were recalculated applying the same  
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3 thermobarometer combination (Nickel & Green, 1985; Taylor, 1998) and oxybarometer  
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5 (Stagno *et al.*, 2013) to enable meaningful comparisons in depth– $fO_2$  space.  
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### 8 **Sr-Nd-Hf isotope ratio determinations**

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11 Clinopyroxene mineral separates were produced from six representative peridotite xenoliths  
12 and the ten megacrysts following coarse crushing steps in an agate mortar and sieving of the  
13 crushed material to collect the grain size fraction between 0.5 and 1.2 mm. Visually clean and  
14 inclusion-free clinopyroxene crystal fragments were hand-selected under a binocular  
15 microscope at the University of Johannesburg to obtain pure mineral fractions weighing  
16 between 18 and 180 mg. Representative crystal fragments from each sample were mounted in  
17 epoxy resin for trace element determinations by LA-ICP-MS analysis (see above).  
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28 The bulk of the clinopyroxene fractions was washed in deionized H<sub>2</sub>O, transferred into  
29 Savillex teflon vials and dissolved in concentrated HF–HNO<sub>3</sub> (3 / 1) on a hotplate at 150°C  
30 for 48 hours (University of Münster). Sample solutions were gently dried down and the  
31 residues were redissolved in 6N HCl alternating between a hotplate and an ultrasonic bath.  
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33 This dissolution step was repeated at least twice until clear solutions were obtained.  
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35 Approximately 10 wt.% of each sample solution was allocated for Ca isotope analytical work  
36 (see below). The other 90 wt.% of the solutions were processed through ion exchange  
37 chromatography to isolate Sr (AG50W-X8 cation resin, 200–400 mesh, (Hart & Brooks,  
38 1977)) and Nd-Hf (LN Spec anion resin, 50–100 mesh, (Pin & Zalduegui, 1997; Münker *et*  
39 *al.*, 2001)) fractions for isotope ratio determinations at the University of Münster. The total  
40 procedural blanks are typically <50 pg Sr, <30 pg Nd and <10 pg Hf, and are negligible  
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53 (Tappe *et al.*, 2020b).  
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Strontium isotope ratio measurements were performed on a Thermo Scientific Triton TIMS instrument ( $^{85}\text{Rb}$  monitored to correct for the potential presence of interfering  $^{87}\text{Rb}$  was negligible), whereas Nd and Hf isotope ratios were measured in solution mode with a Thermo Scientific Neptune Plus MC-ICP-MS instrument in static collection mode. Measured  $^{87}\text{Sr}/^{86}\text{Sr}$ ,  $^{143}\text{Nd}/^{144}\text{Nd}$  and  $^{176}\text{Hf}/^{177}\text{Hf}$  ratios were normalized using an exponential mass fractionation law and the recommended  $^{88}\text{Sr}/^{86}\text{Sr}$  (8.37521),  $^{146}\text{Nd}/^{144}\text{Nd}$  (0.7219) and  $^{179}\text{Hf}/^{177}\text{Hf}$  (0.7325) ratios. The measured Sr-Nd-Hf isotope ratios for all sample and secondary standard runs are reported relative to  $^{87}\text{Sr}/^{86}\text{Sr} = 0.710245$  for NBS987 (Thirlwall, 1991),  $^{143}\text{Nd}/^{144}\text{Nd} = 0.512115$  for JNdi-1 (Tanaka *et al.*, 2000), and  $^{176}\text{Hf}/^{177}\text{Hf} = 0.282160$  for AMES / JMC-475 (Blichert-Toft *et al.*, 1997) standard solutions, respectively. During the course of multiple analytical sessions in Münster in 2018–2019, repeated measurements of  $^{87}\text{Sr}/^{86}\text{Sr}$  for the NBS987 primary standard averaged  $0.710250 \pm 11$  (2S.D.,  $n=5$ ),  $^{143}\text{Nd}/^{144}\text{Nd}$  for the JNdi-1 primary standard averaged  $0.512072 \pm 17$  (2S.D.,  $n=20$ ), and  $^{176}\text{Hf}/^{177}\text{Hf}$  for the AMES primary standard averaged  $0.282146 \pm 9$  (2S.D.,  $n=16$ ). During this analysis period we also processed the USGS basaltic reference materials BCR-2 and BHVO-2 alongside our samples, and the results are in good agreement with previous measurements (Weis *et al.*, 2006; McCoy-West *et al.*, 2010; Chauvel *et al.*, 2011). For BCR-2 the obtained values are:  $^{87}\text{Sr}/^{86}\text{Sr} = 0.705005 \pm 7$  (2S.D.,  $n=2$ ),  $^{143}\text{Nd}/^{144}\text{Nd} = 0.512635 \pm 6$  (2S.D.,  $n=3$ ),  $^{176}\text{Hf}/^{177}\text{Hf} = 0.282873 \pm 22$  (2S.D.,  $n=3$ ). For BHVO-2 the obtained values are:  $^{87}\text{Sr}/^{86}\text{Sr} = 0.703469 \pm 7$  (2S.D.,  $n=2$ ),  $^{143}\text{Nd}/^{144}\text{Nd} = 0.512990 \pm 16$  (2S.D.,  $n=3$ ),  $^{176}\text{Hf}/^{177}\text{Hf} = 0.283108 \pm 13$  (2S.D.,  $n=3$ ) (Tappe *et al.*, 2020a).

Parent-daughter element ratios were calculated using the Rb, Sr, Sm, Nd, Lu and Hf concentrations of clinopyroxene as determined by the LA-ICP-MS method for the same sample aliquots used for solution mode Sr-Nd-Hf isotope analytical work. All initial isotope ratios and notations are reported for the Premier kimberlite magma emplacement age of ca.

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3 1150 Ma (Tappe *et al.*, 2018a). Corrections of measured  $^{87}\text{Sr}/^{86}\text{Sr}$  for 1150 million years of in  
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5 situ  $^{87}\text{Rb}$  decay are small due to the generally low Rb/Sr nature of clinopyroxene, whereas  
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7 calculations of the initial  $^{143}\text{Nd}/^{144}\text{Nd}$  and  $^{176}\text{Hf}/^{177}\text{Hf}$  ratios are associated with a substantial  
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9 error propagation resulting in uncertainties of 0.7–1.3 and 0.5–1.0 epsilon units at the 2-sigma  
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11 level, respectively. The 2-sigma uncertainties of the calculated initial epsilon Nd and Hf  
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13 values entail a propagation of errors assuming normally distributed data (Table 2), for which  
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15 the uncertainties of the 1150 Ma emplacement age and parent-daughter element ratios were  
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17 set at 1% (i.e., 1150  $\pm$ 12 Ma) and 5%, respectively. The following decay constants were used  
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19 for age corrections:  $^{87}\text{Rb}$   $1.42 \times 10^{-11} \text{ a}^{-1}$ ;  $^{147}\text{Sm}$   $6.54 \times 10^{-12} \text{ a}^{-1}$ ;  $^{176}\text{Lu}$   $1.865 \times 10^{-11} \text{ a}^{-1}$  (see Tappe  
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21 *et al.*, 2020b for references).  
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## 25 26 **Ca isotope ratio determinations**

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29 Approximately 10 wt.% of each sample solution prepared for Sr-Nd-Hf isotope analytical  
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31 work was transferred into Savillex teflon vials and mixed with a  $^{42}\text{Ca}$ – $^{43}\text{Ca}$  double-spike to  
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33 enable correction for isotope fractionation during mass spectrometry (Gussone *et al.*, 2011).  
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36 Calcium isotope ratio measurements were performed at the University of Münster on the  
37  
38 same Thermo Scientific Triton TIMS instrument as used for the Sr isotope ratio  
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40 determinations (see above). Purified Ca was loaded onto Re single filaments with a Ta  
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42 activator in sandwich technique (Gussone *et al.*, 2010). The Ca isotope variations are  
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44 expressed as  $\delta^{44/40}\text{Ca}$  values relative to the NIST SRM-915a standard:  $\delta^{44/40}\text{Ca} [\text{‰}] =$   
45  
46  $((^{44}\text{Ca}/^{40}\text{Ca})_{\text{sample}} / (^{44}\text{Ca}/^{40}\text{Ca})_{\text{SRM-915a}} - 1) \times 1000$ .  
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51 All sample data are normalised to the average value of the 3 to 5 measurements of SRM-915a  
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53 per turret during a TIMS analytical session to mitigate potential effects of collector cup-  
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55 degradation. Replicate measurements of samples and standards yielded an average 2S.D. of  
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57 approximately 0.05‰ (an individual analysis consisted of 180 cycles over a ca. 40 minute  
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3 period). The average value of 20 analyses of SRM-915b is  $0.76 \pm 0.02\text{‰}$  (2S.E.) or  $\pm 0.09\%$   
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5 (2S.D.) relative to SRM-915a, which is in good agreement with the recommended value of  
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7  $0.72\text{‰}$  (Heuser *et al.*, 2016). Nine analyses of the IAPSO seawater standard yielded a value  
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9 of  $1.87 \pm 0.03\text{‰}$  (2S.E.) or  $\pm 0.09\%$  (2S.D.) during the analytical sessions in Münster in  
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11 April/May 2018, which is in good agreement with the seawater  $\delta^{44/40}\text{Ca}$  values compiled in  
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13 Heuser *et al.* (2016).  
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## 16 17 **Thermobarometry**

### 18 19 *Pressure–temperature estimations*

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21 Pressures and temperatures (P-T) of last equilibration were calculated from the acquired plus  
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23 quality-screened mineral compositional data for the peridotite xenoliths and clinopyroxene  
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25 megacrysts following the guidelines of Nimis & Grütter (2010). For the garnet peridotite  
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27 xenoliths we applied two well-established thermobarometer combinations: (1) Taylor (1998)  
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29 – Nickel & Green (1985) [*hereafter TNG*], and (2) Brey & Köhler (1990). For both  
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31 combinations the temperature estimates are obtained from two-pyroxene Fe-Mg exchange  
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33 equilibria and the pressure estimates from Al-exchange equilibrium between coexisting  
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35 orthopyroxene and garnet. Although these two thermobarometer combinations yield  
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37 systematically different P-T values for individual data inputs (Supplementary Data Appendix  
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39 C; see also Kopylova *et al.*, 2019), the obtained P-T arrays, or shapes of the resultant  
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41 paleogeotherms, are identical within uncertainties. However, fitting of garnet-facies peridotite  
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43 xenoliths P-T arrays using the numerical *FITPLOT* model (McKenzie & Bickle, 1988) shows  
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45 that the *TNG* thermobarometer combination results in more precise estimates of lithosphere  
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47 thickness, which was also pointed out by Mather *et al.* (2011). This observation is in good  
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49 agreement with the reappraisal of widely used barometers and thermometers by Nimis &  
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51 Grütter (2010), who recommended the *TNG* combination as the most accurate currently  
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3 available. Pressure–temperature estimates of last equilibration for the clinopyroxene  
4 megacrysts were obtained using the single-grain thermobarometer of Nimis & Taylor (2000),  
5 implementing the new data quality-control screens developed by Ziberna et al. (2016).  
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### 10 *Pressure–temperature array ('geotherm') fitting*

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14 The obtained garnet peridotite xenolith and clinopyroxene megacryst P-T arrays were  
15 quantitatively fitted to calculate paleogeotherms utilizing the numerical *FITPLOT* model  
16 (McKenzie & Bickle, 1988; Mather *et al.*, 2011). Besides equations that describe the thermal  
17 properties of peridotite-dominated lithospheric mantle, *FITPLOT* requires the following input  
18 parameters for the crustal and mantle portions of the lithosphere column under investigation:  
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25 (1) thickness and heat production of the upper and lower crust, (2) heat production of the  
26 lithospheric mantle, and (3) potential temperature  $T_p$  of the sublithospheric convecting upper  
27 mantle, *or* asthenosphere.  
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33 For *FITPLOT* geotherm calculations using our P-T dataset for xenoliths and  
34 megacrysts from the ca. 1150 Ma Premier kimberlite we applied the upper (23 km) and lower  
35 (27 km) crustal thicknesses reported for the Rehoboth terrane (Youssof *et al.*, 2013) (Fig. 1).  
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38 These values are the current best estimates of thickened Precambrian crust on and off the  
39 Kaapvaal craton in southern Africa (Stankiewicz & de Wit, 2013), as for example in the  
40 vicinity of the ca. 2056 Ma Bushveld Complex, where 5-10 km of mafic material had been  
41 'underplated' during this large igneous event (Wright *et al.*, 2003). Values for heat production  
42 of the upper ( $1.4 \mu\text{W}/\text{m}^3$ ) and lower ( $0.3 \mu\text{W}/\text{m}^3$ ) cratonic crust, as well as of the continental  
43 lithospheric mantle (zero), are taken from Rudnick & Nyblade (1999) and Michaut et al.  
44 (2007). In our paleogeotherm calculations for the central Kaapvaal craton at Mesoproterozoic  
45 times we applied a range of mantle potential temperatures between 1315 and 1420°C,  
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3 corresponding to estimates of ambient upper mantle temperatures at around 1.1 Ga (Davies,  
4 2009; Ganne & Feng, 2017); i.e., at the time of Premier kimberlite magma eruption.  
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## 8 9 **RESULTS**

### 10 11 12 **Petrography of the mantle-derived peridotite xenoliths from Premier**

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15 The investigated 15 garnet peridotite xenoliths range in size between 3 and 10 cm (Fig. 2).  
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17 Abundances of the primary rock-forming minerals were visually estimated during microscopy  
18 of polished petrographic thin sections (Table 1). The xenoliths are dominated by olivine  
19 (typically >70 vol.%) and contain variably small but significant amounts of orthopyroxene,  
20 clinopyroxene and garnet. On the basis of this mineral assemblage, the ultramafic xenoliths  
21 are identified as garnet lherzolites. Although the studied lherzolite nodules are very fresh,  
22 round to elongate garnet and orthopyroxene porphyroclasts typically exhibit a thin ‘coat’ of  
23 dark brown to black reaction products (Fig. 3). Applying the terminology of Harte (1977), the  
24 14 ‘sheared peridotite’ xenoliths have mosaic–porphyroclastic textures, whereas the ‘coarse  
25 peridotite’ nodule CIM15-40 lacks porphyroclasts and has a relatively homogeneous grain  
26 size distribution between 2–5 mm (Fig. 2D; Table 1). The sheared peridotites contain mm-  
27 sized porphyroclasts comprising single and composite grains of garnet, orthopyroxene,  
28 clinopyroxene and rare olivine, set in a fine-grained matrix of recrystallized olivine, where  
29 neoblast sizes typically range between 30–300 µm across (Figs. 3, 4). Although single-crystal  
30 porphyroclasts, often showing incipient subgrain formation along grain boundaries, may  
31 occur in isolation within the olivine-rich matrix, mm-sized lenses and stringers consisting of  
32 composite garnet–pyroxene assemblages are common (Figs. 3, 4). The lenses may form  
33 sheared and folded bands imparting a ‘fluidal’ rock texture (Fig. 4C), and their clinopyroxene  
34 constituents are commonly porous to spongy along the margins indicative of incipient melting  
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(Kopylova *et al.*, 2021). Orthopyroxene porphyroclasts also show evidence of partial melting such as the development of diffuse clinopyroxene coronas (Fig. 4E). Among the sheared peridotite xenoliths, sample CIM15-35 and CIM15-50 exhibit cm-sized round clusters or ‘pockets’ of ultra-coarse garnet and clinopyroxene assemblages set in a fine-grained olivine-rich matrix (Supplementary Data Figure 1). This ‘coarse-in-sheared’ texture may represent in situ megacryst formation. Ten discrete clinopyroxene megacrysts studied range in size between 1 and 3 cm across.

### Pressure–temperature array

Conventional thermobarometry was applied to estimate the P-T conditions for 14 sheared peridotite xenoliths from Premier (Table 1). The sheared peridotites equilibrated last at 5.2–5.6 GPa (~170–185 km depth) and 1208–1324°C, with the exception of CIM15-35 (4.8 GPa and 1088°C). The coarse peridotite xenolith CIM15-40 equilibrated last at much shallower conditions at ~2.8 GPa (~90 km depth) and ~809°C. Eight clinopyroxene megacrysts equilibrated over a wide P-T range between 4.1–6.3 GPa (~135–210 km depth) and 1026–1382°C (Fig. 5). Sample CUL17-cpx4 falls off the megacryst P-T trend (~7.2 GPa and ~1313°C; see Discussion) and CUL17-cpx8 is not suitable for single-grain thermobarometry due to its very low Cr content (Supplementary Data Appendix D).

All investigated samples, except for the ‘shallow’ coarse peridotite xenolith CIM15-40, are derived from the diamond stability field (Fig. 5) applying the graphite–diamond transition curve of Day (2012). The majority of analysed sheared peridotite xenoliths (n=12) form a cluster in P-T space between 5.2–5.6 GPa and 1294–1324°C (Fig. 5), recording warmer conditions than the 40 mW/m<sup>2</sup> model conductive geotherm of Hasterok & Chapman (2011). The cluster of high-temperature sheared peridotite analyses is equivalent to the ‘hot deformed’ peridotite xenolith suite from Premier discussed in Viljoen *et al.* (2009), for which

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3 we reprocessed the original data with our methodology where possible to ensure data  
4 compatibility. These authors noted that the Premier peridotite xenoliths fall between the 40  
5 and 45 mW/m<sup>2</sup> model geotherms of Pollack & Chapman (1977), which tend to be slightly  
6 warmer near the cratonic LAB than the model of Hasterok & Chapman (2011). Regardless,  
7 the P-T array fit for the peridotite xenoliths *versus* the fit for xenoliths plus clinopyroxene  
8 megacrysts (excluding CUL17-cpx4) yielded very similar geotherm shapes with the *FITPLOT*  
9 algorithm (Supplementary Data Figure 2). This suggests that most of the megacrysts studied  
10 equilibrated with the diamondiferous lithospheric mantle root of the central Kaapvaal craton  
11 by ca. 1150 Ma (see also Bell & Moore, 2004). Recently, Nimis *et al.* (2020) presented a  
12 large P-T dataset for clinopyroxene from Premier pipe, which comprises two subsets of grains  
13 that fall either along a ~38-40 mW/m<sup>2</sup> (*xenocrysts and inclusions in diamond*) or along a ~36-  
14 37 mW/m<sup>2</sup> (*inclusions in diamond*) model conductive geotherm of Hasterok & Chapman  
15 (2011). Importantly, our clinopyroxene megacryst CUL17-cpx4 falls onto the newly  
16 identified enigmatic Premier model geotherm of ~36-37 mW/m<sup>2</sup>. This rather ‘cold’ geotherm  
17 is subordinate to the more representative heat flow of ~40-42 mW/m<sup>2</sup> for the central Kaapvaal  
18 cratonic mantle root (Fig. 5).

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40 The newly devised conductive paleogeotherm of ~40-42 mW/m<sup>2</sup> for the lithospheric  
41 mantle column of the central Kaapvaal craton (Fig. 1), which incorporates the new P-T data  
42 into the existing body of work on peridotite xenoliths, intersects the modern 1315°C mantle  
43 adiabat at approximately 200 km depth, defining a thermal lithosphere–asthenosphere  
44 boundary *or* LAB at 1150 Ma (Fig. 5). If a 1420°C mantle adiabat is applied, catering for a  
45 slightly warmer convecting upper mantle during the Mesoproterozoic (Davies, 2009; Ganne  
46 & Feng, 2017), then the central Kaapvaal lithosphere may have reached down to ~225 km  
47 depth at 1150 Ma (Fig. 5). Note that the pressure–depth conversion applied in this study is  
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3 based on the ‘PREM’ model (Dziewonski & Anderson, 1981), in which 1 GPa lithostatic  
4 pressure is equivalent to ~33 km depth (see also Cammarano, 2013).  
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## 8 **Mineral major and trace element compositions**

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11 The major and trace element concentrations of mineral phases measured during this study are  
12 listed in Supplementary Data Appendix D, including mean and median values for each  
13 xenolith as used in pressure–temperature calculations (see above).  
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### 19 *Olivine*

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22 Sheared peridotites contain two generations of olivine, and the olivine Mg-numbers (atomic  
23  $Mg/[Mg+Fe^{2+}] \times 100$ ) for rare porphyroclasts and the abundant fine-grained neoblasts in the  
24 matrix overlap within the range of 89.9–91.7 (Fig. 6). These high Mg-number values are  
25 coupled to high Ni contents between 2947–3222 ppm. Olivine in sheared peridotite sample  
26 CIM15-23 has a lower Mg-number of ~87.6 and a lower Ni content of ~2750 ppm. Within  
27 individual sheared peridotite nodules the olivine major and minor element compositions are  
28 exceptionally homogenous in keeping with attainment of equilibrium. Olivine in the coarse  
29 peridotite xenolith CIM15-40 has a Mg-number value of ~91.7 and a Ni content of up to 3379  
30 ppm, which fall at the upper end of the ranges displayed by the sheared peridotites (Fig. 6). In  
31 general, the olivine compositions of the peridotite xenoliths from Premier pipe studied here  
32 are notably less refractory than depleted low-temperature peridotites from cratonic mantle  
33 roots worldwide (Fig. 6), which typically have average Mg-number values between 92.4–92.9  
34 (Boyd, 1989; Bernstein *et al.*, 2007; Pearson & Wittig, 2014).  
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### *Orthopyroxene*

Orthopyroxene in the sheared peridotite xenoliths has a near end-member enstatite composition (~90 mol.%) with Mg-number values ranging between 91.1–92.6 (~89.2 for CIM15-23). Higher Mg-numbers of orthopyroxene compared with coexisting olivine have also been reported for garnet peridotite xenoliths from Kimberley and Lesotho on the Kaapvaal craton, which testifies to attainment of orthopyroxene-olivine equilibrium (Simon *et al.*, 2007). As observed for olivine, orthopyroxene in coarse peridotite xenolith CIM15-40 exhibits a Mg-number at the upper end of the range displayed by the sheared peridotites from Premier (~92.6).

The sheared peridotites contain orthopyroxene porphyroclasts with sinusoidal to hump-shaped chondrite normalized REE patterns that show LREE and HREE depletion relative to the MREE (Fig. 7). Although the REE as a group are depleted relative to chondrite, concentrations in orthopyroxene from Premier tend to be elevated compared with orthopyroxene from Lesotho and Kimberley low-temperature garnet peridotite xenoliths, which show much stronger MREE-HREE depletion (Simon *et al.*, 2003; Simon *et al.*, 2007). Orthopyroxene from the coarse peridotite xenolith CIM15-40 has a chondrite normalized REE pattern that shows LREE enrichment over the MREE, and much stronger HREE depletion compared with the sheared peridotites from Premier, more similar to some orthopyroxene patterns identified for Lesotho and Kimberley low-temperature peridotite xenoliths (Simon *et al.*, 2003; Simon *et al.*, 2007) (Fig. 7). All analysed orthopyroxene crystals from Premier garnet peridotite xenoliths show negative Th-U and positive Nb-Ta-Ti anomalies in primitive mantle normalized multi-element diagrams (not shown). Orthopyroxene porphyroclasts from the sheared peridotites show subtle positive anomalies at

Hf in these diagrams, which is not observed for the coarse peridotite sample CIM15-40 (not shown).

