| 1  | The elusive Congo craton margin during Gondwana breakup:  |
|----|---|
| 2  | Insights from lithospheric mantle structure and heat-flow beneath   |
| 3  | the Xaudum kimberlite province, NW Botswana   |
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| 17 | ABSTRACT  |
| 18 | The continental lithospheric mantle (CLM) beneath the southern margin of the Congo craton has                                   |
| 19 | remained elusive mainly because of thick Phanerozoic sedimentary cover concealing possible                                      |
| 20 | kimberlite and lamproite diatremes. In this study, we explore this lithospheric mantle section by                               |

21 using major and trace element compositions of mantle-derived clinopyroxene and garnet

22 xenocrysts from kimberlites of the ca. 84 Ma Nxau Nxau cluster in NW Botswana, which is part 23 of the poorly known Xaudum kimberlite province extending into northern Namibia. We utilize 24 these data to better understand the thermal and compositional evolution of the lithospheric mantle 25 at the southern margin of the Congo craton. The clinopyroxene population (83 individual grains) 26 comprises Cr-rich and Cr-poor diopsides with variable major (Al<sub>2</sub>O<sub>3</sub>, Na<sub>2</sub>O, Mg#) and 27 incompatible trace element (U, Th, Zr, Hf, Nb, Ta, REEs) compositions. The large garnet 28 population studied (496 individual grains) is dominated by lherzolitic G9 (38%) and 'megacrystic' 29 G1 (41%) compositions, with minor contributions from Ti-metasomatized G11 (7%) and eclogitic 30 G3 (6%) cratonic mantle sources. Harzburgitic G10 garnet is very rare (two grains only), consistent 31 with a lherzolite-dominated CLM section in a craton margin position. The eclogitic garnet 32 population has compositions akin to garnet from high-Mg cratonic mantle eclogite xenoliths, and 33 such compositions have recently been interpreted as metasomatic in origin within the mantle 34 xenoliths literature.

35 Pressure-temperature calculations using the single-grain clinopyroxene technique reveal a 36 relatively cold cratonic geotherm of  $37-38 \text{ mW/m}^2$  for the study region during the Late Mesozoic. 37 For peridotitic garnets, projections of calculated Ni-in-garnet temperatures onto the independently 38 constrained regional conductive geotherm suggest that lherzolite dominates at <145 km depths, 39 whereas high-Ti lherzolitic G11 garnets and 'megacrystic' G1 garnets originate mostly from 40 greater depths, down to the lithosphere base at 150 to 210 km depth. The apparent confinement of 41 'megacrystic' G1 garnet to the bottom of the lithosphere suggests formation from infiltrating 42 asthenosphere-derived proto-kimberlite liquids during melt-rock interactions. In general, the data 43 suggest that the CLM beneath NW Botswana is depleted to about 145 km depth, and between 145-44 210 km depths a thick metasomatized layer is identified, representing the transition into the

underlying asthenosphere. A relatively thin lithosphere beneath NW Botswana is consistent with
the proposed craton margin setting, especially when compared to the thicker cratonic roots beneath
the central regions of the Congo and Kalahari cratons in Angola and South Africa, respectively,
reaching down to 250 km depth and possibly even deeper.

49 The compositional dissimilarity between the deepest-derived garnets from kimberlites in 50 NW Botswana (i.e., from the diamond stability field) and garnets that occur as inclusions in 51 diamond from cratons worldwide suggests extensive overprinting of the lowermost cratonic 52 lithosphere by oxidative melt-related metasomatism. This finding, together with the very low 53 diamond grades of the Xaudum kimberlites, points to a diminished diamond potential of the large 54 and mostly unexposed 'cratonic' region (e.g., covered by thick desert sand) located between the 55 major diamond mining districts of the Congo craton to the north (e.g., Catoca) and the Kalahari 56 craton to the south (e.g., Orapa and Jwaneng).