### *Clinopyroxene*

Clinopyroxene porphyroclasts in the sheared peridotite xenoliths from Premier pipe are generally rich in Cr-diopside component with 0.59–1.73 wt.% Cr<sub>2</sub>O<sub>3</sub> and Mg-number values between 89.8–91.6 (CIM15-23 clinopyroxene Mg-number is ~87.5). The Na<sub>2</sub>O and TiO<sub>2</sub> contents are between 1.2–2 wt.% and 0.13–0.59 wt.%, respectively. Clinopyroxene in the coarse peridotite sample CIM15-40 has higher Cr<sub>2</sub>O<sub>3</sub> (~1.8 wt.%) and Na<sub>2</sub>O (~2.4 wt.%) concentrations, and a much higher Mg-number of ~93.5, whereas TiO<sub>2</sub> is relatively low at ~0.16 wt.%. The discrete clinopyroxene megacrysts display a wide range of Cr<sub>2</sub>O<sub>3</sub> (0.02–1.09 wt.%), Na<sub>2</sub>O (0.7–1.7 wt.%) and TiO<sub>2</sub> (0.09–0.51 wt.%) concentrations, with Mg-number values between 83.2–92.7. They share more compositional similarity with clinopyroxene porphyroclasts of the sheared peridotite xenoliths than with clinopyroxene in coarse peridotites from Premier (Viljoen *et al.*, 2009) (Fig. 8). Although the Premier clinopyroxene megacrysts have generally a ‘low-Cr’ nature, some crystals have higher Mg-number values and Cr<sub>2</sub>O<sub>3</sub> contents of >0.7 wt.% partially overlapping the lower end of ‘Cr-rich’ clinopyroxene megacryst suites from cratons worldwide (Fig. 8B).

Clinopyroxene in the sheared peridotite xenoliths from Premier is generally LREE enriched (>10 times chondrite), with maxima at Pr-Nd and consistent levels of HREE depletion in chondrite normalized distribution patterns (Fig. 9). The coarse peridotite xenolith CIM15-40 contains clinopyroxene with even stronger LREE enrichment approaching 100 times chondritic values compared with clinopyroxene in the sheared nodules (Fig. 9A). This LREE enrichment level is more similar to clinopyroxene in low-temperature garnet peridotite xenoliths from Kimberley on the western Kaapvaal craton (Simon *et al.*, 2007). The discrete



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3 clinopyroxene megacrysts from the Premier kimberlite pipe have identical chondrite  
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5 normalized REE patterns that fully overlap those of clinopyroxene porphyroclasts from the  
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7 sheared peridotite xenoliths (Fig. 9B). The only exception is megacryst CUL17-cpx4, which  
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9 has much lower REE and HFSE concentrations (see Discussion). The clinopyroxene  
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11 megacrysts and porphyroclasts from Premier also display very similar primitive mantle  
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13 normalized multi-element patterns with similar concentration levels of Th-U-Nb-Ta, Sr and  
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15 Zr-Hf (not shown).  
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### 18 19 20 *Garnet*

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23 Garnets from the studied Premier peridotite samples have ‘Iherzolitic G9’ affinities in terms  
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25 of their Mg, Ca and Cr compositions (Fig. 10), which is compatible with the presence of  
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27 significant clinopyroxene in these mantle-derived xenoliths (Sobolev *et al.*, 1973; Gurney &  
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29 Zweistra, 1995; Grütter *et al.*, 2004). The sheared peridotites contain garnet porphyroclasts  
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31 with Cr<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> concentrations between 1.9–6.7 wt.% and 0.48–1.56 wt.% respectively,  
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33 compositional ranges that completely overlap with garnet in ‘hot deformed’ peridotites from  
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35 Premier as reported in Viljoen *et al.* (2009). The pyrope-rich garnet porphyroclasts display a  
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37 narrow range of Mg-number values between 82.4–85.7, with the exception of garnet from  
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39 sheared peridotite xenolith CIM15-23, which has a lower Mg-number value of ~79.8, a trend  
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41 that is also observed for olivine and the pyroxenes (see above). Major element compositions  
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43 of garnet in the coarse peridotite xenolith CIM15-40 (e.g., Mg-number of ~82.9; ~2.5 wt.%  
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45 Cr<sub>2</sub>O<sub>3</sub>) fall within the range of garnet compositions from the sheared peridotite xenolith suite  
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47 (Fig. 10), except for the much lower TiO<sub>2</sub> content of ~0.1 wt.% (Fig. 11A), which is similar to  
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49 garnet in other coarse peridotite xenoliths from Premier (Viljoen *et al.*, 2009). The garnet  
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51 porphyroclasts in the sheared peridotite xenoliths resemble high-Cr megacrysts from  
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53 kimberlite localities worldwide with regard to their elevated Mg- and Cr-numbers (Fig. 11B).  
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3 However, their elevated  $\text{TiO}_2$  contents provide a good match to garnet compositions from  
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However, their elevated  $\text{TiO}_2$  contents provide a good match to garnet compositions from metasomatized peridotite xenoliths of the Kaapvaal cratonic root (Fig. 11A).

The garnet porphyroclasts in the sheared peridotite xenoliths have ‘normal’ chondrite normalized REE patterns, with strong LREE depletions and a flat plateau for the MREE–HREE (Fig. 12A). The REE pattern for garnet in the coarse peridotite xenolith CIM15-40 is similar except for a slightly stronger LREE depletion. The enriched and positively correlated Y-Zr systematics of garnet porphyroclasts in the sheared peridotites from Premier follow the global trend of lherzolitic garnets from metasomatized cratonic mantle lithosphere (Griffin & Ryan, 1995; Griffin *et al.*, 1999; Viljoen *et al.*, 2009; Hunt *et al.*, 2012; Gibson *et al.*, 2013; Shu & Brey, 2015; Shaikh *et al.*, 2020) (Fig. 13A). The extremely high Ti/Eu of these garnets, coupled with low Zr/Hf, suggests an involvement of silicate-rich rather than carbonate-dominated metasomatic agents (Shu & Brey, 2015) (Fig. 13B), but these trace element proxies do not always provide meaningful insights into mantle enrichment processes (see Discussion). Regardless, the low Ti/Eu and high Zr/Hf ratios of garnets in the coarse peridotite xenolith CIM15-40, and in previously analysed coarse peridotite nodules from Premier (Viljoen *et al.*, 2009), point to contrasting metasomatic styles in the lithospheric mantle beneath the central Kaapvaal craton (Fig. 13B). Prominent HFSE enrichment of the sheared peridotite garnet porphyroclasts is also apparent in primitive mantle normalized multi-element diagrams, where Zr-Hf forms a plateau and Nb-Ta and Ti display slightly positive spikes (not shown). In contrast, garnet from the coarse peridotite xenolith CIM15-40 is characterized by negative anomalies at Zr-Hf and Ti (not shown).

The REE distribution coefficients for coexisting garnet and clinopyroxene porphyroclasts in sheared peridotites from the Premier kimberlite pipe reveal an only subtle offset from equilibrium  $^{\text{garnet/cpx}}D_{\text{REE}}$  values (Fig. 14), which were determined both in

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3 experiments and in natural rocks for ultramafic-mafic silicate and carbonated silicate systems  
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5 between 920–1600°C and 1.5–10 GPa (Zack *et al.*, 1997; Green *et al.*, 2000; Adam & Green,  
6  
7 2006; Xiong, 2006; Girniss *et al.*, 2013). The offset from equilibrium is larger for garnet–  
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9 clinopyroxene pairs in the coarse peridotite xenolith CIM15-40. In general, the observed  
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11 subtle trace element disequilibrium in the Premier peridotite xenoliths, where clinopyroxene  
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13 is systematically more REE enriched than coexisting garnet, suggests a complex metasomatic  
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15 history of the central Kaapvaal mantle lithosphere (Simon *et al.*, 2003; Burgess & Harte,  
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17 2004), with some enrichment events having occurred only shortly prior to the kimberlite and  
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19 carbonatite magma eruptions at ca. 1150 Ma.  
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### 24 **Garnet $\text{Fe}^{3+}/\Sigma\text{Fe}$ compositions and sheared peridotite $f\text{O}_2$**

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27 Corrected  $\text{Fe}^{3+}/\Sigma\text{Fe}$  ratios for peridotite-derived garnets range between 0.070–0.088 (Table 1),  
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29 and these compositions were used to calculate sheared peridotite  $f\text{O}_2$  values relative to the  
30  
31 FMQ buffer applying the oxybarometer of Stagno *et al.* (2013). The oxybarometer yielded  
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33  $\Delta\log f\text{O}_2$  [FMQ] values between -2.80 and -2.17 (2S.E. of  $\pm 0.5$  log units) for five  
34  
35 representative sheared peridotite xenoliths from the Premier kimberlite pipe (Fig. 15). Four  
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37 high-temperature sheared peridotite xenoliths within the pressure range of 5.2–5.5 GPa  
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39 (1300–1324°C) are more reduced (-2.80 to -2.47  $\Delta\log f\text{O}_2$  units) than the slightly shallower  
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41 and cooler ( $\sim 1088^\circ\text{C}$ ) ‘megacrystic’ sheared peridotite nodule CIM15-35 at  $\sim 4.8$  GPa, which  
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43 has the highest  $\Delta\log f\text{O}_2$  value of FMQ-2.17 (Table 1).  
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49 Although this correlation is in alignment with the theoretically expected reduction of  
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51 peridotite  $f\text{O}_2$  with increasing pressure *or* depth (Woodland & Koch, 2003; Frost &  
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53 McCammon, 2008), the trend observed for the sheared peridotite xenoliths from Premier is  
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55 shifted by less than 0.5 log units toward slightly more oxidized  $\Delta\log f\text{O}_2$  values relative to the  
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57 most reduced garnet peridotite nodules known from the Kaapvaal craton (Fig. 15). The  
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3 oxidation state of the Premier sheared peridotites overlaps with  $fO_2$  values for both coarse and  
4 sheared peridotites from the diamondiferous lithospheric mantle root of the western Kaapvaal  
5 craton (Supplementary Data Appendix B); e.g., from Kimberley and Finsch (Woodland &  
6 Koch, 2003; Creighton *et al.*, 2009; Lazarov *et al.*, 2009; Hanger *et al.*, 2015). Importantly,  
7 the diamondiferous root of the central Kaapvaal craton, as probed by the sheared garnet  
8 peridotite xenoliths from the ca. 1150 Ma Premier kimberlite pipe, is not as highly oxidized as  
9 the deformed mantle roots of the Slave (Creighton *et al.*, 2010; Yaxley *et al.*, 2017) and  
10 Siberian cratons (Goncharov *et al.*, 2012; Yaxley *et al.*, 2012) (Fig. 15).

### 21 22 **Clinopyroxene Sr-Nd-Hf isotopic compositions**

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24 Clinopyroxene porphyroclasts separated from five sheared peridotite xenoliths from the  
25 Premier kimberlite pipe define a subhorizontal array in Sr-Nd isotope space. The initial  
26  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios are between 0.703256–0.707224 and 0.511304–0.511334,  
27 respectively. Only xenolith CIM15-35 falls off the Sr-Nd isotope array due to elevated initial  
28  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{143}\text{Nd}/^{144}\text{Nd}$ . The Nd isotopic compositions of the sheared peridotites are  
29 moderately depleted as revealed by a narrow range of suprachondritic initial  $\epsilon\text{Nd}$  values  
30 between +3 and +3.6. For the Premier peridotite xenoliths, we were only able to determine the  
31 Hf isotopic composition of clinopyroxene from the sheared sample CIM15-50 at high  
32 precision, and the initial  $\epsilon\text{Hf}$  value is suprachondritic at +2.1. Coarse peridotite xenolith  
33 CIM15-40, which equilibrated at much lower pressure at ~2.8 GPa, has similar initial  
34  $^{87}\text{Sr}/^{86}\text{Sr}$  (0.704973) and  $^{143}\text{Nd}/^{144}\text{Nd}$  (0.511303; equivalent to an initial  $\epsilon\text{Nd}$  value of +3)  
35 ratios compared with the deeper derived sheared peridotites at 5.2–5.6 GPa (Fig. 16A).

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53 The ten low-Cr clinopyroxene megacrysts from Premier pipe show a similar range of  
54 initial  $^{87}\text{Sr}/^{86}\text{Sr}$  (0.702235–0.706073), but a much wider range of initial  $^{143}\text{Nd}/^{144}\text{Nd}$   
55 (0.511223–0.511354; equivalent to an initial  $\epsilon\text{Nd}$  range of +1.4 to +4.0) compared to

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3 clinopyroxene hosted in the peridotite xenoliths. The initial  $^{176}\text{Hf}/^{177}\text{Hf}$  compositions of the  
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5 clinopyroxene megacrysts range between 0.282063–0.282154, which is equivalent to an  
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7 initial  $\epsilon\text{Hf}$  range of +0.2 to +3.4 (Fig. 16B). In Nd-Hf isotope space, the clinopyroxene  
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9 megacrysts and the only successfully analysed sheared peridotite nodule from Premier pipe  
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11 fall mostly within the mantle array (initial  $\Delta\epsilon\text{Hf}$  of -2.6 to -6.9; for definition see footnote to  
12  
13 Table 2), in contrast to the more strongly decoupled Nd-Hf isotope systematics of the host  
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15 Premier kimberlite (initial  $\Delta\epsilon\text{Hf}$  of -7.6 to -10.7; Tappe et al., 2020b). There is, however, a  
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17 small overlap between the Nd-Hf isotopic compositions of the clinopyroxene megacrysts and  
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19 enigmatic carbonatite dykes at Premier pipe (initial  $\Delta\epsilon\text{Hf}$  of -6.1 to -8.8; Tappe et al., 2020b)  
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21 (Fig. 16B).  
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27 We note that sheared peridotite xenoliths and clinopyroxene megacrysts from the same  
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29 depth interval at the craton base have similar Sr-Nd isotopic compositions. We note further  
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31 that clinopyroxene megacrysts from >5.5 GPa contain isotopically more enriched Sr with  
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33 initial  $^{87}\text{Sr}/^{86}\text{Sr} > 0.7050$  compared to shallower megacrysts, although this trend with depth is  
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35 less clear for Nd and Hf isotopes (Fig. 17; Table 2).  
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### 39 **Calcium isotopic compositions of clinopyroxene and garnet**

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42 Aliquots of clinopyroxene sample solutions prepared for Sr-Nd-Hf isotope analytical work  
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44 were processed and analysed for stable Ca isotopic compositions, expressed here as  $\delta^{44/40}\text{Ca}$   
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46 in per mil relative to SRM-915a (Table 2). We also analysed garnet grains that were left over  
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48 after Mössbauer spectroscopy  $\text{Fe}^{3+}$  determinations (see above) for their  $\delta^{44/40}\text{Ca}$  compositions.  
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50 Clinopyroxene from five sheared peridotite xenoliths displays a range in  $\delta^{44/40}\text{Ca}$  values  
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52 between 0.78–1.00‰ (Fig. 18), and the  $\delta^{44/40}\text{Ca}$  values for coexisting garnet grains range  
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54 between 1.06–1.24‰, in excellent agreement with the known clinopyroxene–garnet Ca  
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3 isotope equilibrium fractionation of 0.2–0.6‰ at high temperatures (Antonelli *et al.*, 2019;  
4 Huang *et al.*, 2019; Wang *et al.*, 2019; Smart *et al.*, 2021a). The coarse peridotite xenolith  
5 CIM15-40 contains clinopyroxene and garnet with corresponding  $\delta^{44/40}\text{Ca}$  values of 0.95‰  
6 and 1.87‰, which suggests Ca isotopic disequilibrium for this low-temperature peridotite  
7 sample (Table 2). The  $\delta^{44/40}\text{Ca}$  values of the clinopyroxene megacrysts range between 0.72–  
8 0.94‰, with exception of crystal CUL17-cpx4, which has a value of 1.38‰ (Fig. 18). We  
9 note that deeper derived clinopyroxene megacrysts from >5.5 GPa reveal more notable Ca  
10 isotopic heterogeneity compared with the shallower megacrysts (Fig. 17; Table 2). We also  
11 analysed fresh bulk rock materials from four kimberlite dykes at Premier pipe for their  
12  $\delta^{44/40}\text{Ca}$  compositions, which show a wide range between 0.73–1.27‰ (Supplementary Data  
13 Appendix E).  
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## 29 **DISCUSSION: THE DYNAMIC KAAPVAAL CRATON LITHOSPHERE**

### 30 **Sheared peridotite formation across the lithosphere–asthenosphere transition**

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32 Petrological and geophysical models often define different depths for the cratonic LAB.  
33 Whereas results from seismological methods have frequently suggested craton thicknesses in  
34 excess of 250 km (Jordan, 1978; James *et al.*, 2001), geotherms derived from peridotite  
35 xenoliths typically project LAB positions between 150–250 km depths (Sleep, 2003; Sand *et*  
36 *al.*, 2009; Mather *et al.*, 2011; Gibson *et al.*, 2013; Kopylova *et al.*, 2019). O'Reilly & Griffin  
37 (2010) pointed out that petrological–geochemical methods tend to detect the horizon within  
38 the cratonic mantle lithosphere where the shallower depleted mantle transitions into  
39 refertilized *or* less depleted portions of a >250 km thick cratonic mantle root, which is strictly  
40 speaking not equivalent to the LAB, but conforms to earlier concepts of a  
41 mechanical/chemical *versus* a thermal boundary layer (McKenzie & Bickle, 1988). Other  
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3 authors also advocated for the existence of much deeper cratonic roots that extend down to  
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5 400 km depth approaching the mantle transition zone (Jordan, 1988; Masters *et al.*, 1996).  
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7 However, recently improved treatments of global seismological datasets suggest that a  
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9 lithosphere–asthenosphere transition occurs beneath cratons at shallower levels of less than  
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11 230 km depth, roughly overlapping the petrologically defined LAB (Mancinelli *et al.*, 2017;  
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13 Tharimena *et al.*, 2017; Priestley *et al.*, 2019; Rychert *et al.*, 2020).  
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17 We derived a refined paleogeotherm of  $\sim 40\text{--}42$  mW/m<sup>2</sup> for the central Kaapvaal craton  
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19 at ca. 1150 Ma through regression of both previously published and the new P-T data,  
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21 obtained for both coarse and sheared garnet-facies peridotite xenoliths from Premier pipe  
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23 (Fig. 5). The P-T array fitting of exclusively coarse peridotite xenoliths resulted in an only  
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25 marginally cooler steady-state geotherm of  $\sim 41$  mW/m<sup>2</sup> compared to the regression that  
26  
27 incorporated all textural types of garnet-facies peridotite (Fig. 5). The P-T data for discrete  
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29 clinopyroxene megacrysts independently define a conductive geotherm of  $40\text{--}41$  mW/m<sup>2</sup>  
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31 (Supplementary Data Figure 2), which is similar to the  $\sim 38\text{--}40$  mW/m<sup>2</sup> geotherm defined by  
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33 clinopyroxene xenocrysts from the Premier kimberlite (Nimis *et al.*, 2020). Our new  $\sim 40\text{--}42$   
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35 mW/m<sup>2</sup> conductive paleogeotherm for the lithospheric mantle column of the central Kaapvaal  
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37 craton intersects the 1315°C mantle adiabat at approximately 200 km depth, defining the  
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39 thermal LAB at 1150 Ma (Fig. 5). Adjusting the potential temperature of the ambient upper  
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41 mantle to 1420°C, as may be more realistic for the convecting mantle during the Proterozoic  
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43 (Davies, 2009; Ganne & Feng, 2017), increases the thickness of the central Kaapvaal  
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45 lithosphere to  $\sim 225$  km at 1150 Ma (Fig. 5), which is in good agreement with the estimate of  
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47  $\sim 230$  km based on single-grain clinopyroxene xenocryst thermobarometry (Nimis *et al.*,  
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49 2020). Taking these new estimates of the LAB beneath the central Kaapvaal craton into  
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51 account, it appears that the sheared peridotites studied here and by Viljoen *et al.* (2009)  
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53 formed near the craton base in some vertical distance of  $\sim 10\text{--}50$  km above this thermal and  
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3 structural/rheological discontinuity. Three of the analysed clinopyroxene megacrysts are  
4 derived from ~190–210 km depth suggesting formation at the cratonic LAB, if the 1420°C  
5 mantle potential temperature scenario is considered (Fig. 5).  
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10 Clinopyroxene megacryst CUL17-cpx4 is anomalous in that it is derived from great  
11 depth (~230 km) but records a relatively low temperature of ~1300°C (Fig. 5). Similar P-T  
12 systematics corresponding to a ~36-37 mW/m<sup>2</sup> conductive paleogeotherm are reported for  
13 some clinopyroxene xenocrysts and a suite of clinopyroxene inclusions in diamond from  
14 Premier pipe (Supplementary Data Figure 2). This observation has been tentatively  
15 interpreted as a record of deep and old lithospheric mantle that predates Proterozoic thermal  
16 perturbations (Nimis *et al.*, 2020). The much less enriched or even depleted trace element  
17 composition of CUL17-cpx4 supports this idea; e.g., this megacryst only contains 0.05 ppm  
18 Hf (Fig. 9B; Supplementary Data Appendix D). In addition, CUL17-cpx4 has the highest  
19  $\delta^{44/40}\text{Ca}$  value (1.38‰) of the suite analysed (0.72–0.94‰; Fig. 18), and such <sup>44</sup>Ca enriched  
20 clinopyroxene compositions relative to ‘normal’ upper mantle are probably a consequence of  
21 melt extraction (Chen *et al.*, 2019; Smart *et al.*, 2021a). Thus, the Ca isotopic evidence also  
22 supports the presence of remnant deep lithospheric mantle ‘pockets’ that remained largely  
23 unaffected by younger metasomatic (and thermal) events. Such materials may be more  
24 common in the lower cratonic mantle lithosphere than is generally accepted, and a few studies  
25 suggest indeed that ‘kimberlite sampling patterns’ are skewed toward metasomatically  
26 overprinted mantle rocks and minerals (Malkovets *et al.*, 2007; Artemieva *et al.*, 2019).  
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49 A paleogeotherm of ~40-42 mW/m<sup>2</sup> intersects the solidus of CO<sub>2</sub>-H<sub>2</sub>O-fluxed  
50 (Massuyeau *et al.*, 2021) or CO<sub>2</sub>-bearing peridotite (Dasgupta & Hirschmann, 2006) between  
51 ~130–160 km, which means that small volumes of ephemeral carbonated silicate melts may  
52 be ubiquitous at ≥150 km depths at the relatively reducing *f*O<sub>2</sub> conditions recorded by the  
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3 Premier garnet peridotite xenoliths (Stagno *et al.*, 2013) (Fig. 15). The presence of these  
4 volatiles and partial melt component (see the melting-induced textures in Figure 4) decreases  
5 the viscosity of peridotitic mantle and thus enhances plastic flow and heat transfer (Faul,  
6 1997; Tharimena *et al.*, 2017). Our model suggests that the depth interval between ~150–225  
7 km presents a cratonic lithosphere–asthenosphere transition zone, comparable to the concept  
8 of a thermal boundary layer in earlier studies (McKenzie, 1989). Importantly, the majority of  
9 the analysed sheared peridotite xenoliths equilibrated between ~170–185 km depth (see also  
10 Viljoen *et al.*, 2009), firmly within this boundary layer. The most shallowly derived sheared  
11 peridotite xenolith CIM15-35 equilibrated at ~160 km depth in close proximity to the deepest  
12 confirmed coarse peridotite sample of Viljoen *et al.* (2009) (Fig. 5). This textural and spatial  
13 evidence supports the position of the upper limit of the lithosphere–asthenosphere transition  
14 zone as inferred primarily on the basis of petrological constraints.

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31 Baptiste *et al.* (2012) presented OH<sup>-</sup> concentration data for olivine from Kaapvaal  
32 craton peridotite xenoliths from several kimberlite localities of mainly Mesozoic age. These  
33 data demonstrate that the lithospheric mantle at greater than 150 km depth is surprisingly dry  
34 compared to the shallower cratonic mantle (Baptiste *et al.*, 2012), a pattern that had  
35 previously emerged based on a preliminary dataset for the Kaapvaal craton (Peslier *et al.*,  
36 2010). Although the water systematics of the Kaapvaal lithospheric root are generally  
37 opposite to trends observed for other continental mantle columns worldwide (Doucet *et al.*,  
38 2014; Demouchy & Bolfan-Casanova, 2016; Kilgore *et al.*, 2020), experimental data suggest  
39 that melt-driven metasomatism in the presence of abundant mixed CO<sub>2</sub>-H<sub>2</sub>O volatiles results  
40 in rather dry olivine compositions (Baptiste *et al.*, 2015) that may subsequently dominate a  
41 thermally and chemically overprinted cratonic root. Eaton *et al.* (2009) integrated olivine  
42 creep data into thermal models and concluded that if the LAB comprises mainly dry  
43 peridotites, then this boundary layer is probably diffuse and >50 km thick. In this scenario

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3 extremely high shear stresses are required to accommodate tectonic plate motion. In contrast,  
4 a wet LAB would cause this boundary to be <20 km thick and relatively sharp, with much  
5 lower shear stresses caused by the differential motion between the lithospheric plate and the  
6 underlying convecting mantle. Relatively dry sheared peridotites from the Premier kimberlite  
7 pipe, together with the observations from other localities by Baptiste *et al.* (2012), support a  
8 case for a thick and diffuse LAB beneath the Kaapvaal craton at 1150 Ma, where plate  
9 tectonic movements may have created a several 10s of kilometers thick deformed layer at the  
10 base of the lithosphere.  
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22 Some earlier studies of sheared peridotites argued that the high equilibration  
23 temperatures and mylonitic microstructures, such as extremely reduced grain sizes and  
24 laminated-disrupted porphyroclastic textures, record strain rates that are too high to be  
25 explained by mantle flow at the LAB, or plate tectonic movements in general (Green &  
26 Gueguen, 1974; Goetze, 1975; Skemer & Karato, 2008; Baptiste *et al.*, 2012). In these  
27 studies, sheared peridotite formation at >150 km depth within the lower reaches of cratonic  
28 mantle lithosphere had been ascribed to strong plastic deformation associated with ascending  
29 kimberlite magma diapirs [see also Gregoire *et al.* (2006) and Arndt *et al.* (2010)]. However,  
30 mylonite formation is common in crustal shear zones and thrust faults and can be readily  
31 explained by the differential movement of rock packages at strain rates that are comparable to  
32 those in the deep continental mantle lithosphere (Le Roux *et al.*, 2008; Lee *et al.*, 2018). We  
33 argue that rather than being restricted to a narrow zone at the base of the craton where mantle  
34 flow causes viscous drag (Boyd & Nixon, 1975; Kennedy *et al.*, 2002), sheared peridotite  
35 formation may occur in localized meter-thick bands spaced out across the entire width of the  
36 lithosphere–asthenosphere transition zone between ~150–225 km depth. This idea is  
37 supported by the facts that (1) sheared peridotites have protoliths that resemble ancient coarse  
38 garnet peridotites (Harte, 1977; Walker *et al.*, 1989; Pearson *et al.*, 1995), and (2) that such  
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3 coarse garnet peridotites occur across a large portion of the Kaapvaal cratonic mantle column  
4 from 80 km down to 215 km depth (Katayama *et al.*, 2009; Viljoen *et al.*, 2009; Baptiste *et*  
5 *al.*, 2012). Thus, it is reasonable to assume that relatively undeformed coarse peridotite  
6 materials and strongly deformed sheared peridotites are structurally interleaved between  
7 ~150–200 km depth, where most of the tectonic strain may be accommodated by a network of  
8 relatively narrow subhorizontal shear zones (Fig. 19). It should be noted, however, that  
9 modern geobarometry of garnet-bearing peridotites is typically associated with uncertainties  
10 of ~0.2–0.4 GPa or ±10 km (Mather *et al.*, 2011), such that relative depth distributions of  
11 coarse and sheared peridotite varieties as building blocks of cratonic mantle roots cannot be  
12 resolved more precisely.  
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26 Despite the intense plastic deformation of sheared peridotites under extreme deviatoric  
27 stress, most xenoliths of this textural group exhibit a recrystallized olivine-rich matrix that  
28 records evidence of effective recovery from strain, such as grain boundary migration under  
29 static conditions (e.g., olivine tablet formation; Fig. 4). This evidence for annealing has been  
30 used as an argument against a spatial association of sheared peridotites with the cratonic LAB  
31 (Baptiste *et al.*, 2012). However, in our model the observed annealing microstructures are  
32 readily explained by mantle shear zones going through cycles of intense movement and  
33 dormancy (i.e., non-steady state deformation); that is, they are active only intermittently,  
34 similar to major shear zones in the Earth's crust (Fossen & Cavalcante, 2017). In other words,  
35 we suggest that plate tectonically induced stresses onto the lower cratonic mantle are  
36 distributed unevenly (i.e., strain partitioning) over extensive networks of subhorizontal shear  
37 bands across the entire  $\geq 50$  km thick lithosphere–asthenosphere transition zone (Fig. 5, 19).  
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## The enigmatic connection between sheared peridotites and megacrysts

Although olivine in peridotites from the lower Kaapvaal craton lithosphere appears to have lower OH<sup>-</sup> contents than in the shallow cratonic mantle at <150 km depth (Peslier *et al.*, 2010; Baptiste *et al.*, 2012), past activity of fluids and melts in the sheared peridotitic mantle, as probed by the kimberlite-borne xenoliths from Premier pipe derived from ~160–185 km depth, is evidenced by the annealing textures of the olivine-rich matrix. Undeformed mosaic textures, including olivine tablet formation in the matrix of sheared cratonic peridotites (Figs. 3, 4), have been explained by fluid-assisted static recrystallization (Drury & van Roermund, 1989). The former presence of fluid/melt is also recorded in the REE patterns of clinopyroxene in the sheared peridotites and megacrysts from Premier pipe, suggesting a common origin by metasomatic reactions within the lower Kaapvaal craton lithosphere (Fig. 9). This trace element evidence is supported by melting-induced texture development such as the porous tails around and spongy margins along clinopyroxene porphyroclasts in the sheared peridotite xenoliths (Fig. 4).

The Premier paleogeotherm of ~40–42 mW/m<sup>2</sup> intersects the solidus of CO<sub>2</sub>-H<sub>2</sub>O-bearing (Massuyeau *et al.*, 2021) or CO<sub>2</sub>-bearing peridotite (Dasgupta & Hirschmann, 2006) between ~130–160 km depth, which suggests that an influx of CHO volatile mixtures should stabilize low-degree partial melts with carbonate-rich to carbonated silicate compositions at ≥150 km depth (Foley *et al.*, 2009; Aulbach *et al.*, 2017), even under relatively reducing conditions (Stagno & Frost, 2010). Several studies suggest that megacryst suites in kimberlites on cratons worldwide formed from percolating melts within the lower cratonic mantle lithosphere (Hops *et al.*, 1992; Bell & Moore, 2004; Moore & Belousova, 2005; Kopylova *et al.*, 2009; Bussweiler *et al.*, 2018; Cone & Kopylova, 2021). The Premier clinopyroxene megacrysts studied formed over a wide P-T range between ~135–210 km depth

(Fig. 5). In detail, a shallower megacryst subgroup at ~135–150 km depth can be distinguished from a deeper subgroup at ~190–210 km depth. The apparent clustering of clinopyroxene megacrysts near the top and bottom of the central Kaapvaal lithosphere–asthenosphere transition zone may reflect stalling of melt in channels beneath permeability boundaries (e.g., Sleep, 2003; Rychert *et al.*, 2020). Nimis *et al.* (2020) observed a similar bimodal depth distribution for a large clinopyroxene xenocryst suite from Premier pipe that probably comprises a significant portion of fragmented megacrysts. Importantly, the clinopyroxene megacrysts from both depth horizons have similar geochemical and Sr-Nd-Hf isotopic compositions (Figs. 9, 16), which suggests a common melt source and similar magma evolution. We note, however, that the shallower megacryst subgroup shows slightly lower initial  $^{87}\text{Sr}/^{86}\text{Sr}$  and slightly higher initial  $^{176}\text{Hf}/^{177}\text{Hf}$  relative to the deeper clinopyroxene subgroup (Fig. 17; see below). It is noted further that the Ca isotopic compositions of the deepest derived clinopyroxene megacrysts from ~190–210 km depth are more variable compared to their counterparts from mid-lithospheric depth, which exhibit exclusively mantle-like  $\delta^{44/40}\text{Ca}$  values (Figs. 17B, 18).

Clinopyroxene porphyroclasts in sheared peridotite xenoliths from Premier show a striking overlap with the clinopyroxene megacrysts in terms of major and trace element compositions as well as Sr-Nd-Hf-Ca isotope ratios (Figs. 8, 9, 16, 18). The clinopyroxene porphyroclasts (~160–185 km depth) exhibit the same range of  $\delta^{44/40}\text{Ca}$  values as their megacrystic counterparts from ~190–210 km depth; i.e., 0.78–1.00‰ and 0.72–0.94‰  $\delta^{44/40}\text{Ca}$ , respectively (Figs. 17B, 18). These clinopyroxene Ca isotopic compositions are also similar to those of the host kimberlite magmas with  $\delta^{44/40}\text{Ca}$  values as low as 0.73‰ (Fig. 18; Supplementary Data Appendix E). The clinopyroxene Sr-Nd isotopic compositions from both megacrysts and sheared peridotites are moderately depleted to slightly enriched, overlapping the compositions of kimberlite and carbonatite magmas from Premier pipe (Fig. 16A).

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3 However, in Nd-Hf isotope space there is only a match between the mantle-derived  
4 clinopyroxene crystals (megacrysts and sheared peridotite) and carbonatite magmas, falling  
5 slightly below the mantle array (Fig. 16B). In contrast, the Premier kimberlites show strong  
6 Nd-Hf isotope decoupling significantly below the mantle regression line (Tappe *et al.*,  
7  
8 2020b). Combined, the isotope data reveal that clinopyroxene formation or heavy  
9 overprinting by melt/fluid must have been a relatively ‘young’ event that occurred only  
10 shortly prior to kimberlite and carbonatite magmatism on the central Kaapvaal craton at ca.  
11 1150 Ma. The data suggest direct links between passing carbonatitic to kimberlitic melts,  
12 localised plastic deformation and clinopyroxene formation during metasomatic reactions  
13 within the lower lithospheric mantle, as had been proposed for other regions on the Kaapvaal  
14 craton (Gurney & Harte, 1980; Moore & Lock, 2001). Tappe *et al.* (2018a) presented high-  
15 precision geochronology data for Premier pipe and suggested that the kimberlite–carbonatite  
16 magmatic system was active for >3 million years, which provides ample time for metasomatic  
17 overprinting of portions within the lower lithospheric mantle, including megacryst formation.  
18 Although vestiges of undisturbed Archean lower cratonic mantle are preserved in some  
19 peridotitic clinopyroxene and garnet inclusions in diamonds from Premier pipe showing  
20 extremely depleted  $\epsilon\text{Nd}$  values of up to +66 (Richardson *et al.*, 1993), no such ancient  
21 material has been traced in the formation of metasomatic clinopyroxene within sheared  
22 peridotitic mantle and associated megacrysts (Fig. 16). However, clinopyroxene crystal  
23 CUL17-cpx4 is anomalous and falls on a relatively cold 36–37 mW/m<sup>2</sup> cratonic geotherm  
24 (Fig. 5), similar to several clinopyroxene inclusions in diamond and few clinopyroxene  
25 xenocrysts from the Premier kimberlite, which have apparently not adjusted to the elevated  
26 heat flow of the central Kaapvaal cratonic mantle root, with values of ~39–42 mW/m<sup>2</sup> at 1150  
27 Ma (Viljoen *et al.*, 2009; Nimis *et al.*, 2020; this study).  
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3 To further constrain possible links between clinopyroxene in sheared peridotites and  
4 the discrete megacrysts via kimberlitic to carbonatitic melt metasomatism, we calculated the  
5 REE contents of hypothetical melts in equilibrium with clinopyroxene (Fig. 9C). Both the  
6 REE contents of hypothetical melts in equilibrium with clinopyroxene (Fig. 9C). Both the  
7 effects of carbonate-dominated (cpx/melt partition coefficients from Dasgupta *et al.*, 2009 and  
8 Girnir *et al.*, 2013) and carbonated silicate (Keshav *et al.*, 2005) melts at conditions  
9 equivalent to those near cratonic LABs are shown in Figure 9C. The reconstructed  
10 equilibrium melt compositions that may have been involved in metasomatic clinopyroxene  
11 formation are highly enriched in LREE–MREE, with concentration levels up to an order of  
12 magnitude higher than in the natural carbonatite and kimberlite dyke magmas known from  
13 Premier pipe (Tappe *et al.*, 2020b). The best fit between model and nature is provided by the  
14 extremely high LREE–MREE concentrations of saline and high-Mg carbonatitic high-density  
15 fluids (HDFs) entrapped in diamonds from lithospheric mantle columns of cratons worldwide  
16 (Weiss *et al.*, 2011; Weiss *et al.*, 2018). Hence, carbonatitic to kimberlitic melts plus  
17 associated high-density fluids may have been responsible for metasomatic clinopyroxene  
18 formation within the lithosphere–asthenosphere transition zone of the central Kaapvaal  
19 cratonic root at ca. 1150 Ma.

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41 Several other studies also ascribed metasomatic origins to garnet in cratonic mantle  
42 peridotites (Malkovets *et al.*, 2007; Simon *et al.*, 2007; Klein-BenDavid & Pearson, 2009;  
43 Chepurov *et al.*, 2019; Kopylova *et al.*, 2019), and the high TiO<sub>2</sub> contents of ≥0.5 wt.% as  
44 well as elevated Mg- and Cr-number values of garnet in sheared peridotites from Premier lend  
45 support to this idea (Fig. 11). We used a range of equilibrium  $Grt/CpxD$  values for the REE  
46 (Zack *et al.*, 1997; Green *et al.*, 2000; Adam & Green, 2006; Xiong, 2006; Girnir *et al.*, 2013)  
47 to evaluate trace element equilibrium between metasomatic garnet and clinopyroxene in the  
48 sheared peridotite xenoliths from Premier. Our data show that clinopyroxene is slightly more  
49 REE enriched than predicted by the composition of coexisting garnet (Fig. 14), which

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3 indicates a subtle trace element disequilibrium between these phases and suggests distinct  
4 origins during multiple metasomatic events. The Y-Zr and Zr/Hf-Ti/Eu systematics of garnet  
5 in the sheared peridotite xenoliths reveal strong HFSE enrichments at constant and low Zr/Hf  
6 (Fig. 13), a geochemical signature that is typically ascribed to silicate melt metasomatism  
7 (Griffin & Ryan, 1995; Griffin *et al.*, 2003; Viljoen *et al.*, 2009; Shaikh *et al.*, 2020). High  
8 Zr/Hf ratios at low Ti/Eu are observed for garnet in the coarse low-temperature peridotite  
9 xenoliths from Premier (Fig. 13B), and this contrasting signature points to carbonate-  
10 dominated melt metasomatism of the shallow cratonic mantle beneath Premier pipe (Rudnick  
11 *et al.*, 1993; McCoy-West *et al.*, 2015; Shu & Brey, 2015). However, Tappe *et al.* (2017)  
12 showed that natural primitive carbonated silicate magmas with up to 25 wt.% CO<sub>2</sub> derived  
13 from >200 km depth can be extremely enriched in Ti and Zr, which complicates use of the  
14 HFSE systematics of mantle-derived xenoliths in our quest for the nature and origin of  
15 metasomatic agents within the lower cratonic lithosphere. For example, both the action of  
16 carbonate-dominated (garnet/melt partition coefficients from Dasgupta *et al.*, 2009 and Girmis  
17 *et al.*, 2013) and carbonated silicate *or* kimberlitic (Brey *et al.*, 2008) melts at P-T conditions  
18 equivalent to those of lower cratonic lithosphere and slightly deeper can explain the observed  
19 REE compositions of metasomatic garnet in sheared peridotites from Premier, without  
20 involvement of pure silicate melts such as basaltic liquid compositions. Regardless of the  
21 exact nature of the garnet-forming metasomatic agent, in the absence of radiogenic isotope  
22 data for garnet from the sheared peridotite xenoliths it is impossible to constrain the timing of  
23 this mantle enrichment event. However, on the basis of attained major element equilibrium  
24 between garnet and clinopyroxene (e.g., Ca isotope equilibrium, compatibility of Fe-Mg  
25 exchange temperatures with other thermometer results), but subtle trace element  
26 disequilibrium (Fig. 14), it can be inferred that all of these identified metasomatic events  
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3 occurred shortly prior to kimberlite magmatism during the Mesoproterozoic (see Jollands *et*  
4 *al.*, 2018).  
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### 8 **Bushveld mantle plume impact on the Kaapvaal cratonic root revisited**

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11 Several studies link the slightly elevated cratonic geotherm and relatively fertile nature of the  
12 central Kaapvaal mantle lithosphere, as probed by the ca. 1150 Ma Premier kimberlite, to the  
13 magmatic events associated with the ca. 2056 Ma Bushveld layered mafic–ultramafic  
14 intrusion (Danchin, 1979; Griffin *et al.*, 2003; Gregoire *et al.*, 2005; Richardson & Shirey,  
15 2008; Viljoen *et al.*, 2009; Viljoen *et al.*, 2014; Korolev *et al.*, 2018b). However, some of the  
16 previously discussed evidence is ambiguous or circumstantial, and simpler explanations exist.  
17 Because impingement of the putative Bushveld mantle plume on thick diamondiferous  
18 continental lithosphere frequently serves in metallogenesis models (Arndt, 2013; Griffin *et*  
19 *al.*, 2013; Fiorentini *et al.*, 2020), a brief re-examination of this contentious topic in light of  
20 our new evidence appears warranted.  
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35 For Premier pipe, a high abundance of Ti-rich garnet in sheared peridotite xenoliths  
36 (Fig. 11A) and from heavy mineral concentrate has been ascribed previously to melt  
37 metasomatism that was potentially related to basaltic magmatism of the Bushveld event  
38 (Griffin *et al.*, 2003; Kobussen *et al.*, 2009; Viljoen *et al.*, 2009). New research shows,  
39 however, that the parental magmas to the giant Bushveld layered intrusion had a low-Ti  
40 komatiitic nature (Wilson, 2012; Maier *et al.*, 2016; Solovova *et al.*, 2021) and were probably  
41 incapable of causing the observed Ti-enrichment of the lower cratonic mantle. Instead, our  
42 major and trace element results for such metasomatic garnet porphyroclasts show that an  
43 origin from carbonated silicate melts, such as kimberlitic liquids, is more likely (see the  
44 previous section). Moreover, the observed subtle trace element disequilibrium between  
45 metasomatic garnet and clinopyroxene suggests that volatile-driven refertilization events  
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3 occurred shortly prior to Mesoproterozoic kimberlite and carbonatite magmatism on the  
4 central Kaapvaal craton. Based on the garnet  $\text{Fe}^{3+}$  compositions, it appears that the oxidative  
5 effects of this volatile-rich mantle metasomatism were only minor when compared with the  
6 highly oxidized and deformed mantle roots beneath some regions on the Slave and Siberian  
7 cratons (Creighton *et al.*, 2010; Goncharov *et al.*, 2012; Yaxley *et al.*, 2012; Yaxley *et al.*,  
8 2017) (Fig. 15). Recently, Moussallam *et al.* (2019) demonstrated the highly oxidized nature  
9 of mafic–ultramafic silicate melts from major modern mantle plumes worldwide, which have  
10 primary  $f\text{O}_2$  values of up to two log units above the FMQ buffer. Several Paleoproterozoic  
11 large igneous events were also supplied with magma from relatively oxidized mantle sources  
12 (Nicklas *et al.*, 2019). However, the putative Bushveld plume impact on the central Kaapvaal  
13 cratonic root left no such anomalous imprint on the lithospheric mantle redox composition.  
14 This argument is also consistent with the observation that Premier garnet-facies peridotite  
15 xenoliths have similar  $f\text{O}_2$  values to deep-sourced peridotite xenolith suites from other regions  
16 of southern Africa, which have been more distal to the possible effects of the Bushveld large  
17 igneous event at ca. 2056 Ma (Woodland & Koch, 2003; Creighton *et al.*, 2009; Lazarov *et*  
18 *al.*, 2009; Hanger *et al.*, 2015) (Fig. 15). Similar to Premier, these peridotite xenolith suites  
19 may also show strong Ti-enrichment (e.g., Rehfeldt *et al.*, 2008).

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42 A frequently cited line of evidence for Fe-Ti-enrichment of the lower Kaapvaal  
43 cratonic mantle lithosphere by plume-related mafic–ultramafic Bushveld magmas is the  
44 occurrence of so-called ‘igneous textured mantle xenoliths’, which contain biotite and  
45 ilmenite (Hoal, 2003). Our own ongoing work on this type of xenolith from the Premier  
46 kimberlite pipe shows that the textures and mineral assemblages plus compositions are similar  
47 to ultramafic lamprophyre dykes, which are common on the central Kaapvaal craton in the  
48 vicinity of the ca. 1400 Ma Pilanesberg alkaline complex (Verwoerd, 2006) (Fig. 1). Hence, it  
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3 is doubtful that this xenolith type from Premier has a direct mantle origin and we also  
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5 challenge the previously inferred connection to Bushveld magmatic activity (cf., Hoal, 2003).  
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9 Ernst *et al.* (2018) modelled the thermal impact of sizeable mantle plumes on thick  
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11 diamond-bearing lithospheres and concluded that excess heat of  $>150^{\circ}\text{C}$  above ambient  
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13 mantle temperature dissipates within approximately 100 million years, after which the pre-  
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15 plume thermal structure of the affected cratonic mantle would be restored. Hence, it is  
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17 probable that the thermal effects of the putative Bushveld mantle plume on the central  
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19 Kaapvaal cratonic root had vanished before the Mesoproterozoic. In other words, the mantle-  
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21 derived xenoliths and megacrysts brought to surface by the ca. 1150 Ma Premier kimberlite  
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23 magma eruptions, some 900 million years after the Bushveld event, are highly unlikely to  
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25 retain a record of this large-scale thermal disturbance. Some of these materials, such as  
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27 incompatible trace element enriched clinopyroxene megacrysts, may even post-date Bushveld  
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29 magmatism.  
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33 Korolev *et al.* (2018b) determined anomalously high formation and equilibration  
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35 temperatures of up to  $1500^{\circ}\text{C}$  for olivine inclusions in some lithosphere-derived peridotitic  
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37 diamonds from Premier. Likewise, some lithosphere-derived eclogitic diamonds also yielded  
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39 high formation temperatures of up to  $1570^{\circ}\text{C}$ . These high formation temperatures of some  
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41 diamonds were linked to ‘hot’ carbon-bearing fluids associated with the putative Bushveld  
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43 mantle plume (Nimis, 2002; Korolev *et al.*, 2018b). The fact that single mineral inclusions or  
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45 non-touching inclusion pairs shielded by diamond cannot re-equilibrate to the steady-state  
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47 thermal conditions of cooling mantle lithosphere suggests indeed that rare vestiges of a  
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49 Bushveld mantle plume – or of any other known regional large igneous event during the  
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51 Paleoproterozoic or Neoproterozoic (Gumsley *et al.*, 2020) – survived within the central  
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53 Kaapvaal cratonic root. Rhenium depletion model ages between 2.8–1.7 Ga for peridotite  
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3 xenoliths from Premier are in agreement with both Neoproterozoic and Paleoproterozoic  
4 overprinting of the cratonic mantle lithosphere (Pearson *et al.*, 1995). However, the thermal,  
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7 compositional and microstructural records of the sheared peridotite xenoliths and  
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10 clinopyroxene megacrysts studied relate more plausibly to dynamic processes within the  
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12 lithosphere–asthenosphere transition zone at ~150–225 km depth beneath the central  
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14 Kaapvaal craton during the Mesoproterozoic.