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*Keywords:* Congo craton, continental mantle lithosphere, garnet and clinopyroxene xenocrysts,
cratonic geotherm, Cretaceous kimberlites, thermobarometry, asthenospherization

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#### 64 **INTRODUCTION**

Although kimberlites represent only small volumes of mantle-derived magma, they play an
essential role in our understanding of the nature and origin of the continental lithospheric mantle
(CLM) because they carry materials from otherwise inaccessible parts of Earth's interior. The

68 abundance of xenoliths and xenocrysts in kimberlites and related rocks is strongly biased, with 69 xenolith-rich and xenolith-poor occurrences typically existing within a single volcanic field or 70 cluster (O'Reilly and Griffin, 2006), also mirrored by the skewed diamond grades of individual 71 magmatic bodies within a given kimberlite field (Tappe et al., 2018a). Significant sampling bias 72 is also known from the discrete magmatic pulses that led to different kimberlite units within a 73 single diatreme structure (Moss et al., 2018). For most xenolith-deficient kimberlites, mantle-74 derived xenocrysts can be used as a proxy for mantle rocks to determine their compositions and pressure-temperature evolution (e.g., Griffin et al., 2004; Grütter et al., 2004). Using this 75 76 approach, a large dataset for mantle-derived xenocrysts and micro-xenoliths from cratons 77 worldwide has been produced and improved our understanding of ancient CLM and its diamond 78 potential (e.g., Schulze, 1989; Griffin and Ryan, 1995; Griffin et al., 2004; Ashchepkov et al., 79 2012; Hunt et al., 2012; Nimis et al., 2020).

80 The  $84 \pm 4$  Ma Nxau Nxau kimberlites in NW Botswana are located at the southern margin 81 of the Congo craton near the Damara mobile belt (Farr et al., 2018; Tappe et al., 2020), which 82 formed during Pan-African crustal reworking as part of the Gondwana assembly between 600 and 83 500 Ma (Jelsma et al., 2018). However, the present-day position of the southern craton boundary 84 has remained elusive. The 31 known diatremes and sills of the Nxau Nxau kimberlite cluster 85 contain only small amounts of mantle-derived xenoliths, which are mostly altered (Farr et al., 86 2018). Diamond exploration activity between 2010 and 2015 produced significant quantities of 87 mantle-derived xenocrysts from Nxau Nxau kimberlite drill cores, including garnet and 88 clinopyroxene, which are the subject of this study.

89 We report a large major and trace element dataset for mantle-derived garnet and 90 clinopyroxene xenocrysts (~550 grains) to obtain first insights into the lithospheric mantle 91 architecture beneath this poorly explored cratonic region of southern Africa. Our investigation of 92 the deeper lithosphere beneath the Nxau Nxau kimberlite cluster includes an assessment of the 93 compositional and thermal evolution of the regional CLM, including its diamond potential. The 94 results are compared with the petrological information available for key mantle sections on the 95 Congo craton to the northwest and the Kalahari craton to the southeast. The findings reported 96 herein provide essential pieces in the puzzle of the cratonic assembly of former Gondwanaland in 97 sub-Saharan Africa, because it is shown for the first time that the mantle lithosphere beneath NW 98 Botswana has a cratonic origin, albeit strongly overprinted during tectonomagmatic processes in 99 the 'buffer zone' between the Congo and Kalahari cratons.

100

## 101 GEOLOGICAL SETTING

## 102 Congo craton

The Congo craton (also known as 'Congo Shield', 'Central African Shield', 'Kasai Shield' and 'Angola–Kasai Craton') comprises vast regions in central and southwest Africa. In the south, it is separated from the Kalahari craton (including the Zimbabwe and Kaapvaal cratons plus the intervening Limpopo belt) by the Late Proterozoic Damara orogenic belt, and in the east from the Tanzania craton by the Early Proterozoic Kibaran orogenic belt (Fig. 1). Other orogenic belts flanking the Congo craton include the Lufilian Arc in the southeast, the Kaoko and West Congolian belts in the west, and the Central African fold belt in the north (e.g., Key and Ayres, 2000).

The Archean crust of the Congo craton (3.4–2.6 Ga) comprises granulites, gneisses, granites and amphibolites, and some gabbro–charnockite and migmatite complexes (e.g., Walraven and Rumvegeri, 1993). A large portion of the Congo craton is covered by Phanerozoic strata, with metamorphic and igneous basement rocks being exposed principally at four craton

114 edges, known as the Angolan, Kasai, Mbomou and Ntem blocks (Jelsma et al., 2018). Additional 115 information about the architecture of the Congo craton and its margins can be obtained from the 116 study of kimberlite-borne xenoliths and xenocrysts, with three principal study regions: (1) the 117 Kasai block (Mbuji Mayi – Tshibwe kimberlite clusters), (2) the central Angola block (Catoca and 118 Lunda kimberlite clusters), and (3) the Bangweulu block (Kundelungu kimberlite and Kapamba 119 lamproite clusters) (Batumike et al., 2009; Ashchepkov et al., 2012; Robles-Cruz et al., 2012; 120 Nikitina et al., 2014; Kosman et al., 2016; Korolev et al., 2021; Ngwenya and Tappe, 2021; Tappe 121 et al., 2023). These relatively few studies provide the current knowledge base for our 122 understanding of the nature and evolution of the CLM beneath the Congo craton and its margins.