### 15 16 17 **Significance for kimberlite and carbonatite magma petrogenesis**

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21 The petrogenesis of the Premier kimberlite may hold important clues to the origins and  
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23 evolution of volatile-rich low-volume melts beneath cratons, because this locality is  
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25 associated with some of the largest and deepest derived ‘sublithospheric’ diamonds (Nestola  
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27 *et al.*, 2018; Smith *et al.*, 2018), and also with one of the most strongly decoupled Nd-Hf  
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29 isotope signatures identified for mantle-derived magmas (Tappe *et al.*, 2020a). Tappe *et al.*  
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31 (2020b) used the highly siderophile element systematics of fresh kimberlite and carbonatite  
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33 dykes from Premier pipe to argue that the observed Nd-Hf isotope decoupling, with initial  
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35  $\Delta\epsilon_{\text{Hf}}$  values as low as -10.7 (Fig. 16B), is probably not created during melt–rock interactions  
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37 within the peridotite-dominated mantle lithosphere. These authors suggested a primary origin  
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39 of this feature that either relates to the composition of the convecting mantle source (e.g.,  
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41 presence of ancient recycled oceanic crust components), or to the melting process within the  
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43 convecting mantle source such as mixing of melt increments derived from a highly  
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45 heterogeneous domain.  
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51 Megacryst formation in the lower cratonic lithosphere has frequently been linked to  
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53 ‘failed’ proto-kimberlite magma eruptions (Hops *et al.*, 1992; Moore & Belousova, 2005;  
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55 Giuliani *et al.*, 2013), and the partially overlapping Sr-Nd-Hf-Pb isotopic compositions  
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57 between many megacryst suites and their host kimberlites from localities worldwide support  
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3 this inference (Nowell *et al.*, 2004; Kopylova *et al.*, 2009; Tappe *et al.*, 2011; Bussweiler *et*  
4 *al.*, 2018). At Premier, the analyzed clinopyroxene megacrysts show significantly less Nd-Hf  
5 isotope decoupling than the host kimberlite, with initial  $\Delta\epsilon_{\text{Hf}}$  values of -2.6 to -6.9 *versus* -7.6  
6 to -10.7, respectively. Importantly, the carbonatite dykes have  $\Delta\epsilon_{\text{Hf}}$  values of -6.1 to -8.8 and  
7 provide a better Nd-Hf isotopic match to the megacrysts and clinopyroxene porphyroclasts of  
8 sheared peridotite xenolith CIM15-50 ( $\Delta\epsilon_{\text{Hf}}$  -4.6), although the clinopyroxene crystals extend  
9 the data array toward slightly higher  $^{176}\text{Hf}/^{177}\text{Hf}$  (Fig. 16B). Taken at face value, the Nd-Hf  
10 isotope data, together with P-T estimates and trace element compositions (Figs. 5, 9), suggest  
11 that the megacrysts formed from percolating carbonate-rich melts near the top and bottom of  
12 the lithosphere–asthenosphere transition zone beneath the central Kaapvaal craton at around  
13 1150 Ma. The upward percolating carbonate-rich melts (carbonatitic to proto-kimberlitic?)  
14 appear to have interacted with ancient cratonic mantle lithosphere to attain the slightly more  
15 radiogenic Hf isotopic compositions measured for some of the shallower clinopyroxene  
16 megacrysts from ~135–145 km depth (Fig. 17; Table 2; Supplementary Data Appendix E).  
17 The main Hf repositories within Archean cratonic mantle are clinopyroxene and garnet  
18 (Griffin *et al.*, 2000), and both peridotitic (Simon *et al.*, 2007; Aulbach *et al.*, 2013; Shu *et al.*,  
19 2019) and eclogitic (Jacob *et al.*, 2005; Aulbach *et al.*, 2007; Schmidberger *et al.*, 2007)  
20 assemblages show the high  $^{176}\text{Hf}/^{177}\text{Hf}$  required to explain the small isotopic shifts by subtle  
21 interactions between the carbonate-rich melts and depleted ancient cratonic mantle lithosphere  
22 (Fig. 16B).  
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49 Previous authors also suggested roles for carbonate-rich melts in the origin of  
50 clinopyroxene megacrysts (e.g., Cone & Kopylova, 2021), and some workers argued that such  
51 carbonated liquids evolve into carbonated silicate magmas of kimberlitic affinity by  
52 assimilation processes within the lower cratonic lithosphere (van Achtebergh *et al.*, 2002,  
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2004; Bussweiler *et al.*, 2016; Bussweiler *et al.*, 2018). In the case of Premier, however, such melt evolution from carbonatitic to kimberlitic would require assimilation of significant amounts of refractory peridotite material that has very low  $^{176}\text{Hf}/^{177}\text{Hf}$ , as well as  $\epsilon\text{Nd}-\epsilon\text{Hf}$  systematics below the mantle array (Fig. 16B). Cratonic peridotites with  $\epsilon\text{Nd}-\epsilon\text{Hf}$  values below the mantle array are volumetrically very minor, and the continental lithospheric mantle reservoir is characterized by strongly depleted Nd-Hf isotopic compositions above the mantle array (Pearson & Wittig, 2014). Hence, we argue that the Premier kimberlite magmas originated as carbonated silicate melts from the  $>1300^\circ\text{C}$  warm convecting mantle beneath the  $>200$  km thick continental root, a notion that finds support in the compositions of silicate glass-rich melt inclusions within kimberlite-borne olivine megacrysts derived from the base of the Kaapvaal craton farther south (Howarth & Büttner, 2019). In contrast, the thermal regime of the cooler lithosphere–asthenosphere transition zone between  $\sim 150$ – $200$  km depth additionally can stabilize relatively ‘pure’ carbonate melts in the presence of CHO fluid phases (Foley *et al.*, 2009; Massuyeau *et al.*, 2021), but this process requires – at least temporarily – fairly oxidizing mantle redox conditions (Stagno & Frost, 2010; Stagno *et al.*, 2013). Such carbonate-rich melts and associated high-density fluids are extremely mobile and present potent metasomatic agents upon interactions with the lower cratonic mantle lithosphere (Rudnick *et al.*, 1993; Weiss *et al.*, 2011; Kopylova *et al.*, 2021). In conjunction with local melt ponding at permeability boundaries (Rabinowicz *et al.*, 2002), and also during strong shear movements (Katz *et al.*, 2006; Holtzman & Kohlstedt, 2007; Le Roux *et al.*, 2008) (Supplementary Data Figure 1), megacrysts may form from such low-volume carbonate-rich melts by deformation-assisted segregation and channeling within the lower reaches of cratonic lithosphere.

Ubiquitous volatile-rich melts and high-density fluids also contribute to the accommodation of extremely high shear stresses across the lithosphere–asthenosphere

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3 transition zone caused by plate tectonic motions (Tharimena *et al.*, 2017). Hence, low-degree  
4 carbonate-rich melts within the lowermost lithosphere may – contrary to previous models  
5 (e.g., Tappe *et al.*, 2007; Foley, 2011; Holtzman *et al.*, 2012) – lend overall stability to  
6 cratonic mantle keels while they are dragged over the convecting mantle for >2.5 billion years  
7 as part of drifting tectonic plates. In other words, the fabrics of and metasomatic imprints on  
8 the sheared peridotite xenoliths and clinopyroxene megacrysts entrained by the Premier  
9 kimberlite eruptions at ca. 1150 Ma may present snapshots of lithosphere–asthenosphere  
10 interactions at the early phase of the 1220–1090 Ma Namaqua–Natal orogeny, during which  
11 the Kaapvaal craton was on collision course with several adjacent more juvenile terranes  
12 (Jacobs *et al.*, 2008; Spencer *et al.*, 2015; Van Schijndel *et al.*, 2020) (Fig. 1). These  
13 deformation events may have also facilitated the creation of new, or the reactivation of  
14 existing, translithospheric magma ascent pathways through the central Kaapvaal craton (see  
15 Holtzman *et al.*, 2012), along which both kimberlitic and carbonatitic melts were successfully  
16 extracted from the lithosphere–asthenosphere boundary over the course of several millions of  
17 years (Tappe *et al.*, 2018a; Tappe *et al.*, 2018b).

## 37 CONCLUSIONS

- This study shows that strongly deformed and refertilized sheared peridotites occur  
42 between ~160–185 km depth beneath the central Kaapvaal craton, falling firmly within  
43 a lithosphere–asthenosphere transition zone that reaches from ~150 to ~225 km depth.  
44 This  $\geq 50$  km thick boundary layer is defined here by intersections of the ~40–42 mW/m<sup>2</sup>  
45 Premier paleogeotherm with (1) CO<sub>2</sub>-H<sub>2</sub>O-present solidus curves for peridotite (upper  
46 bound), and (2) typical mantle adiabats with potential temperatures between 1315 and  
47 1420°C (lower bound).

- At 150–225 km depth, the central Kaapvaal cratonic mantle root should be able to stabilize small volumes of ephemeral carbonate-rich melts and associated high-density fluids, which may be a critical factor in causing diffuse, several 10s of kilometers thick lithosphere–asthenosphere ‘transitions’ beneath cratons. Permeability contrasts at the top and bottom of the lithosphere–asthenosphere transition zone may cause local ponding of carbonate-rich melts, which can trigger the formation of megacryst suites.
- We suggest that sheared peridotite formation occurs in localized deformation zones spaced out across the entire width of the lithosphere–asthenosphere transition zone between ~150–225 km depth, rather than being restricted to a single thin layer at the craton base where mantle flow causes viscous drag. Thus, plate tectonic stresses acting on the lower cratonic lithosphere may be accommodated by networks of narrow shear zones, which – when active – provide pathways and sinks for percolating volatile-rich melts, linking the formation of megacrysts and sheared peridotites (i.e., deformation-assisted melt segregation).
- The central Kaapvaal cratonic root has frequently been portrayed as thermally and compositionally ‘anomalous’, and a common explanation is heavy overprinting of the mantle lithosphere during magmatic activity associated with the giant Bushveld layered intrusion at ca. 2056 Ma. However, the vast majority of thermal, compositional and microstructural properties of the lower cratonic mantle beneath the ca. 1150 Ma Premier pipe can be linked to dynamic processes across the  $\geq 50$  km thick lithosphere–asthenosphere transition zone, shortly prior to or during Mesoproterozoic kimberlite and carbonatite magmatism. Thus, it is unlikely that after 900 million years the impact of a putative Bushveld mantle plume on the Kaapvaal cratonic root would



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3 still be evident, with the possible exception of ‘remnant’ high formation temperatures  
4 of some lithospheric diamonds that retain a memory of long vanished heat anomalies.  
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9 • The Nd-Hf isotopic compositions of the clinopyroxene megacrysts, falling largely  
10 within the mantle array, are best explained by an origin from percolating carbonate-  
11 rich melts that underwent subtle interactions with ancient depleted cratonic mantle  
12 components on their way through the lowermost lithosphere. On the basis of this Nd-  
13 Hf isotope evidence, these particular melt–rock interactions cannot transform the  
14 observed carbonate-rich liquid compositions into kimberlitic magmas. Thus, the  
15 strongly decoupled Nd-Hf isotope systematics of the Premier kimberlite, falling below  
16 the mantle array, are consistent with an ultimate magma origin as carbonated silicate  
17 melts from deep within the convecting upper mantle, fully compatible with the  
18 presence of ‘ultradeep’ sublithospheric diamonds.  
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## 31 32 **FUNDING**

33  
34  
35 This work was supported by the National Research Foundation (NRF) of South Africa (IPRR  
36 grant to ST) and the Geological Society of South Africa (REI grant to ST). The DEEP  
37 Research Group at the University of Johannesburg is financially supported by the DSI-NRF  
38 CIMERA Centre of Excellence, South Africa. Calcium isotope work in Münster was  
39 supported through grant GU1035/10-1 by the German Research Foundation (DFG).  
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## 48 49 **ACKNOWLEDGMENTS**

50  
51 We gratefully acknowledge support of our research on Cullinan Diamond Mine by Petra  
52 Diamonds Ltd and the Geological Society of South Africa. Christian Reinke and Bradley Guy  
53 are thanked for valuable help with the EPMA and MLA analyses in Johannesburg. This study  
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2  
3 has benefited from informal discussions with Fanus Viljoen about the Premier mantle and  
4  
5 Jeremie Lehmann about shear zones. Yannick Bussweiler is sincerely thanked for sharing his  
6  
7 megacryst database and for discussions pertaining to this topic. The highly constructive  
8  
9 criticism and valuable scientific feedback by Maya Kopylova, Alex McCoy-West, Pedro  
10  
11 Waterton and an anonymous reviewer are much appreciated. We thank the science editors  
12  
13  
14 Tod Waight and Georg Zellmer for consummate handling of the review process.  
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## 17 18 **DATA AVAILABILITY STATEMENT**

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21 The data underlying this article are available in the article and in its online supplementary  
22  
23 material.  
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## 26 27 **SUPPLEMENTARY DATA**

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30 Supplementary data are available at *Journal of Petrology* online.  
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## 33 34 **FIGURE CAPTIONS**

### 35 36 37 **Figure 1:**

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40 Geological map of southern Africa showing the outline of the Archean Kaapvaal craton and  
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42 surrounding Proterozoic terranes plus mobile belts (adopted from Tappe *et al.*, 2020b). The  
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44 1153.3 ± 5.3 Ma Premier kimberlite pipe (Cullinan Diamond Mine) is located in the Republic  
45  
46 of South Africa on the central Kaapvaal craton at the southern margin of the ca. 2056 Ma  
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48 Bushveld Complex. The ca. 1400 Ma Pilanesberg alkaline complex (PAC) that intrudes the  
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50 western lobe of the Bushveld Complex, as well as several renowned Group-1 kimberlite  
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52 localities on and around the Kaapvaal craton, ranging in age between 1835 and 90 Ma, are  
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54 shown for reference.  
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**Figure 2:**

Photographs of representative sheared peridotite xenoliths (**A-C**) and coarse peridotite 'nodule' CIM15-40 (**D**) from the ca. 1150 Ma old Premier kimberlite pipe, Cullinan Diamond Mine, on the central Kaapvaal craton. **Panel A** reveals the contact between sheared peridotite xenolith CIM15-8 and 'Grey' volcanoclastic kimberlite that fills large volumes of the ~32 hectares large Premier diatreme structure (Tappe *et al.*, 2018a). The scale bar in **Panel A** applies to all panels. Ol – olivine, Grt – garnet, Opx – orthopyroxene, Cpx – clinopyroxene.

**Figure 3:**

(**A**) Back-scattered electron image map of sheared peridotite xenolith CIM15-24 from the Premier kimberlite pipe at the scale of an entire standard petrographic thin section. The BSE map was created with a FEI Quanta 600F MLA system that is equipped with two Bruker 4010 Xflash X-ray detectors. The laminated and disrupted peridotite fabric with isolated garnet–orthopyroxene porphyroclasts set in a matrix of olivine neoblasts can be identified. Note at the image centre the coarser olivine porphyroclasts that underwent grain-size-reduction and neoblast formation along their margins. (**B-C**) Photomicrographs of sheared peridotite xenoliths from Premier pipe under plane-polarized light. Note the strong ductile deformation textures with development of recrystallization tails that can be used as kinematic shear-sense indicators. Note further touching relationships between coexisting garnet and orthopyroxene grains enabling pressure estimations based on Al-exchange. The scale bar in **Panel B** also applies to **Panel C**. Ol – olivine, Grt – garnet, Opx – orthopyroxene, Cpx – clinopyroxene, alt – alteration (e.g., kelyphite).

**Figure 4:**

Back-scattered electron images (A-E) of sheared peridotite xenoliths recovered from the Premier kimberlite pipe, Cullinan Diamond Mine, in South Africa. (A) Stretched clinopyroxene porphyroclast with a spongy-textured halo and fine-grained recrystallization tail engulfed by a matrix that is dominated by olivine neoblasts. (B) Touching pair of garnet and orthopyroxene as part of a porphyroclast set within a fine-grained matrix that consists mainly of neoblastic olivine including undeformed euhedral ‘tablets’. (C) Laminated–disrupted porphyroclastic texture of sheared peridotite xenolith ST16-CUL26 showing a tightly folded clinopyroxene crystal at the image centre. The image is a composite comprising four individual BSE images taken at the smallest possible magnification. (D) Touching pair of garnet and orthopyroxene as part of a porphyroclast set within a fine-grained olivine matrix. Note the significant grain-size-reduction and neoblast formation that affected the larger olivine crystal enclosed by porphyroclastic orthopyroxene. (E) Relatively small orthopyroxene porphyroclast with a diffuse melting-induced clinopyroxene halo ‘floating’ within a neoblastic olivine matrix. Ol – olivine, Grt – garnet, Opx – orthopyroxene, Cpx – clinopyroxene.

**Figure 5:**

Pressure–temperature systematics of peridotite xenoliths and Cr-poor clinopyroxene megacrysts from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Table 1 and Supplementary Data Appendix E). The ‘Premier’ paleogeotherm (thick solid red curve) was obtained by regression of P-T data for the here investigated and previously reported coarse and sheared / ‘deformed’ peridotite xenoliths using *FITPLOT*. The long-dashed red curve represents geotherm fitting that included only coarse peridotite xenoliths. All xenolith data, including those from Viljoen *et al.* (2009), are calculated or recalculated

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3 using the preferred *TNG* thermobarometer combination (Nickel & Green, 1985; Taylor,  
4 1998). The P-T data for discrete clinopyroxene megacrysts were calculated according to  
5 Nimis & Taylor (2000), but are not included into the displayed geotherm fitting (outlier  
6 CUL17-cpx4 is discussed in the main text). The brown field for rare ‘cold’ clinopyroxene  
7 xenocrysts and inclusions in some Premier diamonds is based on the data reported in Nimis *et*  
8 *al.* (2020) (the complete clinopyroxene P-T dataset is displayed in Supplementary Data Figure  
9 2). The graphite/diamond phase transition curve of Day (2012) and conductive model  
10 geotherms from Hasterok & Chapman (2011) are shown for reference. The CO<sub>2</sub>-H<sub>2</sub>O-present  
11 peridotite solidus curve is adopted from Foley *et al.* (2009) and has been extrapolated beyond  
12 6 GPa applying the data compilations for high-P melting experiments in Tappe *et al.* (2018b)  
13 and Massuyeau *et al.* (2021). Adiabats for ambient mantle potential temperatures of 1315,  
14 1360 and 1420°C are shown together with corresponding estimations of the petrological  
15 lithosphere–asthenosphere boundary (LAB) beneath the central Kaapvaal craton at ca. 1150  
16 Ma. The blue vertical bar indicates the depth location of a lithosphere–asthenosphere  
17 transition zone (LAT), in which low-degree carbonate-rich melts may be ubiquitous (see main  
18 text).

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39 **Figure 6:**

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42 Average forsterite contents *versus* corresponding olivine modal abundances of sheared and  
43 coarse garnet-facies peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond  
44 Mine (all data are listed in Supplementary Data Appendix D, E). For the sheared peridotite  
45 xenoliths from Premier, both porphyroclastic and neoblastic ‘matrix’ olivine compositions are  
46 shown, without discernable differences in their Mg-numbers within a single nodule. The field  
47 for olivine inclusions within Premier diamonds is based on data presented in Viljoen *et al.*  
48 (2014) and Korolev *et al.* (2018b), with the mean forsterite content shown as an ‘X’. Fields  
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3 for low- and high-temperature peridotite xenoliths from the Kaapvaal and Zimbabwe cratons  
4 are adopted from van der Meer *et al.* (2013). Averages for low-temperature peridotites from  
5 the Kaapvaal craton (*golden star 1*) and other major cratons worldwide (*red star 2*), as well as  
6 for modern abyssal peridotites (*green star 3*), are taken from Pearson & Wittig (2014).  
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8 Olivine in sheared peridotites from Premier pipe and other kimberlite occurrences on the  
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10 Kaapvaal craton resembles olivine from abyssal peridotites of the modern ocean basins, as is  
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12 also apparent from the proximity to the ‘oceanic melting trend’ of Boyd (1989).  
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20 **Figure 7:**

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23 Chondrite normalized REE distributions for orthopyroxene in sheared and coarse peridotite  
24 xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in  
25 Supplementary Data Appendix D). The REE patterns for orthopyroxene in garnet-facies  
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27 peridotite xenoliths derived from ca. 90 Ma old kimberlite pipes of Lesotho and Kimberley on  
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29 the Kaapvaal craton (*grey squares*) are shown for reference (Simon *et al.*, 2003, 2007).  
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31 Chondrite values for normalization are from Palme & Jones (2003).  
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38 **Figure 8:**

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40 TiO<sub>2</sub> (**A**) and Cr<sub>2</sub>O<sub>3</sub> (**B**) contents *versus* Mg-number for clinopyroxene in sheared and coarse  
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42 peridotite xenoliths, as well as Cr-poor clinopyroxene megacrysts, from the Premier  
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44 kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix  
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46 D). Premier pipe literature data for clinopyroxene from sheared (*filled black circles*) and  
47  
48 coarse (*open black circles*) peridotite xenoliths are taken from Viljoen *et al.* (2009). The data  
49  
50 compilation for clinopyroxene megacrysts from kimberlite localities worldwide is based on  
51  
52 Bussweiler *et al.* (2018). The field for clinopyroxene compositions in metasomatized  
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54 peridotite xenoliths from cratons worldwide is based on data provided by Kopylova *et al.*  
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3 (1999), Simon *et al.* (2003, 2007), Aulbach *et al.* (2007), Rehfeldt *et al.* (2008), Kargin *et al.*  
4  
5 (2017), and Busweiler *et al.* (2018).  
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8 **Figure 9:**  
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11 Chondrite normalized REE distributions for clinopyroxene in sheared and coarse peridotite  
12 xenoliths (**A**) and for Cr-poor clinopyroxene megacrysts (**B**) from the Premier kimberlite  
13 pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D). (**C**)  
14 Reconstructed equilibrium melt compositions that may have been responsible for the  
15 metasomatic nature of clinopyroxene in garnet-facies peridotite xenoliths from the Premier  
16 kimberlite pipe, and may have also caused clinopyroxene megacryst formation. Both the  
17 effects of ‘carbonate-dominated’ (cpx/melt partition coefficients from Dasgupta *et al.*, 2009  
18 and Girmis *et al.*, 2013) and ‘carbonated silicate’ (Keshav *et al.*, 2005) melts at P-T conditions  
19 equivalent to the lithosphere–asthenosphere boundary beneath the central Kaapvaal craton are  
20 shown. Average kimberlite (red line) and carbonatite (blue line) dyke compositions from  
21 Premier pipe are based on the data presented in Tappe *et al.* (2020b). The extremely high  
22 LREE–MREE concentrations of saline and high-Mg carbonatitic high-density fluids (HDFs)  
23 entrapped in diamonds from lithospheric mantle columns of cratons worldwide (Weiss *et al.*,  
24 2011, 2018) suggest their possible involvement in the origin of enriched clinopyroxene  
25 signatures within the Kaapvaal continental root. The REE patterns for clinopyroxene from  
26 garnet-facies peridotite xenoliths of the Kimberley kimberlite cluster on the western Kaapvaal  
27 craton (*grey lines*) are shown for reference in **Panels A-B-C** (Simon *et al.*, 2007). Chondrite  
28 values for normalization are from Palme & Jones (2003).  
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**Figure 10:**

Cr<sub>2</sub>O<sub>3</sub> versus CaO contents (wt.%) of garnet in sheared and coarse peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D). The garnet ‘G-type’ classification and the ‘graphite–diamond constraint’ are according to Grütter *et al.* (2004). Literature data for garnet from sheared (*filled black circles*) and coarse (*open black circles*) peridotite xenoliths, as well as from diamond inclusions (*filled grey diamond symbol*) are taken from Viljoen *et al.* (2009) and Viljoen *et al.* (2014), respectively. Note the partial overlap between G9 lherzolitic garnet grains from Premier pipe and the compositional field of G1 low-Cr garnet megacrysts.

**Figure 11:**

TiO<sub>2</sub> content (**A**) and Cr-number (**B**) versus Mg-number for garnet in sheared and coarse peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D). Premier pipe literature data for garnet from sheared (*filled black circles*) and coarse (*open black circles*) peridotite xenoliths, as well as from diamond inclusions (*filled grey diamond symbol*) are taken from Viljoen *et al.* (2009) and Viljoen *et al.* (2014), respectively. The data compilation for garnet megacrysts from localities worldwide is based on Bussweiler *et al.* (2018), and dominated by southern African occurrences for the low-Cr suite (*filled dark grey circles*), as opposed to occurrences located mainly on the Slave craton in Canada for the high-Cr megacrysts (*filled light grey circles*). The field for garnet compositions from metasomatized peridotite xenoliths of the Kaapvaal craton is based on data compiled in Bussweiler *et al.* (2018).



**Figure 12:**

(A) Chondrite normalized REE distributions for garnet in sheared and coarse peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D). Premier pipe literature data for garnet from sheared / deformed and coarse peridotite xenoliths (*grey lines*) are taken from Viljoen *et al.* (2009) and display both ‘normal’ and ‘sinusoidal’ patterns. (B) Reconstructed equilibrium melt compositions that may have been responsible for the metasomatic nature of garnet in peridotite xenoliths from the Premier kimberlite pipe. Both the effects of ‘carbonate-dominated’ (garnet/melt partition coefficients from Dasgupta *et al.*, 2009 and Giris *et al.*, 2013) and ‘carbonated silicate’ (Brey *et al.*, 2008) melts at P-T conditions equivalent to the lithosphere–asthenosphere boundary beneath the central Kaapvaal craton are shown. Data for fresh kimberlite (red lines) and carbonatite (blue lines) dykes from Premier pipe are from Tappe *et al.* (2020b). Chondrite values for normalization are from Palme & Jones (2003).

**Figure 13:**

(A) Y versus Zr contents, and (B) Zr/Hf versus Ti/Eu ratios of garnet in sheared and coarse peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D). Premier pipe literature data for garnet from sheared (*filled black circles*) and coarse (*open black circles*) peridotite xenoliths, as well as from diamond inclusions (*filled grey diamond symbol*) are taken from Viljoen *et al.* (2009) and Viljoen *et al.* (2014), respectively. Data for fresh kimberlite (*filled grey triangle*) and carbonatite (*filled grey inverted triangle*) dykes from Premier pipe are from Tappe *et al.* (2020b). The layout of **Panel A** is adopted from Griffin & Ryan (1995), with some modification to the metasomatic trend. The layout of **Panel B** is adopted from Shu & Brey (2015), with modifications of the metasomatic trends. Note that the Ti/Eu values have been

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3 divided by a hundred for simplicity's sake. Primitive mantle composition is from Palme &  
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5 O'Neill (2003).  
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9 **Figure 14:**

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11 REE distribution coefficients (chondrite-normalized, Palme & Jones, 2003) for coexisting  
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13 garnet and clinopyroxene in sheared and coarse peridotite xenoliths from the Premier  
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15 kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix  
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17 D). A range of equilibrium  $Gr/Cpx$  D values determined either experimentally (Green *et al.*,  
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19 2000; Adam & Green, 2006; Xiong, 2006; Giris *et al.*, 2013) or on natural rocks (Zack *et al.*,  
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21 2000; Adam & Green, 2006; Xiong, 2006; Giris *et al.*, 2013) or on natural rocks (Zack *et al.*,  
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23 1997) is shown for comparison, indicating trace element equilibrium to subtle disequilibrium  
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25 for the Premier sheared peridotite xenoliths, and notable LREE disequilibrium for coarse  
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27 peridotite sample CIM15-040. For this sample, clinopyroxene is more LREE enriched than  
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29 predicted by the composition of coexisting garnet.  
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33 **Figure 15:**

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35 The pressure *versus*  $\Delta \log fO_2$  systematics (relative to the FMQ buffer) of sheared garnet  
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37 peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are  
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39 listed in Table 1 and Supplementary Data Appendix E). The error bars entail the 2S.E.  
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41 uncertainties of the  $fO_2$  calculations using the formulation by Stagno *et al.* (2013) and an  
42  
43 estimated uncertainty of 0.2 GPa for pressure calculations using the calibration by Nickel &  
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45 Green (1985). The previously published data for garnet peridotite xenoliths from the  
46  
47 Kaapvaal (Woodland & Koch, 2003; Creighton *et al.*, 2009; Lazarov *et al.*, 2009; Hanger *et*  
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49 *al.*, 2015), Slave (Creighton *et al.*, 2010; Yaxley *et al.*, 2017) and Siberian (Goncharov *et al.*,  
50  
51 2012; Yaxley *et al.*, 2012) cratons are recalculated using the Stagno *et al.* (2013) and Nickel  
52  
53 & Green (1985) methods for  $fO_2$  and pressure estimations, respectively (Supplementary Data  
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3 Appendix B). The graphite/diamond phase transition and graphite/diamond *versus* carbonate  
4 stability reaction in a peridotitic system [EMOG/D] at cratonic thermal conditions are adopted  
5 from Yaxley *et al.* (2017).  
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### 10 **Figure 16:**

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14 Initial  $\epsilon\text{Nd}$  *versus*  $^{87}\text{Sr}/^{86}\text{Sr}$  (**A**) and  $\epsilon\text{Hf}$  *versus*  $\epsilon\text{Nd}$  (**B**) for clinopyroxene from sheared and  
15 coarse peridotite xenoliths, as well as for discrete Cr-poor clinopyroxene megacrysts from the  
16 Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Table 2 and  
17 Supplementary Data Appendix E). Published data for Cr-poor megacrysts (cpx –  
18 clinopyroxene, grt – garnet, ilm – ilmenite) from Premier pipe are taken from Nowell *et al.*  
19 (2004). The data for Premier kimberlite and carbonatite dykes are from Tappe *et al.* (2020b).  
20  
21 In **Panel A**, the Sr-Nd isotope data for clinopyroxene and garnet inclusions within peridotitic  
22 diamonds from Premier pipe (Richardson *et al.*, 1993), the ca. 1400 Ma Pilanesberg alkaline  
23 complex (Cawthorn *et al.*, 2012), and basaltic lavas from the Rooiberg Group of the ca. 2056  
24 Ma Bushveld Complex on the central Kaapvaal craton (Günther *et al.*, 2018) are shown for  
25 comparison. All data in **Panel A** and **B** are corrected to the kimberlite magma emplacement  
26 age of ca. 1150 Ma for Premier pipe (Tappe *et al.*, 2018a) and they entail a full propagation of  
27 uncertainties shown by the error bars (see Methods and the footnote to Table 2). In **Panel B**,  
28 the ‘mantle array’ is after Chauvel *et al.* (2008) and the vectors pointing toward depleted  
29 ancient cratonic mantle peridotites are after Tappe *et al.* (2020b).  
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### 48 **Figure 17:**

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51 Initial  $^{87}\text{Sr}/^{86}\text{Sr}$  (**A**),  $\delta^{44/40}\text{Ca}$  (**B**), initial  $^{176}\text{Hf}/^{177}\text{Hf}$  (**C**), and initial  $\Delta\epsilon\text{Hf}$  (**D**) compositions  
52 *versus* the equilibration depths of clinopyroxene porphyroclasts from sheared peridotite  
53 xenoliths and of discrete clinopyroxene megacrysts from the Premier kimberlite pipe,  
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Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix E). No hafnium isotope data are available for megacryst CUL17-cpx4 due to its very low Hf concentration.

The upper mantle  $\delta^{44/40}\text{Ca}$  composition is after Chen *et al.* (2019).

### Figure 18:

The  $\delta^{44/40}\text{Ca}$  values for clinopyroxene from sheared and coarse peridotite xenoliths, as well as for discrete Cr-poor clinopyroxene megacrysts from the Premier kimberlite pipe, Cullinan Diamond Mine, reported relative to SRM-915a (all data are listed in Table 2 and Supplementary Data Appendix E). The Ca isotopic compositions of four kimberlite dykes from Premier pipe have also been analysed for comparative purposes. Published  $\delta^{44/40}\text{Ca}$  data for relevant rock types and geochemical reservoirs are taken from the following literature sources and compilations (averages  $\pm$  2S.D.): continental peridotite xenoliths (Kang *et al.*, 2017), cratonic eclogites/pyroxenites (Smart *et al.*, 2021), modern oceanic basalts (Schiller *et al.*, 2016; Chen *et al.*, 2019), modern to 3 Ga old Ca-Mg-Fe carbonatites (Amsellem *et al.*, 2020; Sun *et al.*, 2021) and 1-3 Ga old Precambrian marine carbonates (Blättler & Higgins, 2017). The average  $\delta^{44/40}\text{Ca}$  value for the upper mantle of  $0.94 \pm 0.1\%$  (dashed vertical line plus error envelop) is based on Chen *et al.* (2019).

### Figure 19:

Schematic model of the central Kaapvaal cratonic mantle lithosphere during the ca. 1150 Ma kimberlite and carbonatite magma eruptions at Premier/Cullinan. This study proposes a  $\geq 50$  km thick lithosphere–asthenosphere transition zone between  $\sim 150$ –225 km depth, where strong plastic deformation creates a network of narrow subhorizontal shear zones as represented by abundant sheared peridotite xenoliths. Stress-driven segregation of percolating carbonate-rich melts may provide a link between the common association of megacrysts and

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2  
3 sheared peridotites as part of kimberlite mantle cargo. Both lithospheric and sublithospheric  
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5 (i.e., ‘ultradeep’) diamonds are shown for reference, but they are not discussed in detail as  
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7 part of this model. CLM – cratonic lithospheric mantle, LAB – lithosphere–asthenosphere  
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9 boundary.  
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## 13 **SUPPLEMENTARY DATA FILES**

### 14 15 16 17 **Supplementary Data Appendix A**

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19 LA-ICP-MS trace element results for in-house garnet standard GHR-1 and clinopyroxene  
20  
21 standard KBY17-cpx4.  
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### 23 **Supplementary Data Appendix B**

24  
25 Recalculated oxygen fugacity data for garnet peridotite xenoliths from literature sources as  
26  
27 used in this study for comparison (see Figure 15).  
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### 29 **Supplementary Data Appendix C**

30  
31 Comparison of pressure–temperature results obtained with different thermobarometer  
32  
33 combinations.  
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### 35 **Supplementary Data Appendix D**

36  
37 Complete major and trace element dataset for minerals analysed in-situ during this study.  
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### 40 **Supplementary Data Appendix E**

41  
42 Combined petrology and Sr-Nd-Hf-Ca isotope dataset for minerals analysed during this study  
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44 (Table 1 & 2 of the main article).  
45

### 46 **Supplementary Data Figure 1**

47  
48 Clinopyroxene megacryst formation (melt pool) within sheared peridotite xenolith CIM15-50.  
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### 51 **Supplementary Data Figure 2**

52  
53 *FITPLOT* P-T data arrays for mantle-derived materials from the Premier kimberlite (this  
54  
55 study and literature sources).  
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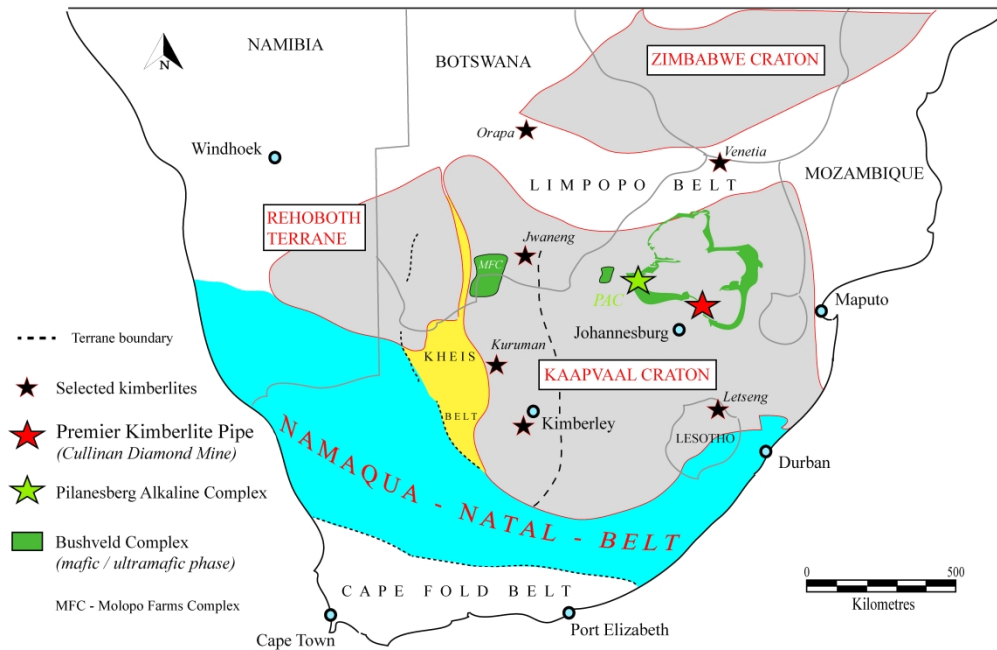


Figure 1:  
 Geological map of southern Africa showing the outline of the Archean Kaapvaal craton and surrounding Proterozoic terranes plus mobile belts (adopted from Tappe et al., 2020b). The  $1153.3 \pm 5.3$  Ma Premier kimberlite pipe (Cullinan Diamond Mine) is located in the Republic of South Africa on the central Kaapvaal craton at the southern margin of the ca. 2056 Ma Bushveld Complex. The ca. 1400 Ma Pilanesberg alkaline complex (PAC) that intrudes the western lobe of the Bushveld Complex, as well as several renowned Group-1 kimberlite localities on and around the Kaapvaal craton, ranging in age between 1835 and 90 Ma, are shown for reference.

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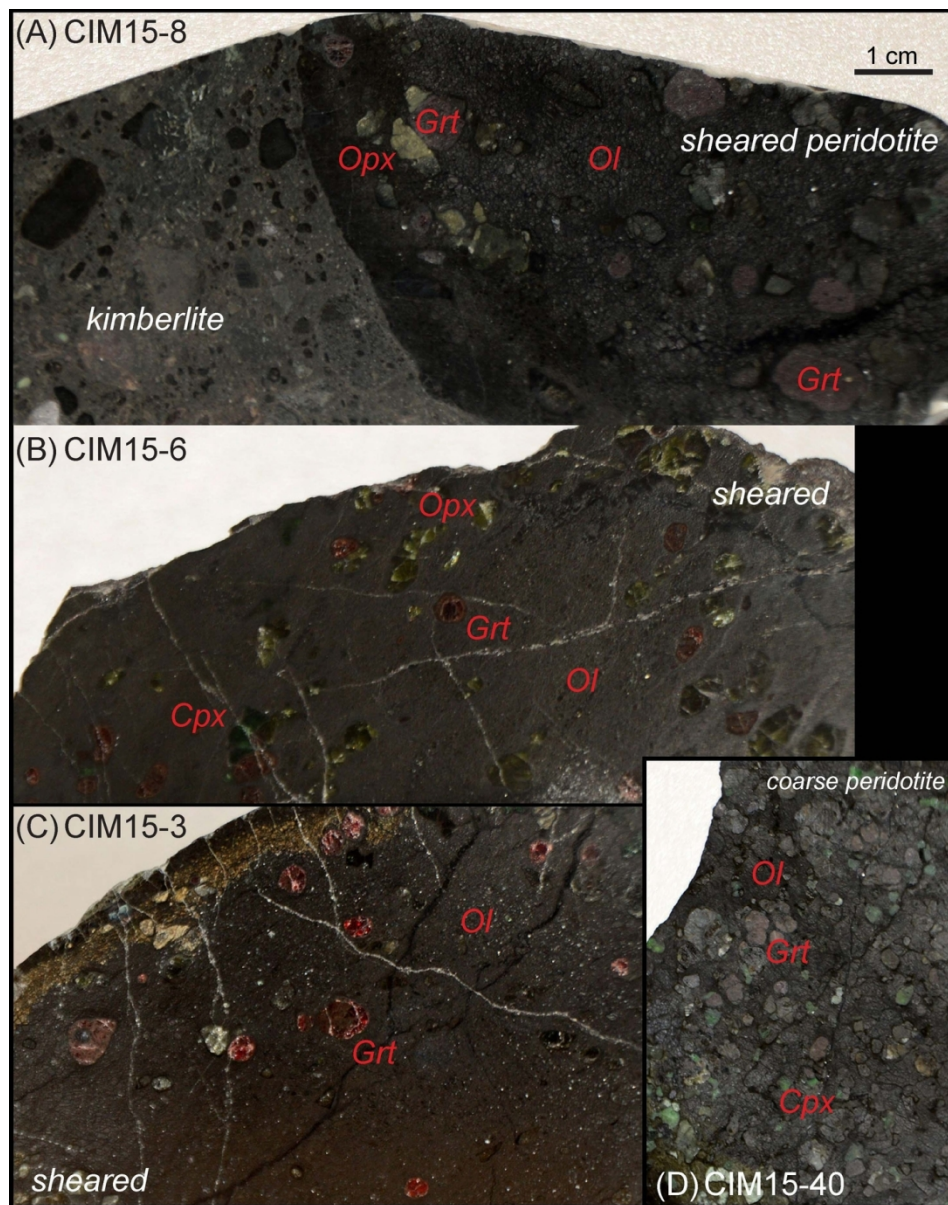


Figure 2:

Photographs of representative sheared peridotite xenoliths (A-C) and coarse peridotite 'nodule' CIM15-40 (D) from the ca. 1150 Ma old Premier kimberlite pipe, Cullinan Diamond Mine, on the central Kaapvaal craton. Panel A reveals the contact between sheared peridotite xenolith CIM15-8 and 'Grey' volcanoclastic kimberlite that fills large volumes of the ~32 hectares large Premier diatreme structure (Tappe et al., 2018a). The scale bar in Panel A applies to all panels. Ol – olivine, Grt – garnet, Opx – orthopyroxene, Cpx – clinopyroxene.

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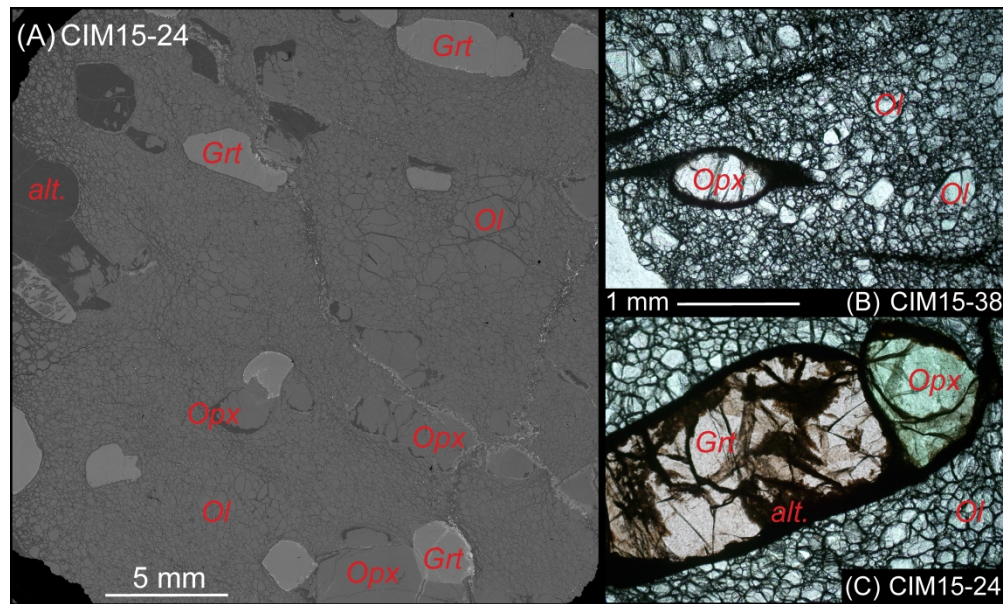


Figure 3:

(A) Back-scattered electron image map of sheared peridotite xenolith CIM15-24 from the Premier kimberlite pipe at the scale of an entire standard petrographic thin section. The BSE map was created with a FEI Quanta 600F MLA system that is equipped with two Bruker 4010 Xflash X-ray detectors. The laminated and disrupted peridotite fabric with isolated garnet–orthopyroxene porphyroclasts set in a matrix of olivine neoblasts can be identified. Note at the image centre the coarser olivine porphyroclasts that underwent grain-size-reduction and neoblast formation along their margins. (B-C) Photomicrographs of sheared peridotite xenoliths from Premier pipe under plane-polarized light. Note the strong ductile deformation textures with development of recrystallization tails that can be used as kinematic shear-sense indicators. Note further touching relationships between coexisting garnet and orthopyroxene grains enabling pressure estimations based on Al-exchange. The scale bar in Panel B also applies to Panel C. Ol – olivine, Grt – garnet, Opx – orthopyroxene, Cpx – clinopyroxene, alt – alteration (e.g., kelyphite).

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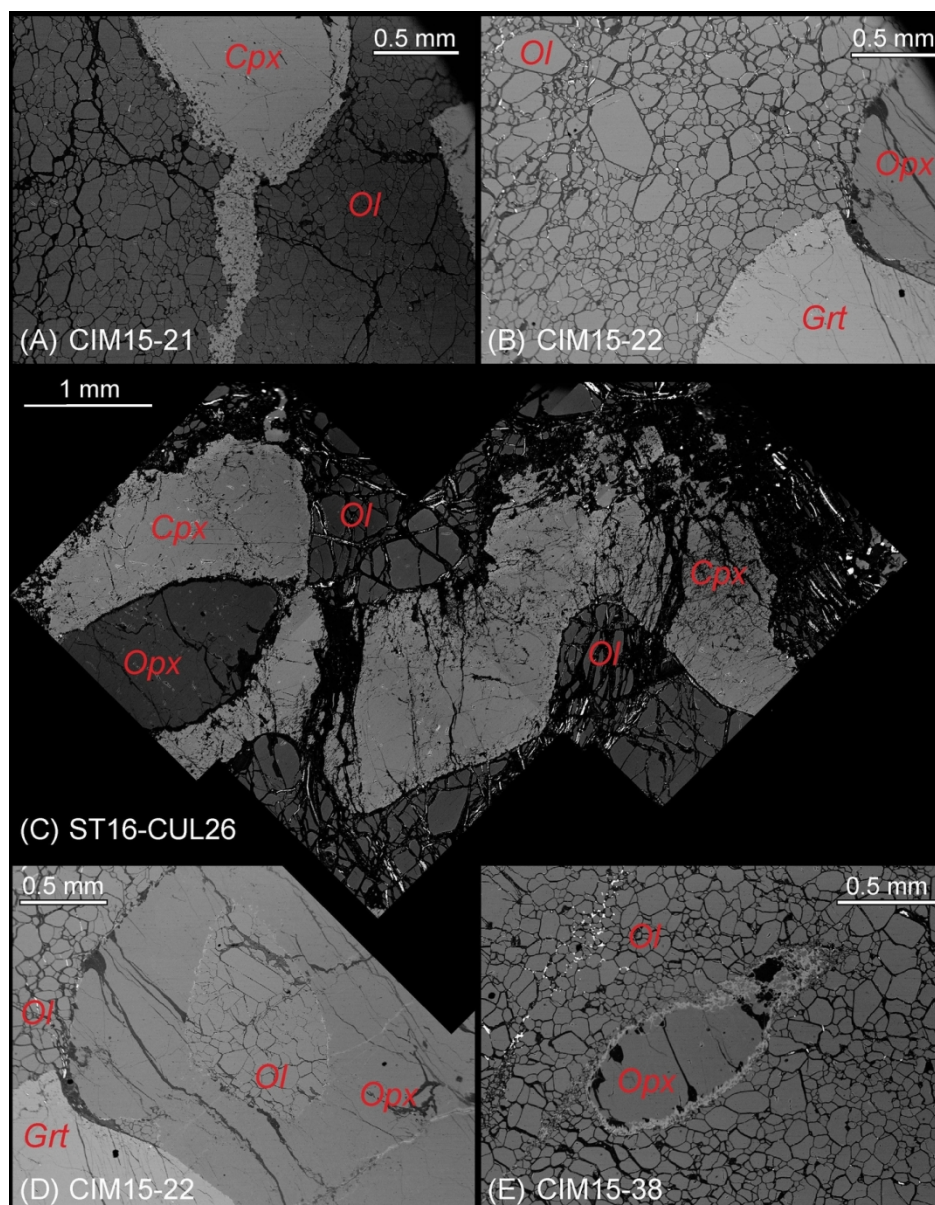


Figure 4:

Back-scattered electron images (A-E) of sheared peridotite xenoliths recovered from the Premier kimberlite pipe, Cullinan Diamond Mine, in South Africa. (A) Stretched clinopyroxene porphyroclast with a spongy-textured halo and fine-grained recrystallization tail engulfed by a matrix that is dominated by olivine neoblasts. (B) Touching pair of garnet and orthopyroxene as part of a porphyroclast set within a fine-grained matrix that consists mainly of neoblastic olivine including undeformed euhedral 'tablets'. (C) Laminated-disrupted porphyroclastic texture of sheared peridotite xenolith ST16-CUL26 showing a tightly folded clinopyroxene crystal at the image centre. The image is a composite comprising four individual BSE images taken at the smallest possible magnification. (D) Touching pair of garnet and orthopyroxene as part of a porphyroclast set within a fine-grained olivine matrix. Note the significant grain-size-reduction and neoblast formation that affected the larger olivine crystal enclosed by porphyroclastic orthopyroxene. (E) Relatively small orthopyroxene porphyroclast with a diffuse melting-induced clinopyroxene halo 'floating' within a neoblastic olivine matrix. Ol - olivine, Grt - garnet, Opx - orthopyroxene, Cpx - clinopyroxene.