123 Demarcation of the southern boundary of the Congo craton is especially complicated 124 because thick sedimentary cover sequences, including Damara-age thrust sheets, conceal this 125 portion of the craton in southern Angola and northern Namibia and Botswana (e.g., Key and Ayers, 126 2000). Although geophysical (aeromagnetic and gravity) and borehole data suggest that the 127 southern margin of the Congo craton is located in NW Botswana (Khoza et al., 2013), supporting 128 evidence from the petrology of lithospheric mantle-derived rocks and minerals from beneath this 129 key region is lacking thus far. We obtained such insightful materials from several small kimberlite 130 bodies of the Nxau Nxau volcanic cluster in NW Botswana, and they were investigated to further 131 constrain the elusive southern margin of the Congo craton.

132

## 133 The Nxau Nxau kimberlites of the Xaudum province

134 The Xaudum kimberlite province is located on the inferred southernmost extent of the Congo 135 craton. It comprises four kimberlite clusters, namely Sikereti, Gura, Kaudom and Nxau Nxau (de 136 Wit, 2013) (Fig. 1). Whereas the Nxau Nxau kimberlites are located in Botswana, the other three nearby clusters occur in Namibia. The Nxau Nxau cluster contains at least 31 discrete kimberlite
bodies out of the 45 known magmatic bodies for the entire Xaudum kimberlite province (Fig. 2).
The Xaudum kimberlite province is generally poorly explored by comparison to other kimberlite
fields in southern and central Africa (de Wit et al., 2016).

141 In the Nxau Nxau cluster, kimberlite magmas were emplaced in the form of maar-diatremes 142 (i.e., volcanic pipes), dykes and sills. They intruded the Phanerozoic sedimentary rocks and 143 dolerites of the Karoo Supergroup (Smith, 1984), the Late Proterozoic sedimentary rock sequence of the Damara Supergroup, and Late Archean basement granitoids (Batumike et al., 2009). The 144 145 emplacement age of the Nxau Nxau kimberlites (ca. 84 Ma; Farr et al., 2018) overlaps with the 146 prominent Late Cretaceous peak of kimberlite magmatic activity across southern Africa and Brazil 147 (Griffin et al., 2014; Tappe et al., 2018b), probably in response to plate reorganization during the 148 late phase of West Gondwana breakup. The Nxau Nxau kimberlites typically consist of olivine 149 macrocrysts (mostly altered) set in a fine-grained groundmass of olivine, spinel, perovskite, 150 apatite, phlogopite, calcite and serpentine (Farr et al., 2018), i.e., a typical hypabyssal Group-1 151 kimberlite mineral assemblage and texture (Mitchell, 2008). In agreement with the petrography, 152 the major and trace element compositions and Sr-Nd-Hf isotopic signatures of the Nxau Nxau 153 kimberlites reflect a typical Group-1 kimberlite petrogenesis, and in the absence of anomalous <sup>182</sup>W an asthenospheric upper mantle source for this volcanism appears most likely (Tappe et al., 154 155 2020).

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#### 157 SAMPLES AND ANALYTICAL TECHNIQUES

Garnet (496 grains) and clinopyroxene (83 grains) xenocrysts recovered from heavy mineral
concentrates of the K20 and K21 kimberlite pipes in the Nxau Nxau cluster were mounted in epoxy