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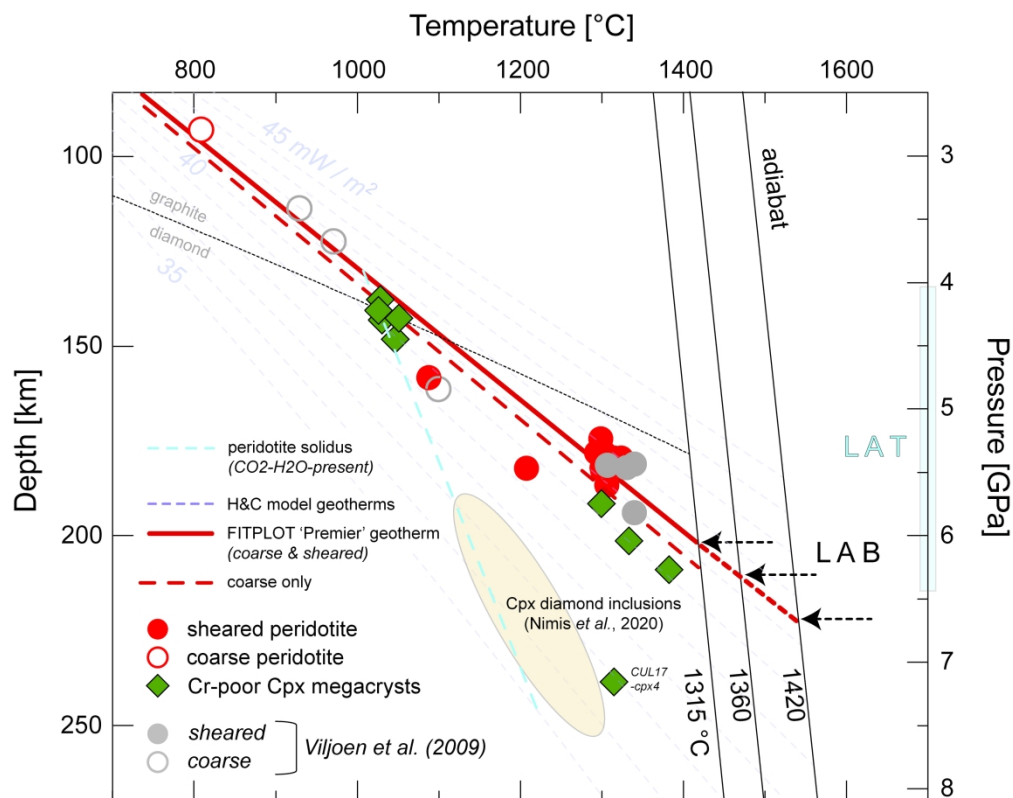


Figure 5:

Pressure–temperature systematics of peridotite xenoliths and Cr-poor clinopyroxene megacrysts from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Table 1 and Supplementary Data Appendix E). The ‘Premier’ paleogeotherm (thick solid red curve) was obtained by regression of P–T data for the here investigated and previously reported coarse and sheared / ‘deformed’ peridotite xenoliths using FITPLOT. The long-dashed red curve represents geotherm fitting that included only coarse peridotite xenoliths. All xenolith data, including those from Viljoen et al. (2009), are calculated or recalculated using the preferred TNG thermobarometer combination (Nickel & Green, 1985; Taylor, 1998). The P–T data for discrete clinopyroxene megacrysts were calculated according to Nimis & Taylor (2000), but are not included into the displayed geotherm fitting (outlier CUL17-cpx4 is discussed in the main text). The brown field for rare ‘cold’ clinopyroxene xenocrysts and inclusions in some Premier diamonds is based on the data reported in Nimis et al. (2020) (the complete clinopyroxene P–T dataset is displayed in Supplementary Data Figure 2). The graphite/diamond phase transition curve of Day (2012) and conductive model geotherms from Hasterok & Chapman (2011) are shown for reference. The CO<sub>2</sub>-H<sub>2</sub>O-present peridotite solidus curve is adopted from Foley et al. (2009) and has been extrapolated beyond 6 GPa applying the data compilations for high-P melting experiments in Tappe et al. (2018b) and Massuyeau et al. (2021). Adiabats for ambient mantle potential temperatures of 1315, 1360 and 1420°C are shown together with corresponding estimations of the petrological lithosphere–asthenosphere boundary (LAB) beneath the central Kaapvaal craton at ca. 1150 Ma. The blue vertical bar indicates the depth location of a lithosphere–asthenosphere transition zone (LAT), in which low-degree carbonate-rich melts may be ubiquitous (see main text).

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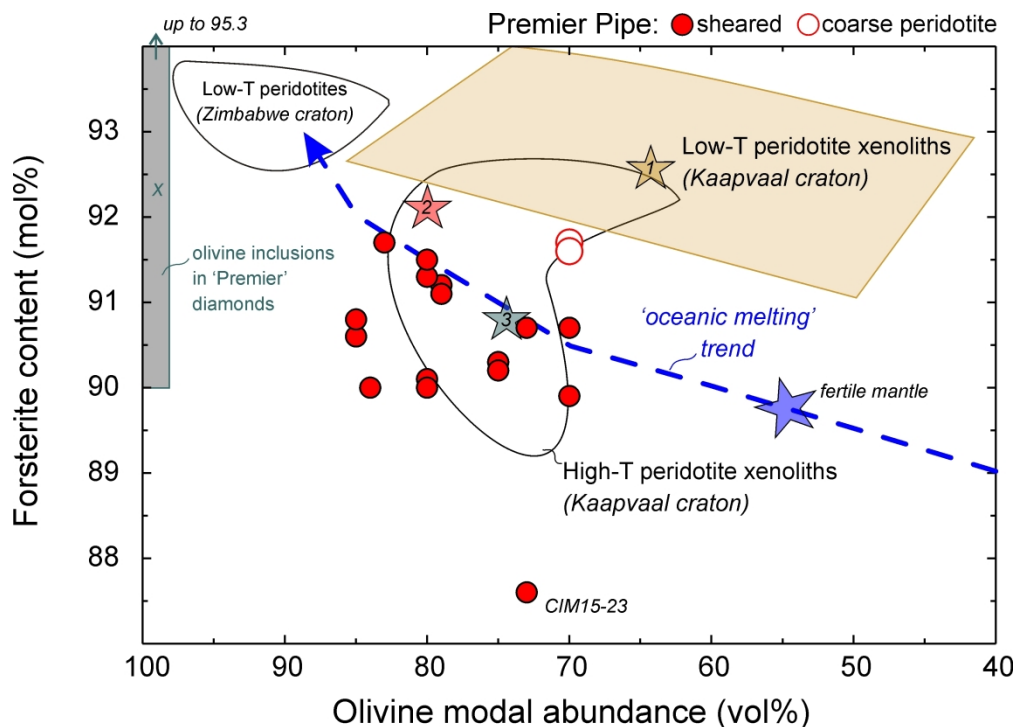


Figure 6:

Average forsterite contents versus corresponding olivine modal abundances of sheared and coarse garnet-facies peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D, E). For the sheared peridotite xenoliths from Premier, both porphyroclastic and neoblastic 'matrix' olivine compositions are shown, without discernable differences in their Mg-numbers within a single nodule. The field for olivine inclusions within Premier diamonds is based on data presented in Viljoen et al. (2014) and Korolev et al. (2018b), with the mean forsterite content shown as an 'X'. Fields for low- and high-temperature peridotite xenoliths from the Kaapvaal and Zimbabwe cratons are adopted from van der Meer et al. (2013). Averages for low-temperature peridotites from the Kaapvaal craton (golden star 1) and other major cratons worldwide (red star 2), as well as for modern abyssal peridotites (green star 3), are taken from Pearson & Wittig (2014). Olivine in sheared peridotites from Premier pipe and other kimberlite occurrences on the Kaapvaal craton resembles olivine from abyssal peridotites of the modern ocean basins, as is also apparent from the proximity to the 'oceanic melting trend' of Boyd (1989).

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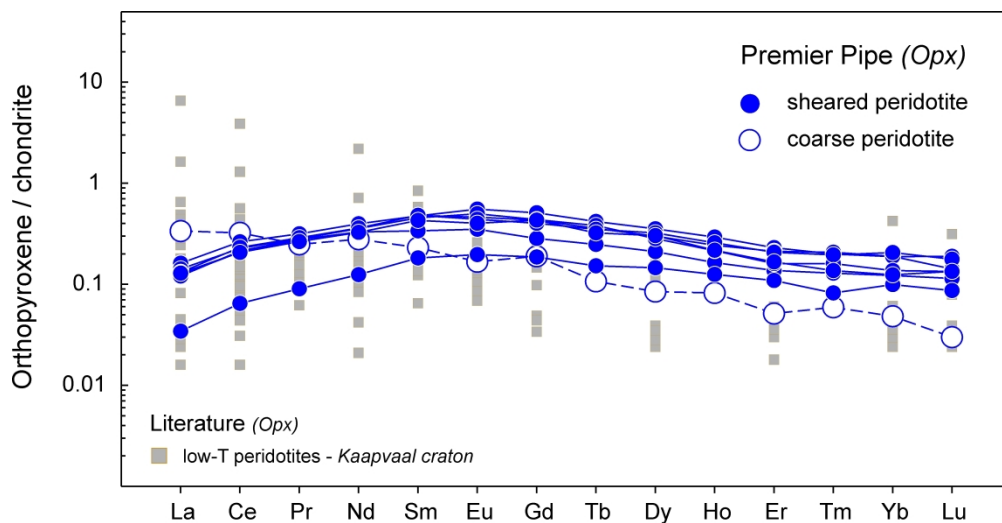


Figure 7:

Chondrite normalized REE distributions for orthopyroxene in sheared and coarse peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D).

The REE patterns for orthopyroxene in garnet-facies peridotite xenoliths derived from ca. 90 Ma old kimberlite pipes of Lesotho and Kimberley on the Kaapvaal craton (grey squares) are shown for reference (Simon et al., 2003, 2007). Chondrite values for normalization are from Palme & Jones (2003).

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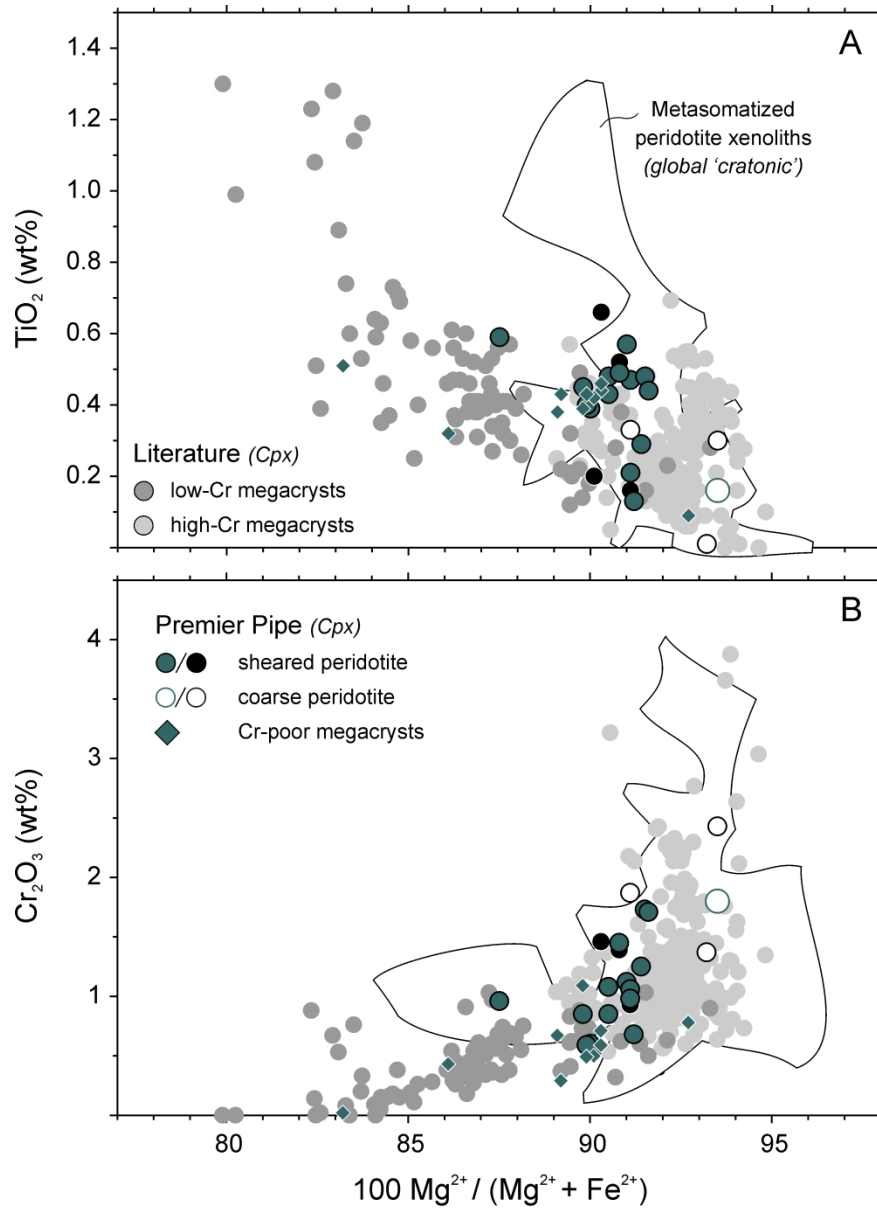


Figure 8:

TiO<sub>2</sub> (A) and Cr<sub>2</sub>O<sub>3</sub> (B) contents versus Mg-number for clinopyroxene in sheared and coarse peridotite xenoliths, as well as Cr-poor clinopyroxene megacrysts, from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D). Premier pipe literature data for clinopyroxene from sheared (filled black circles) and coarse (open black circles) peridotite xenoliths are taken from Viljoen et al. (2009). The data compilation for clinopyroxene megacrysts from kimberlite localities worldwide is based on Busweiler et al. (2018). The field for clinopyroxene compositions in metasomatized peridotite xenoliths from cratons worldwide is based on data provided by Kopylova et al. (1999), Simon et al. (2003, 2007), Aulbach et al. (2007), Rehfeldt et al. (2008), Kargin et al. (2017), and Busweiler et al. (2018).

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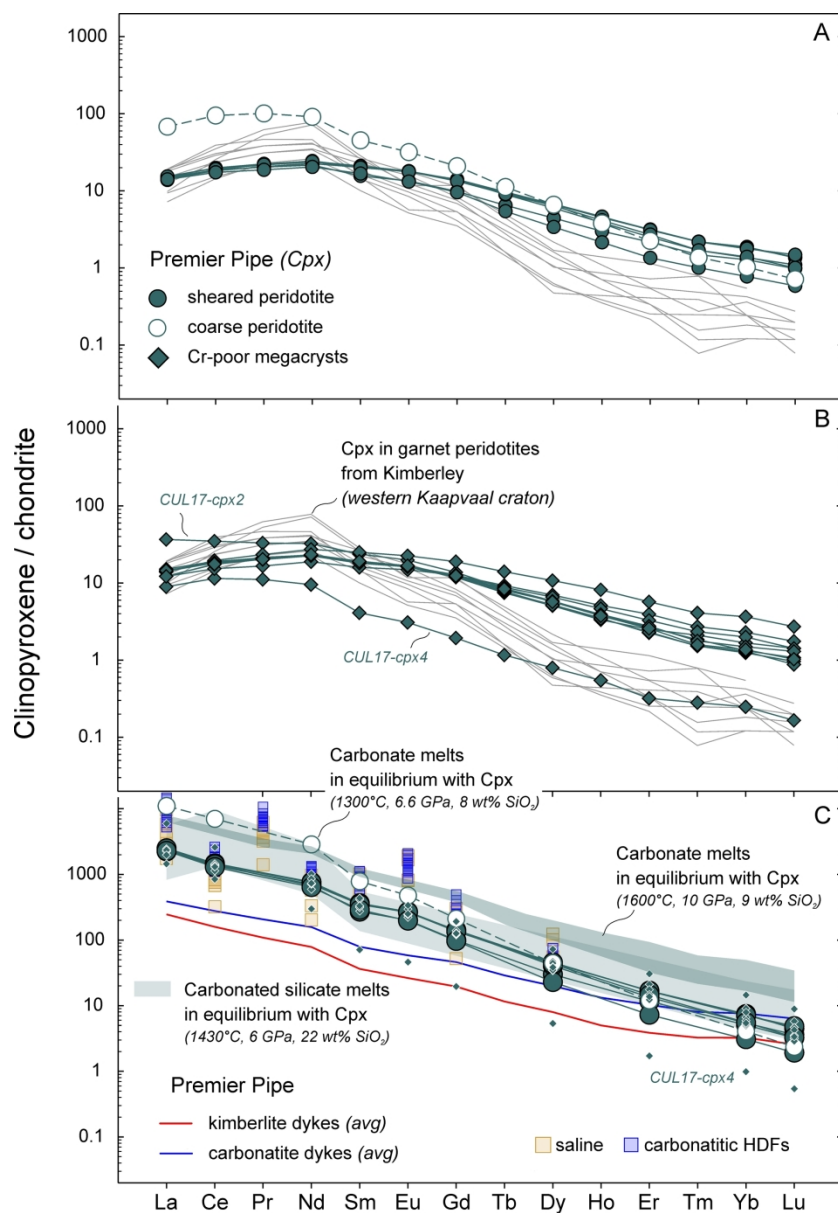


Figure 9:

Chondrite normalized REE distributions for clinopyroxene in sheared and coarse peridotite xenoliths (A) and for Cr-poor clinopyroxene megacrysts (B) from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D). (C) Reconstructed equilibrium melt compositions that may have been responsible for the metasomatic nature of clinopyroxene in garnet-facies peridotite xenoliths from the Premier kimberlite pipe, and may have also caused clinopyroxene megacryst formation. Both the effects of 'carbonate-dominated' (cpx/melt partition coefficients from Dasgupta et al., 2009 and Giris et al., 2013) and 'carbonated silicate' (Keshav et al., 2005) melts at P-T conditions equivalent to the lithosphere-asthenosphere boundary beneath the central Kaapvaal craton are shown. Average kimberlite (red line) and carbonatite (blue line) dyke compositions from Premier pipe are based on the data presented in Tappe et al. (2020b). The extremely high LREE-MREE concentrations of saline and high-Mg carbonatitic high-density fluids (HDFs) entrapped in diamonds from lithospheric mantle columns of cratons worldwide (Weiss et al., 2011, 2018) suggest their possible involvement in the origin of enriched clinopyroxene signatures within the Kaapvaal continental root. The REE patterns for clinopyroxene from garnet-facies peridotite xenoliths of the

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3 Kimberley kimberlite cluster on the western Kaapvaal craton (grey lines) are shown for reference in Panels  
4 A-B-C (Simon et al., 2007). Chondrite values for normalization are from Palme & Jones (2003).

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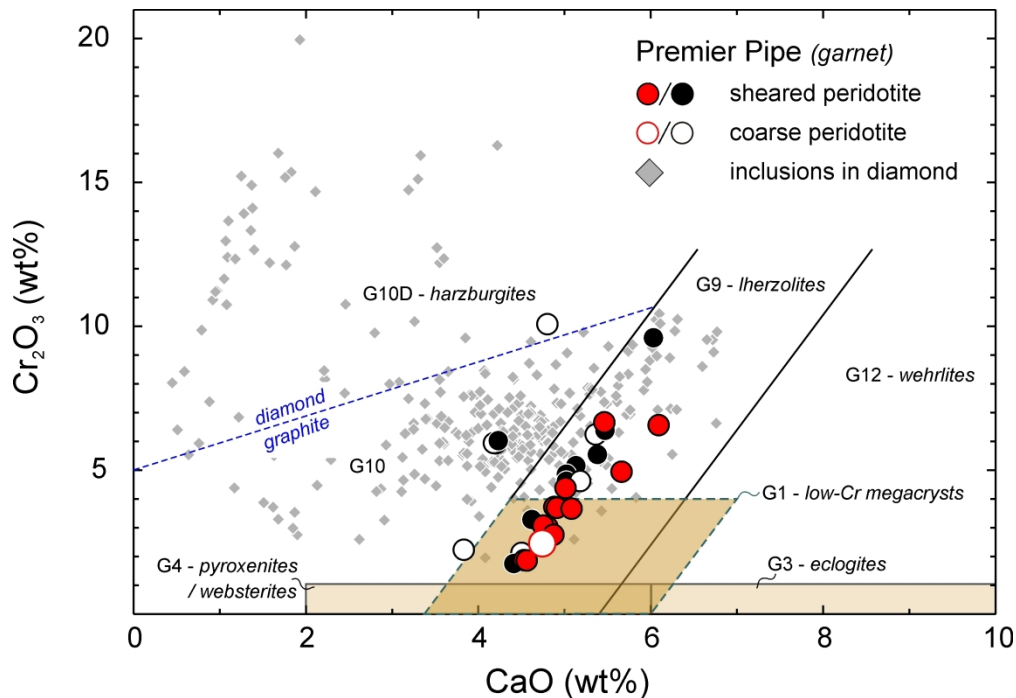


Figure 10:

$\text{Cr}_2\text{O}_3$  versus  $\text{CaO}$  contents (wt.%) of garnet in sheared and coarse peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D). The garnet 'G-type' classification and the 'graphite-diamond constraint' are according to Grütter et al. (2004).

Literature data for garnet from sheared (filled black circles) and coarse (open black circles) peridotite xenoliths, as well as from diamond inclusions (filled grey diamond symbol) are taken from Viljoen et al. (2009) and Viljoen et al. (2014), respectively. Note the partial overlap between G9 lherzolitic garnet grains from Premier pipe and the compositional field of G1 low-Cr garnet megacrysts.

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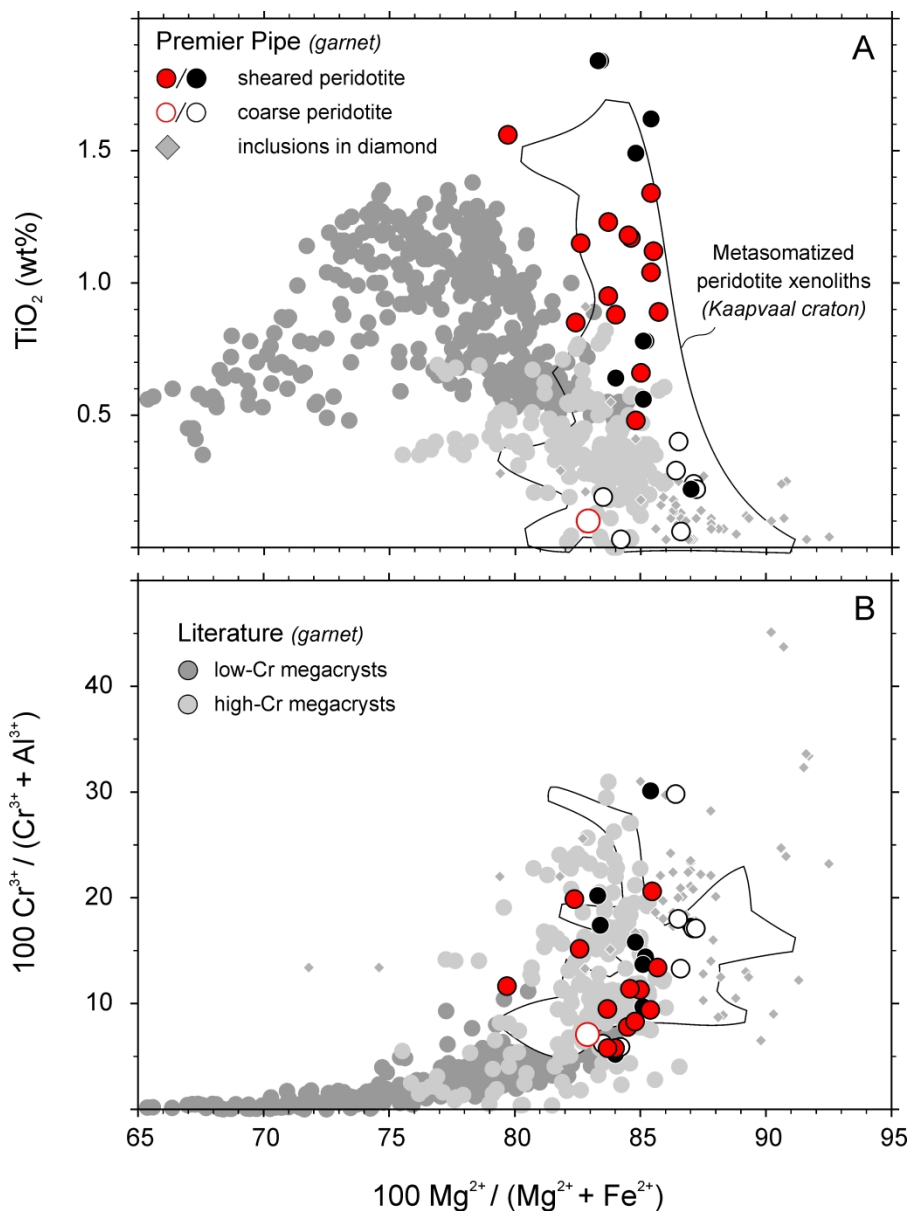


Figure 11:

TiO<sub>2</sub> content (A) and Cr-number (B) versus Mg-number for garnet in sheared and coarse peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D). Premier pipe literature data for garnet from sheared (filled black circles) and coarse (open black circles) peridotite xenoliths, as well as from diamond inclusions (filled grey diamond symbol) are taken from Viljoen et al. (2009) and Viljoen et al. (2014), respectively. The data compilation for garnet megacrysts from localities worldwide is based on Bussweiler et al. (2018), and dominated by southern African occurrences for the low-Cr suite (filled dark grey circles), as opposed to occurrences located mainly on the Slave craton in Canada for the high-Cr megacrysts (filled light grey circles). The field for garnet compositions from metasomatized peridotite xenoliths of the Kaapvaal craton is based on data compiled in Bussweiler et al. (2018).

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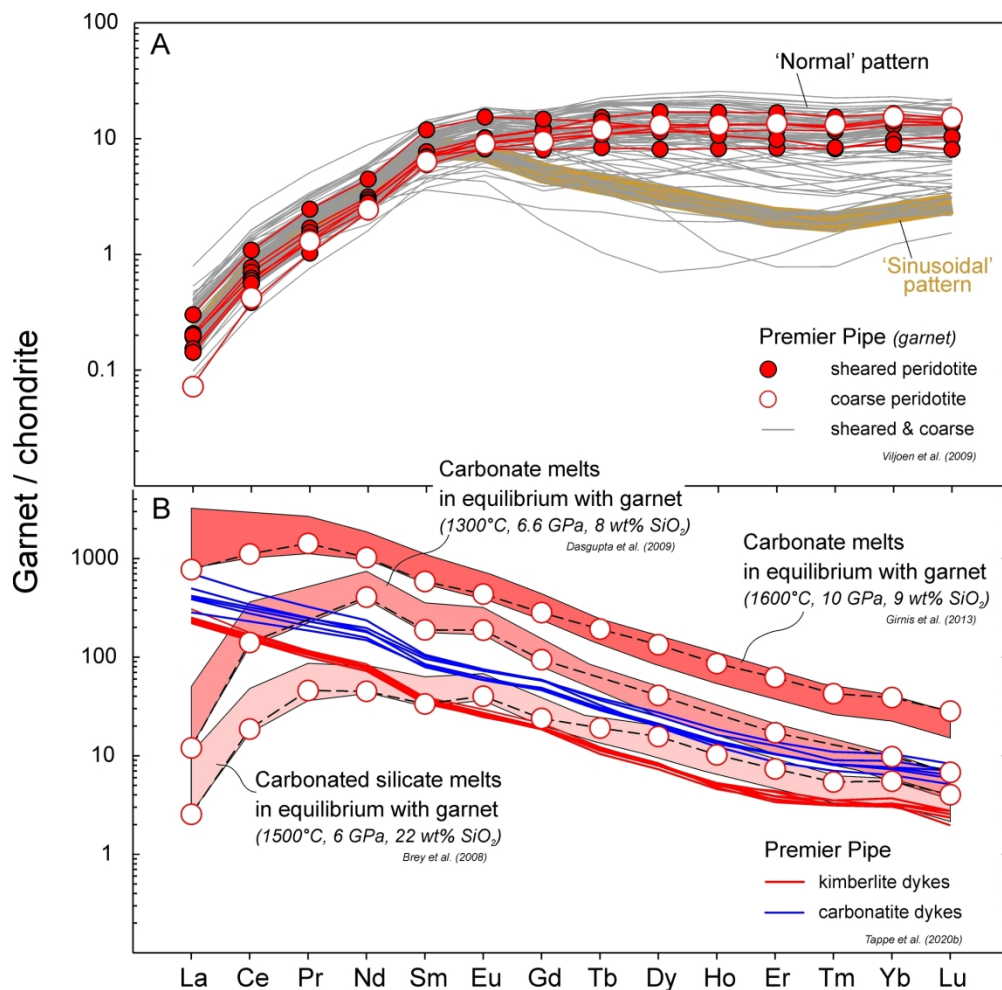


Figure 12:

(A) Chondrite normalized REE distributions for garnet in sheared and coarse peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D). Premier pipe literature data for garnet from sheared / deformed and coarse peridotite xenoliths (grey lines) are taken from Viljoen et al. (2009) and display both 'normal' and 'sinusoidal' patterns. (B) Reconstructed equilibrium melt compositions that may have been responsible for the metasomatic nature of garnet in peridotite xenoliths from the Premier kimberlite pipe. Both the effects of 'carbonate-dominated' (garnet/melt partition coefficients from Dasgupta et al., 2009 and Giris et al., 2013) and 'carbonated silicate' (Brey et al., 2008) melts at P-T conditions equivalent to the lithosphere-asthenosphere boundary beneath the central Kaapvaal craton are shown. Data for fresh kimberlite (red lines) and carbonatite (blue lines) dykes from Premier pipe are from Tappe et al. (2020b). Chondrite values for normalization are from Palme & Jones (2003).

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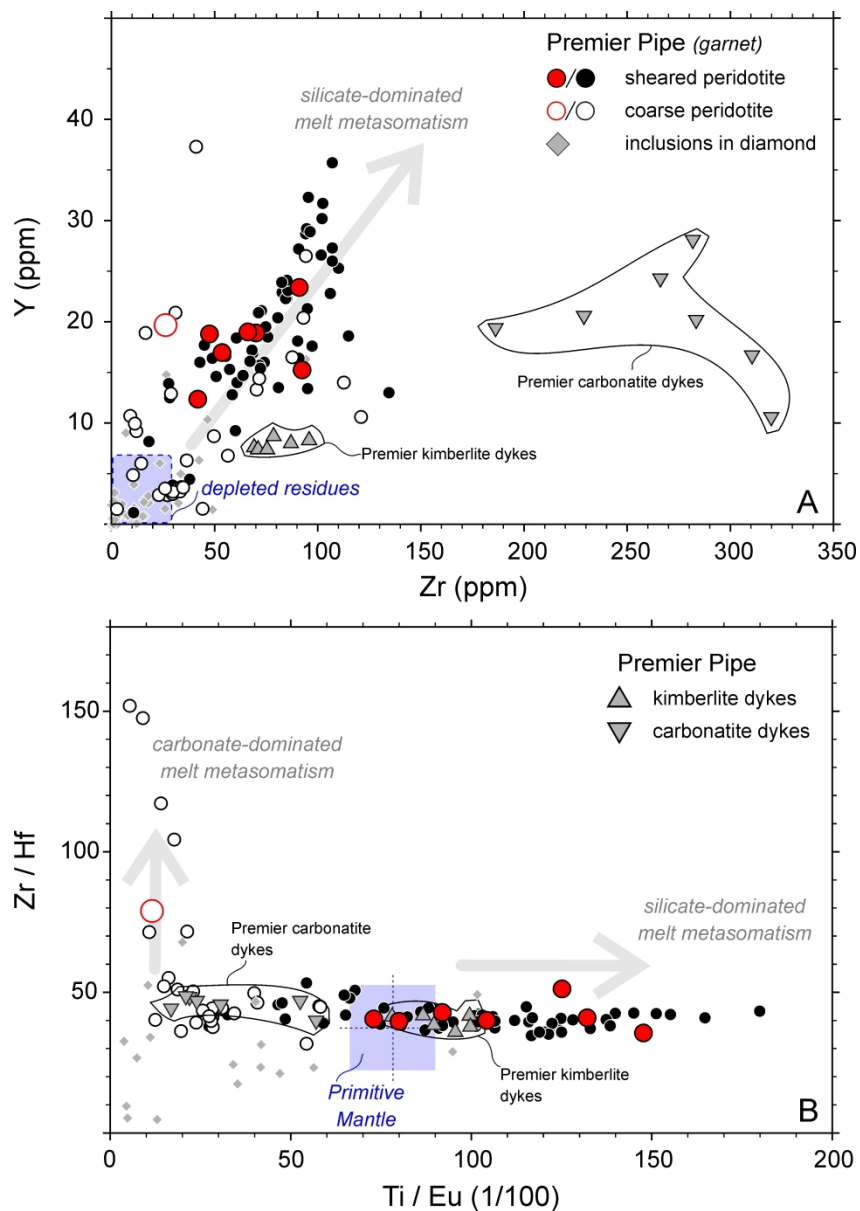


Figure 13:

(A) Y versus Zr contents, and (B) Zr/Hf versus Ti/Eu ratios of garnet in sheared and coarse peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D). Premier pipe literature data for garnet from sheared (filled black circles) and coarse (open black circles) peridotite xenoliths, as well as from diamond inclusions (filled grey diamond symbol) are taken from Viljoen et al. (2009) and Viljoen et al. (2014), respectively. Data for fresh kimberlite (filled grey triangle) and carbonatite (filled grey inverted triangle) dykes from Premier pipe are from Tappe et al. (2020b). The layout of Panel A is adopted from Griffin & Ryan (1995), with some modification to the metasomatic trend. The layout of Panel B is adopted from Shu & Brey (2015), with modifications of the metasomatic trends. Note that the Ti/Eu values have been divided by a hundred for simplicity's sake. Primitive mantle composition is from Palme & O'Neill (2003).

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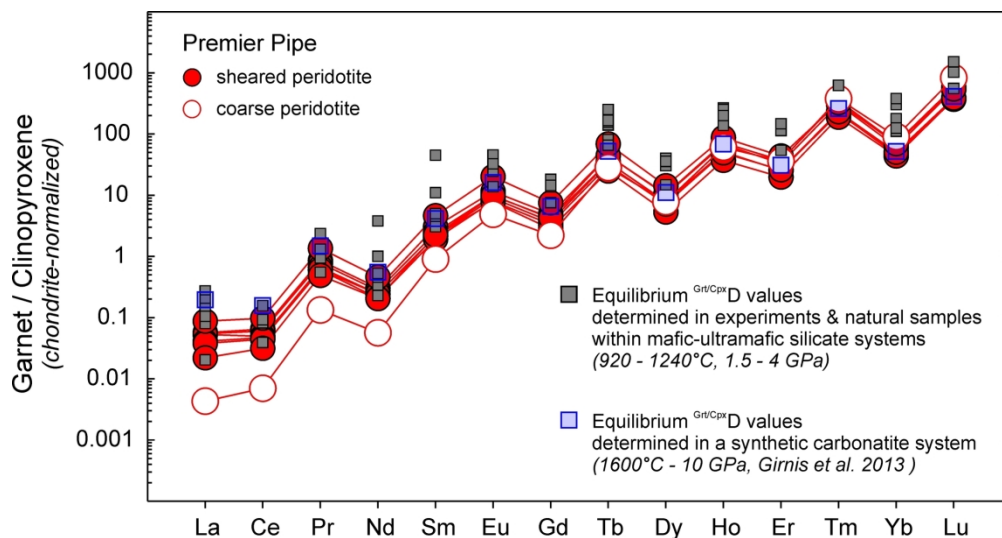


Figure 14:

REE distribution coefficients (chondrite-normalized, Palme & Jones, 2003) for coexisting garnet and clinopyroxene in sheared and coarse peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix D). A range of equilibrium Gd/CpxD values determined either experimentally (Green et al., 2000; Adam & Green, 2006; Xiong, 2006; Girmis et al., 2013) or on natural rocks (Zack et al., 1997) is shown for comparison, indicating trace element equilibrium to subtle disequilibrium for the Premier sheared peridotite xenoliths, and notable LREE disequilibrium for coarse peridotite sample CIM15-040. For this sample, clinopyroxene is more LREE enriched than predicted by the composition of coexisting garnet.

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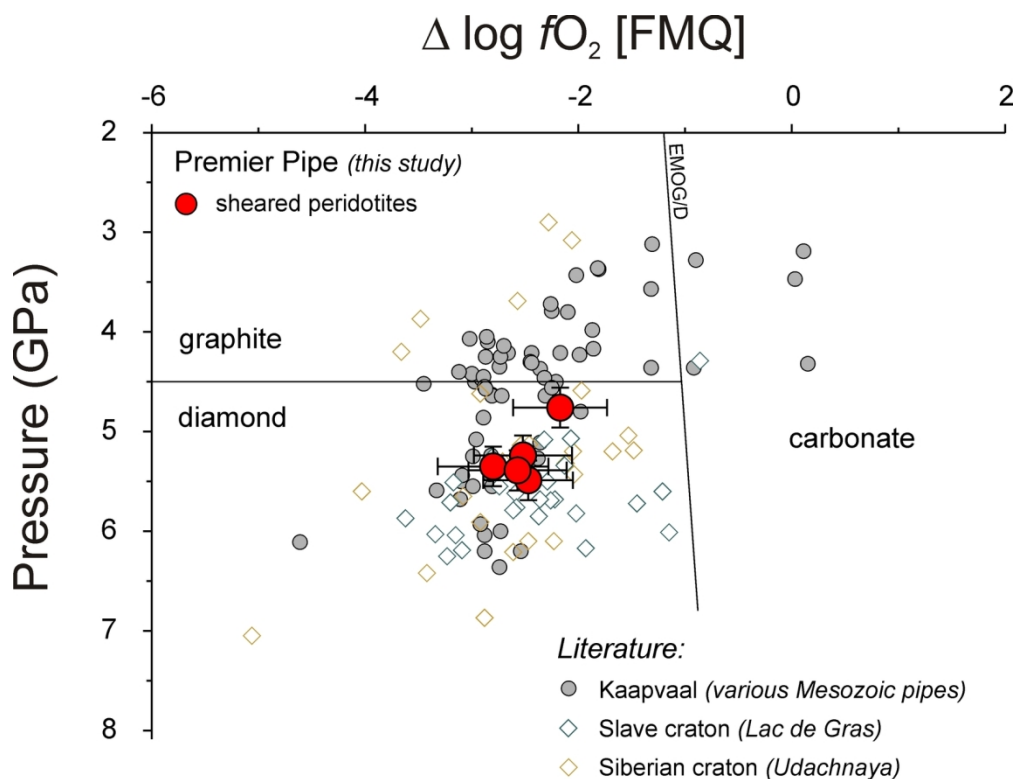


Figure 15:

The pressure versus  $\Delta \log fO_2$  systematics (relative to the FMQ buffer) of sheared garnet peridotite xenoliths from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Table 1 and Supplementary Data Appendix E). The error bars entail the 2S.E. uncertainties of the  $fO_2$  calculations using the formulation by Stagno et al. (2013) and an estimated uncertainty of 0.2 GPa for pressure calculations using the calibration by Nickel & Green (1985). The previously published data for garnet peridotite xenoliths from the Kaapvaal (Woodland & Koch, 2003; Creighton et al., 2009; Lazarov et al., 2009; Hanger et al., 2015), Slave (Creighton et al., 2010; Yaxley et al., 2017) and Siberian (Goncharov et al., 2012; Yaxley et al., 2012) cratons are recalculated using the Stagno et al. (2013) and Nickel & Green (1985) methods for  $fO_2$  and pressure estimations, respectively (Supplementary Data Appendix B). The graphite/diamond phase transition and graphite/diamond versus carbonate stability reaction in a peridotitic system [EMOG/D] at cratonic thermal conditions are adopted from Yaxley et al. (2017).

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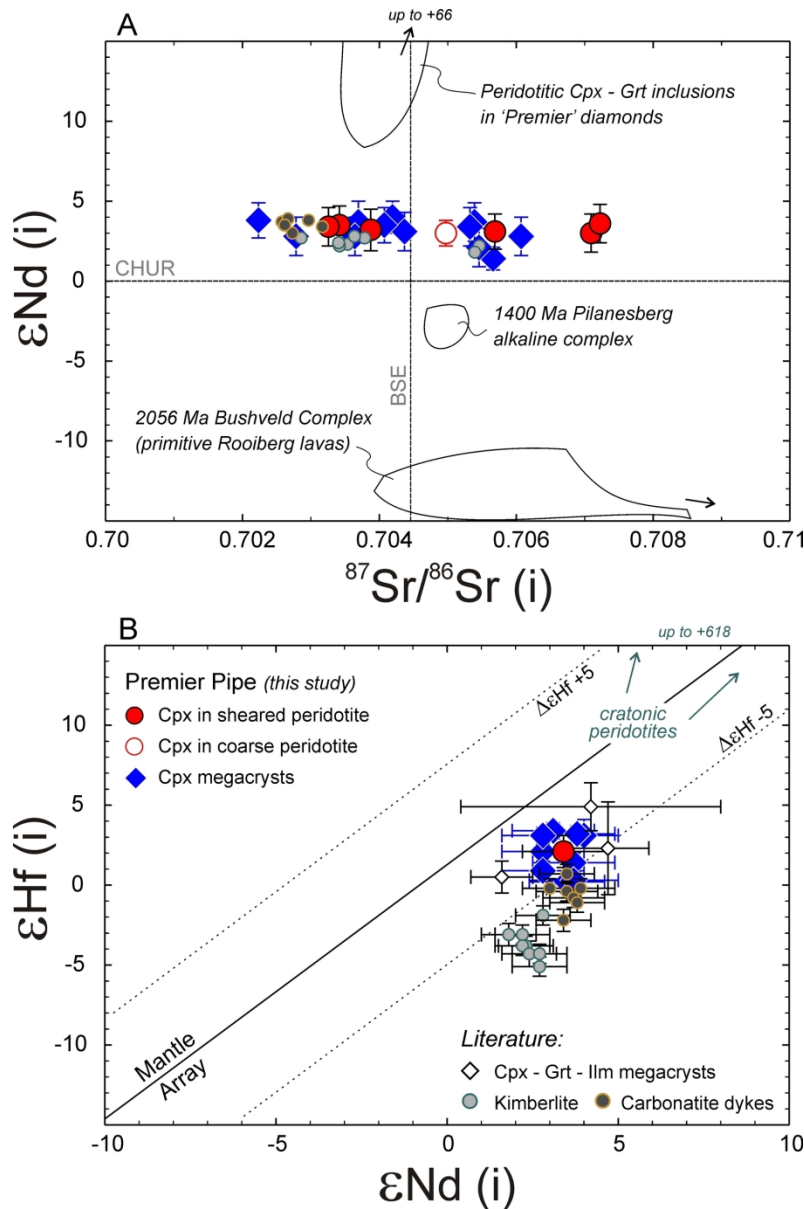


Figure 16:

Initial  $\epsilon_{\text{Nd}}$  versus  $^{87}\text{Sr}/^{86}\text{Sr}$  (A) and  $\epsilon_{\text{Hf}}$  versus  $\epsilon_{\text{Nd}}$  (B) for clinopyroxene from sheared and coarse peridotite xenoliths, as well as for discrete Cr-poor clinopyroxene megacrysts from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Table 2 and Supplementary Data Appendix E). Published data for Cr-poor megacrysts (cpx – clinopyroxene, grt – garnet, ilm – ilmenite) from Premier pipe are taken from Nowell et al. (2004). The data for Premier kimberlite and carbonatite dykes are from Tappe et al. (2020b). In Panel A, the Sr-Nd isotope data for clinopyroxene and garnet inclusions within peridotitic diamonds from Premier pipe (Richardson et al., 1993), the ca. 1400 Ma Pilanesberg alkaline complex (Cawthorn et al., 2012), and basaltic lavas from the Rooiberg Group of the ca. 2056 Ma Bushveld Complex on the central Kaapvaal craton (Günther et al., 2018) are shown for comparison. All data in Panel A and B are corrected to the kimberlite magma emplacement age of ca. 1150 Ma for Premier pipe (Tappe et al., 2018a) and they entail a full propagation of uncertainties shown by the error bars (see Methods and the footnote to Table 2). In Panel B, the 'mantle array' is after Chauvel et al. (2008) and the vectors pointing toward depleted ancient cratonic mantle peridotites are after Tappe et al. (2020b).

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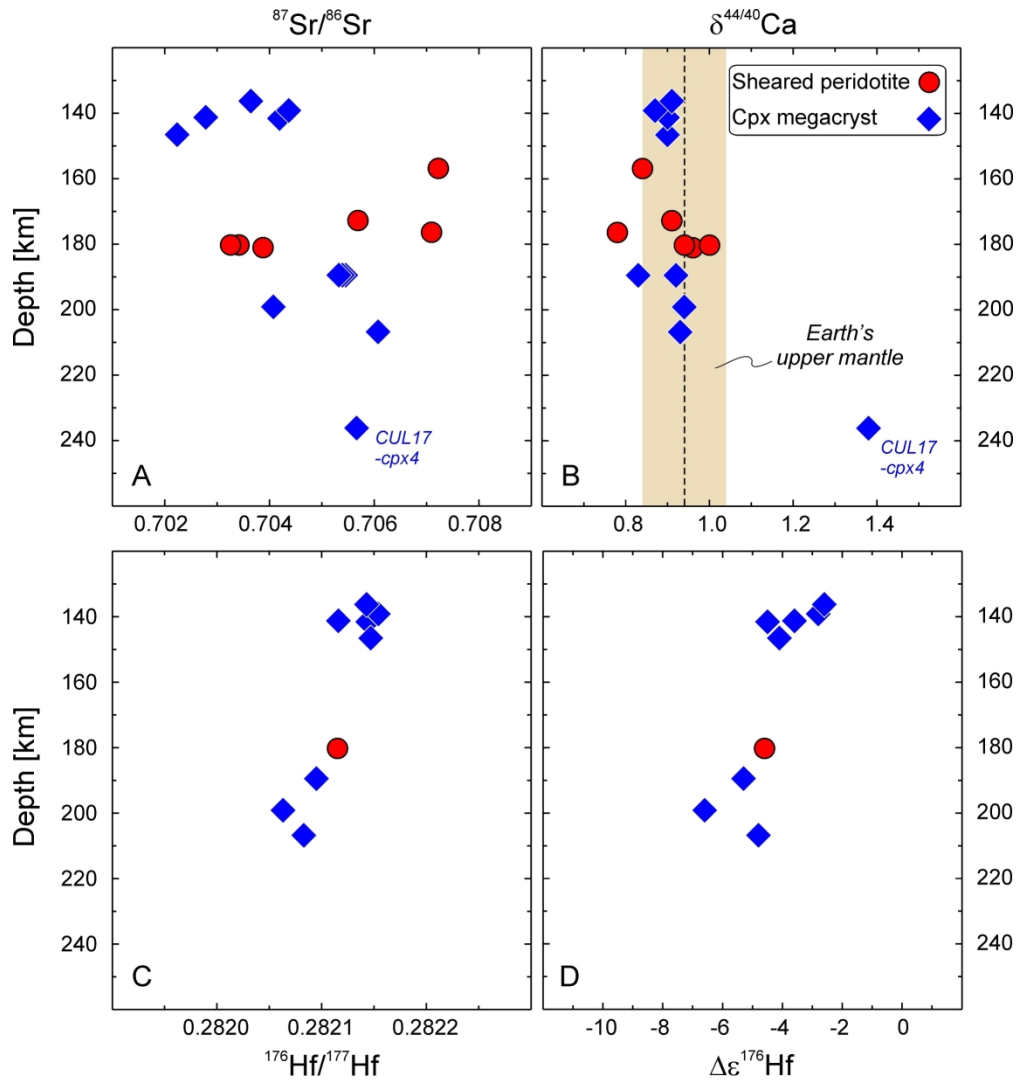


Figure 17:

Initial  $^{87}\text{Sr}/^{86}\text{Sr}$  (A),  $\delta^{44/40}\text{Ca}$  (B), initial  $^{176}\text{Hf}/^{177}\text{Hf}$  (C), and initial  $\Delta\epsilon^{176}\text{Hf}$  (D) compositions versus the equilibration depths of clinopyroxene porphyroclasts from sheared peridotite xenoliths and of discrete clinopyroxene megacrysts from the Premier kimberlite pipe, Cullinan Diamond Mine (all data are listed in Supplementary Data Appendix E). No hafnium isotope data are available for megacryst CUL17-cpx4 due to its very low Hf concentration. The upper mantle  $\delta^{44/40}\text{Ca}$  composition is after Chen et al. (2019).

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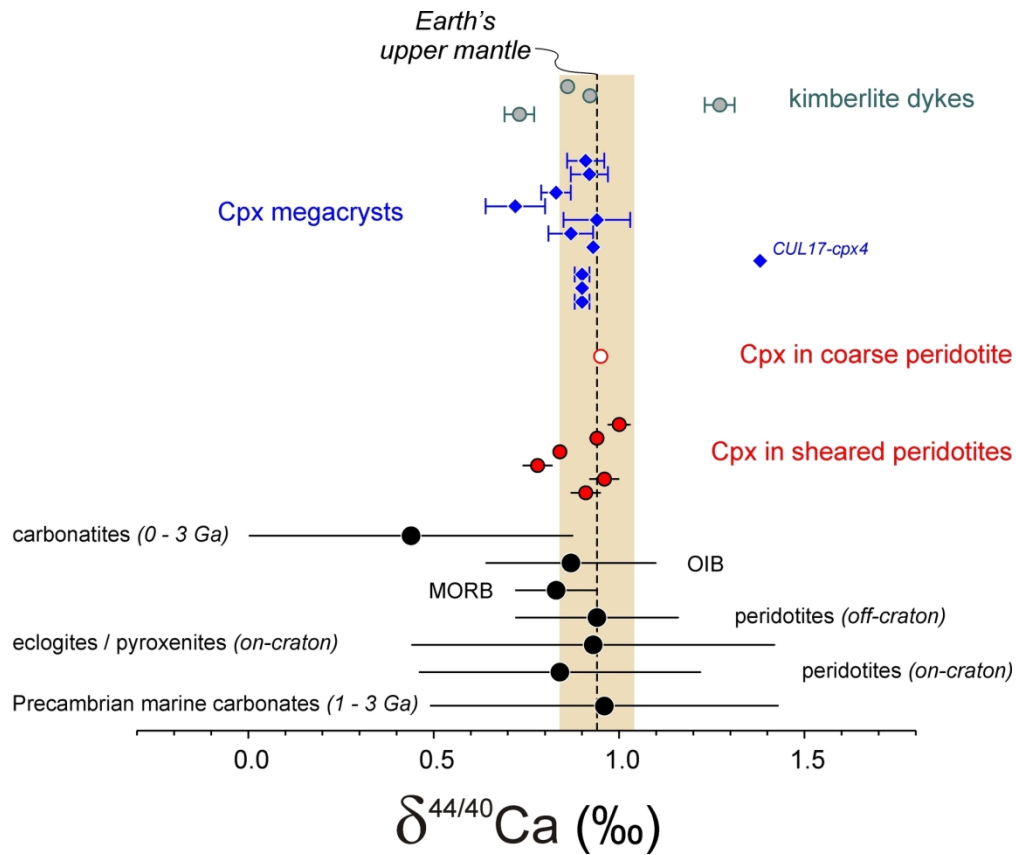


Figure 18:

The  $\delta^{44/40}\text{Ca}$  values for clinopyroxene from sheared and coarse peridotite xenoliths, as well as for discrete Cr-poor clinopyroxene megacrysts from the Premier kimberlite pipe, Cullinan Diamond Mine, reported relative to SRM-915a (all data are listed in Table 2 and Supplementary Data Appendix E). The Ca isotopic compositions of four kimberlite dykes from Premier pipe have also been analysed for comparative purposes. Published  $\delta^{44/40}\text{Ca}$  data for relevant rock types and geochemical reservoirs are taken from the following literature sources and compilations (averages  $\pm$  2S.D.): continental peridotite xenoliths (Kang et al., 2017), cratonic eclogites/pyroxenites (Smart et al., 2021), modern oceanic basalts (Schiller et al., 2016; Chen et al., 2019), modern to 3 Ga old Ca-Mg-Fe carbonatites (Amsellem et al., 2020; Sun et al., 2021) and 1-3 Ga old Precambrian marine carbonates (Blättler & Higgins, 2017). The average  $\delta^{44/40}\text{Ca}$  value for the upper mantle of  $0.94 \pm 0.1\text{‰}$  (dashed vertical line plus error envelop) is based on Chen et al. (2019).

162x137mm (300 x 300 DPI)



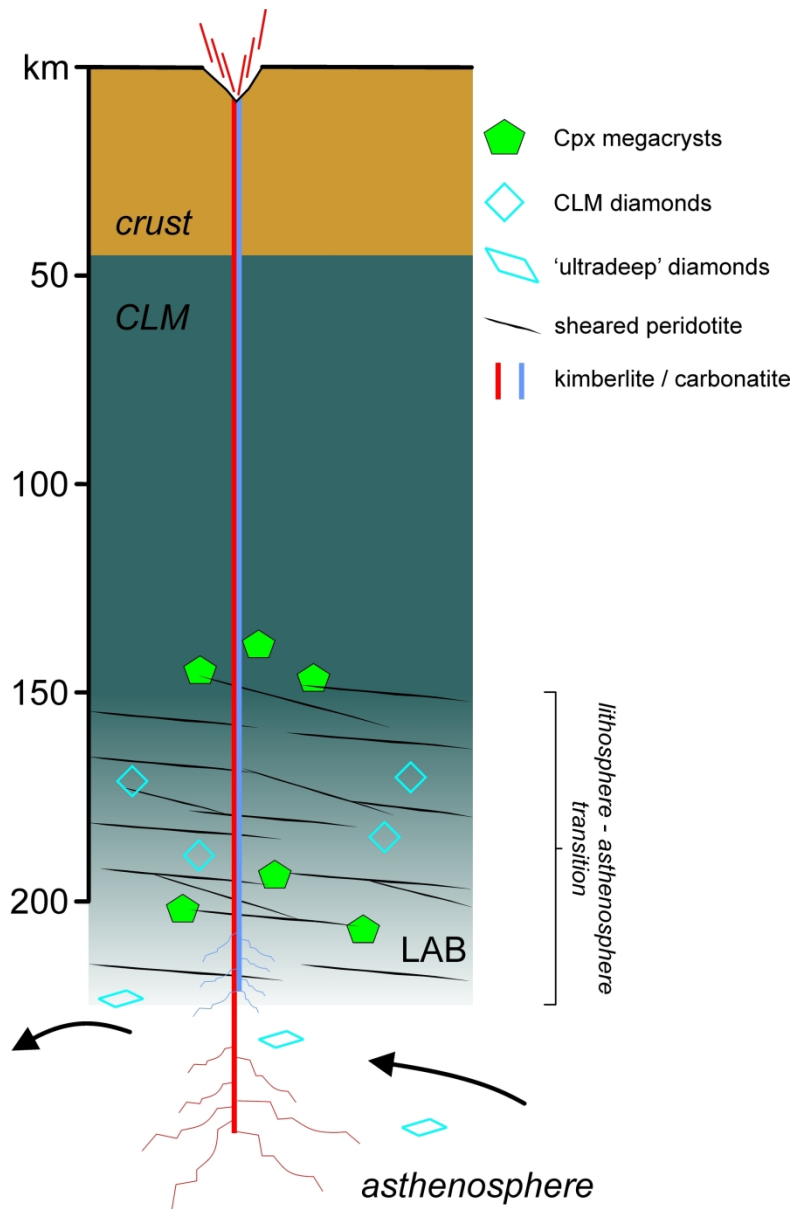


Figure 19:

Schematic model of the central Kaapvaal cratonic mantle lithosphere during the ca. 1150 Ma kimberlite and carbonatite magma eruptions at Premier/Cullinan. This study proposes a  $\geq 50$  km thick lithosphere–asthenosphere transition zone between  $\sim 150$ –225 km depth, where strong plastic deformation creates a network of narrow subhorizontal shear zones as represented by abundant sheared peridotite xenoliths. Stress-driven segregation of percolating carbonate-rich melts may provide a link between the common association of megacrysts and sheared peridotites as part of kimberlite mantle cargo. Both lithospheric and sublithospheric (i.e., ‘ultradeep’) diamonds are shown for reference, but they are not discussed in detail as part of this model. CLM – cratonic lithospheric mantle, LAB – lithosphere–asthenosphere boundary.

187x287mm (300 x 300 DPI)

Table 1: Petrography, pressure-temperature and oxygen fugacity estimates for peridotite xenoliths and clinopyroxene megacrysts from the Premier kimberlite, Kaapvaal craton, South Africa.											
ID	Texture	Grain size	Mineral abundance (%)				*Thermobarometry		Garnet log $fO_2$		
			Ol	Opx	Cpx	Grt	P (GPa)	T (°C)	$Fe^{3+}/\Sigma Fe$	$\Delta FMQ^{**}$	1SE
<b>Peridotite xenoliths</b>											
CIM15-001	sheared	VF (<100 $\mu m$ )	79	10	4	7	5.6	1307			
CIM15-003	sheared	F (100-500 $\mu m$ )	79	8	7	6	5.2	1299	0.085	-2.52	0.23
CIM15-008	sheared	M (>500 $\mu m$ )	73	12	8	7	5.5	1315			
CIM15-015	sheared	F (100-500 $\mu m$ )	80	7	7	6	5.3	1294			
CIM15-016	sheared	F (100-500 $\mu m$ )	84	6	6	4	5.4	1324	0.088	-2.57	0.23
CIM15-018	sheared	F (100-500 $\mu m$ )	85	5	7	3	5.5	1303			
CIM15-021	sheared	VF (<100 $\mu m$ )	80	9	7	4	5.5	1302	0.080	-2.47	0.21
CIM15-022	sheared	F (100-500 $\mu m$ )	70	11	12	7	5.3	1303	0.080	-2.80	0.26
CIM15-023	sheared	F (100-500 $\mu m$ )	73	13	8	6	5.5	1303			
CIM15-024	sheared	F (100-500 $\mu m$ )	80	8	7	5	5.4	1315			
CIM15-038	sheared	VF (<100 $\mu m$ )	85	7	5	3	5.4	1306			
ST16-26-CUL	sheared	M (>500 $\mu m$ )	75	13	9	3	5.5	1208			
CIM15-035	<sup>a</sup> coarse in sheared	F (100-500 $\mu m$ ), up to 1.2 cm	70	10	15	5	4.8	1088	0.070	-2.17	0.22
CIM15-050	<sup>a</sup> coarse in sheared	F (100-500 $\mu m$ ), up to 1.2 cm	83	5	10	2	5.5	1301			
CIM15-050re	<sup>a</sup> coarse in sheared	F (100-500 $\mu m$ ), up to 1.2 cm	83	5	10	2	5.5	1301			
CIM15-040	coarse	>2 mm (2-5 mm)	70	14	11	5	2.8	809			
<b>Clinopyroxene megacrysts</b>											
CUL17-CPX01	megacrystic	1 $\times$ 3 cm					4.3	1030			
CUL17-CPX02	megacrystic	1 $\times$ 3 cm					4.4	1046			
CUL17-CPX03	megacrystic	1 $\times$ 3 cm					4.3	1051			
CUL17-CPX04	megacrystic	2 $\times$ 2 cm					7.2	1313			
CUL17-CPX05	megacrystic	1 $\times$ 2 cm					6.3	1382			
CUL17-CPX06	megacrystic	2 $\times$ 2 cm					4.2	1026			
CUL17-CPX07	megacrystic	2 $\times$ 2 cm					6.0	1333			
CUL17-CPX08	megacrystic	1 $\times$ 2 cm					<i>no solution</i>	<i>no solution</i>			
CUL17-CPX09	megacrystic	2 $\times$ 3 cm					5.7	1299			
CUL17-CPX09re1	megacrystic	2 $\times$ 3 cm					5.7	1299			
CUL17-CPX09re2	megacrystic	2 $\times$ 3 cm					5.7	1299			
CUL17-CPX10	megacrystic	1 $\times$ 2 cm					4.1	1028			
*Preferred pressure and temperature estimations for garnet-facies peridotites based on formulations in <a href="#">Nickel &amp; Green (1985)</a> and <a href="#">Taylor (1998)</a> , respectively.											
*Pressure and temperature calculations for clinopyroxene megacrysts in equilibrium with garnet-facies peridotites based on formulations in <a href="#">Nimis &amp; Taylor (2000)</a> .											
**log $fO_2$ calculations for garnet peridotites based on formulation in <a href="#">Stagno et al. (2013)</a> .											
Texture and grain size descriptions follow <a href="#">Harte (1977)</a> : VF - very fine grained, F - fine grained, M - medium grained.											
<sup>a</sup> Cpx megacrystic domains in sheared Ol-rich matrix.											
<a href="http://www.petrology.oupjournals.org/">http://www.petrology.oupjournals.org/</a>											
Ol - olivine, Opx - orthopyroxene, Cpx - clinopyroxene, Grt - garnet.											

**Table 2:** Sr-Nd-Hf-Ca isotopic compositions of garnet peridotite xenoliths and clinopyroxene megacrysts from the Premier kimberlite, Kaapvaal craton, South Africa.

ID	Texture	Clinopyroxene isotopic compositions														Garnet isotopes						
		<sup>87</sup> Sr/ <sup>86</sup> Sr <sub>m</sub>	2SE	<sup>87</sup> Rb/ <sup>87</sup> Sr	<sup>87</sup> Sr/ <sup>86</sup> Sr <sub>i</sub>	<sup>143</sup> Nd/ <sup>144</sup> Nd <sub>m</sub>	2SE	<sup>147</sup> Sm/ <sup>144</sup> Nd	<sup>143</sup> Nd/ <sup>144</sup> Nd <sub>i</sub>	εNd <sub>i</sub>	*2SD	<sup>176</sup> Hf/ <sup>177</sup> Hf <sub>m</sub>	2SE	<sup>176</sup> Lu/ <sup>177</sup> Hf	<sup>176</sup> Hf/ <sup>177</sup> Hf <sub>i</sub>	εHf <sub>i</sub>	*2SD	** ΔεHf <sub>i</sub>	δ <sup>44</sup> Ca <sub>SRM915a</sub> (‰)	2SE	δ <sup>44</sup> Ca <sub>SRM915a</sub> (‰)	2SE
<b>Peridotite xenoliths</b>																						
CIM15-001	sheared																					
CIM15-003	sheared	0.705728	0.000009	0.00241	0.705688	0.512406	0.000005	0.14504	0.511311	3.1	1.1							0.91	0.04		1.24	0.02
CIM15-008	sheared																					
CIM15-015	sheared																					
CIM15-016	sheared																					
CIM15-018	sheared																					
CIM15-021	sheared	0.704232	0.000009	0.02164	0.703875	0.512595	0.000006	0.16997	0.511312	3.2	1.3							0.96	0.04		1.06	0.04
CIM15-022	sheared	0.707104	0.000007	0.00062	0.707094	0.512541	0.000006	0.16381	0.511304	3.0	1.2							0.78	0.04		1.12	0.03
CIM15-023	sheared																					
CIM15-024	sheared																					
CIM15-038	sheared																					
ST16-26-CUL	sheared																					
CIM15-035	<sup>a</sup> coarse in sheared	0.707364	0.000007	0.00850	0.707224	0.512586	0.000007	0.16578	0.511334	3.6	1.2							0.84	0.003		1.19	0.05
CIM15-050	<sup>a</sup> coarse in sheared	0.703453	0.000006	0.00223	0.703416	0.512560	0.000005	0.16274	0.511331	3.5	1.2							0.94	0.02			
CIM15-050re	<sup>a</sup> coarse in sheared	0.703292	0.000009	0.00223	0.703256	0.512553	0.000006	0.16274	0.511325	3.4	1.2	0.282183	0.000024	0.00312	0.282115	2.1	1.0	-4.6	1.00	0.03		
CIM15-040	coarse	0.705421	0.000007	0.02719	0.704973	0.512040	0.000006	0.09762	0.511303	3.0	0.8							0.95	0.01		1.87	0.03
<b>Clinopyroxene megacrysts</b>																						
CUL17-CPX01	megacrystic	0.704376	0.000006	0.01119	0.704192	0.512363	0.000005	0.13366	0.511354	4.0	1.0	0.282169	0.000025	0.00116	0.282144	3.1	1.0	-4.5	0.90	0.02		
CUL17-CPX02	megacrystic	0.703590	0.000006	0.08229	0.702235	0.512475	0.000007	0.14973	0.511345	3.8	1.1	0.282174	0.000007	0.00126	0.282147	3.2	0.5	-4.1	0.90	0.01		
CUL17-CPX03	megacrystic	0.703050	0.000006	0.01616	0.702784	0.512511	0.000006	0.16145	0.511292	2.8	1.2	0.282169	0.000016	0.00243	0.282116	2.1	0.7	-3.6	0.90	0.02		
CUL17-CPX04	megacrystic	0.705687	0.000005	0.00140	0.705664	0.511875	0.000006	0.08647	0.511223	1.4	0.7								1.38	0.003		
CUL17-CPX05	megacrystic	0.706135	0.000007	0.00373	0.706073	0.512553	0.000005	0.16668	0.511294	2.8	1.2	0.282274	0.000011	0.00882	0.282083	0.9	0.6	-4.8	0.93	0.01		
CUL17-CPX06	megacrystic	0.704666	0.000005	0.01810	0.704368	0.512522	0.000005	0.16042	0.511311	3.1	1.2	0.282176	0.000008	0.00103	0.282154	3.4	0.5	-2.8	0.87	0.06		
CUL17-CPX07	megacrystic	0.704317	0.000007	0.01484	0.704073	0.512406	0.000006	0.14246	0.511330	3.5	1.1	0.282175	0.000015	0.00516	0.282063	0.2	0.7	-6.6	0.94	0.09		
CUL17-CPX08	megacrystic	0.703704	0.000013	0.00055	0.703695	0.512629	0.000005	0.17080	0.511339	3.7	1.3	0.282141	0.000006	0.00355	0.282064	0.3	0.5	-6.9	0.72	0.08		
CUL17-CPX09	megacrystic	0.705467	0.000006	0.00068	0.705456	0.512454	0.000007	0.15869	0.511256	2.1	1.2								0.83	0.04		
CUL17-CPX09re1	megacrystic	0.705402	0.000006	0.00068	0.705391	0.512539	0.000006	0.15869	0.511341	3.7	1.2	0.282201	0.000011	0.00489	0.282095	1.4	0.6	-5.3	0.92	0.05		
CUL17-CPX09re2	megacrystic	0.705336	0.000008	0.00068	0.705325	0.512523	0.000005	0.15869	0.511325	3.4	1.2											
CUL17-CPX10	megacrystic	0.703708	0.000007	0.00399	0.703643	0.512527	0.000005	0.16366	0.511291	2.8	1.2	0.282166	0.000009	0.00109	0.282143	3.1	0.5	-2.6	0.91	0.05		
*The 2-sigma uncertainties of the epsilon Nd and Hf values entail a full error propagation (uncertainties of the 1150 Ma age and parent-daughter element ratios are set at 1% and 5%, respectively).																						
**Initial ΔεHf is defined as εHf <sub>i</sub> - (1.59*εNd <sub>i</sub> +1.28), such that analyses with negative values fall by definition below the Nd-Hf isotope regression line of Chauvel et al. (2008).																						
<sup>a</sup> Clinopyroxene megacrystic domains in sheared olivine-rich matrix.																						
Initial isotope ratios calculated for the Premier kimberlite emplacement age of 1150 Ma (Tappe et al., 2018a) using the following decay constants: <sup>87</sup> Rb 1.42*10 <sup>-11</sup> a <sup>-1</sup> , <sup>147</sup> Sm 6.54*10 <sup>-12</sup> a <sup>-1</sup> , <sup>176</sup> Lu 1.865*10 <sup>-11</sup> a <sup>-1</sup> .																						
Initial epsilon Nd values are calculated using <sup>147</sup> Sm decay constant of 6.54*10 <sup>-12</sup> a <sup>-1</sup> (Lugmair & Marti, 1978); ( <sup>143</sup> Nd/ <sup>144</sup> Nd) <sub>CHUR</sub> = 0.512630 and ( <sup>147</sup> Sm/ <sup>144</sup> Nd) <sub>CHUR</sub> = 0.1960 (Bouvier et al., 2008).																						
Initial epsilon Hf values are calculated using <sup>176</sup> Lu decay constant of 1.865*10 <sup>-11</sup> a <sup>-1</sup> (Scherer et al., 2001); ( <sup>176</sup> Hf/ <sup>177</sup> Hf) <sub>CHUR</sub> = 0.282785 and ( <sup>176</sup> Lu/ <sup>177</sup> Hf) <sub>CHUR</sub> = 0.0336 (Bouvier et al., 2008).																						
Measured Sr, Nd, and Hf isotope ratios are normalized to the following standard values:																						
NBS987 = <sup>87</sup> Sr/ <sup>86</sup> Sr value of 0.710245 (Thirlwall, 1991); JNd-1 = <sup>143</sup> Nd/ <sup>144</sup> Nd value of 0.512115 (Tanaka et al., 2000); JMC475 = <sup>176</sup> Hf/ <sup>177</sup> Hf value of 0.282160 (Blichert-Toft et al., 1997).																						
Powders of BCR-2 and BHVO-2 basalt reference materials were processed alongside the Premier/Cullinan xenoliths and megacrysts, and the following values were obtained during this study in 2018/2019:																						
BCR-2 = <sup>87</sup> Sr/ <sup>86</sup> Sr value of 0.705005(±7), n=2; BHVO-2 = <sup>87</sup> Sr/ <sup>86</sup> Sr value of 0.703469(±7), n=2.																						
BCR-2 = <sup>143</sup> Nd/ <sup>144</sup> Nd value of 0.512635(±6), n=3; BHVO-2 = <sup>143</sup> Nd/ <sup>144</sup> Nd value of 0.512990(±16), n=3.																						
BCR-2 = <sup>176</sup> Hf/ <sup>177</sup> Hf value of 0.282873(±22), n=3; BHVO-2 = <sup>176</sup> Hf/ <sup>177</sup> Hf value of 0.283108(±13), n=3.																						