160 resin and polished for *in-situ* major and trace element analysis at the University of Johannesburg. 161 The major and minor element compositions were measured using a CAMECA SX100 electron 162 microprobe (EPMA), equipped with four WDS spectrometers. The measurements were performed 163 with a beam current of 20 nA and an accelerating voltage of 20 kV at an electron beam size of 1 164 μm. Trace element concentrations were measured for 443 garnet grains and for a selection of 59 165 clinopyroxene xenocrysts by LA-ICP-MS using a Thermo Scientific iCAP RQ mass spectrometer 166 attached to a 193 nm ArF RESOlution SE155 excimer laser. The NIST612 glass was the primary calibration standard, and <sup>29</sup>Si was used for internal standardization. Two in-house matrix-matched 167 168 standards (GHR1 garnet and KBY17 clinopyroxene; Tappe et al., 2021) and several USGS basaltic 169 glasses (BCR2G, BHVO2, BIR1G) were analyzed repeatedly alongside our samples to monitor 170 data quality. A slightly higher variance of up to 30% was observed between the recommended 171 values for KBY17 clinopyroxene and the trace element concentrations determined for this material 172 in this study. However, the concentrations of most elements in GHR1 garnet and USGS basaltic 173 glasses are in excellent agreement (i.e., <15% variability) with the recommended values (Supp. 174 Table 1). Trace element data for clinopyroxene xenocrysts with  $>1 \mu g/g$  Ba are rejected because 175 of a possible imprint from the trace element enriched host kimberlite melt (Shaikh et al., 2020). 176 Full details about the analytical techniques are provided in Appendix 1, and the complete dataset 177 is given in Supp. Table 1.

178

179 **RESULTS** 

180 Major and trace element compositions

181 *Clinopyroxene* 

182 Clinopyroxene xenocrysts collected for this study are mostly diopside (Wo41.1-49.9En46.5-53Fs2.7-8) 183 with variable Cr<sub>2</sub>O<sub>3</sub> (0.03–3.48 wt.%) and Al<sub>2</sub>O<sub>3</sub> (0.30–5.87 wt.%) contents and a wide range of  $Mg\# (=Mg/(Mg+Fe^{2+}))$  (0.86–0.95) (Fig. 3a–c; Supp. Table 1). Among these xenocrysts, Cr-rich 184 185 clinopyroxenes (1.03-2.85 wt.% Cr<sub>2</sub>O<sub>3</sub>) that are eligible for P-T calculations (see the 186 thermobarometry section) have moderate Na<sub>2</sub>O (1.06-2.98 wt.%) and Al<sub>2</sub>O<sub>3</sub> (1.23-3.97 wt.%) 187 contents. The trace element concentrations show a wide range:  $Zr (1-144 \mu g/g)$ ,  $Y (0.1-8.9 \mu g/g)$ , 188 Hf (0.1–6.8  $\mu$ g/g) and Ce (6.2–40.1  $\mu$ g/g) (Supp. Table 1). The normalized extended trace element 189 patterns (using the primitive mantle values recommended by Palme and O'Neill, 2003) for 190 clinopyroxenes have a humped shape with negative anomalies at Nb-Ta, Zr, Pb, Ti, and Y with 191 respect to REEs (Fig. 4a, b). In contrast, the clinopyroxene xenocrysts that were excluded from P-192 T calculations because of their low Cr contents show a wide range of incompatible trace element 193 concentrations, with humped trace element patterns exhibiting positive Zr-Hf anomalies (Fig. 4a, 194 b).

195

196 Garnet

197 Garnets analyzed in this study (n=496) show a wide range of compositions with Mg# between 0.68-0.86 and Ca# (= Ca/(Ca+Mg+Fe<sup>2+</sup>+Mn)) between 0.01-0.30 (Supp. Table 1). The wide range 198 199 of compositions is also reflected in the G-type classification scheme of Grütter et al. (2004), which 200 indicates the presence of at least eight different garnet types (Fig. 5a). The garnet assemblage 201 studied is dominated by megacrystic G1 (41%) and lherzolitic G9 (38%) types, along with a minor 202 proportion of high-Ti lherzolitic (metasomatized peridotite) G11 garnets (7%), eclogitic G3 203 garnets (6%), high-Cr pyroxenite garnets (G5) and pyroxenite/low-Ca eclogite garnets (G4) (3%), 204 as well as unclassified G0 garnets (4%) (Fig. 5b). Only three garnet grains were identified as

wehrlitic G12 and two grains as harzburgitic G10 varieties, which amounts to <2% of the total</li>
garnet xenocryst population studied.

207 Lherzolitic G9 garnets are relatively depleted in Zr (mostly  $<50 \mu g/g$ ) and Y (mostly 208 between 0–20 µg/g) compared to megacrystic G1 and Ti-metasomatized G11 garnets (mostly 209 between 20–80 µg/g Zr, 10–25 µg/g Y) (Supp. Table 1). Megacrystic G1 and Ti-metasomatized 210 G11 garnets also show elevated Zr/Y ratios. Similarly, Ni concentrations in lherzolitic G9 garnets 211 are lower (<50 µg/g Ni) than in megacrystic G1 and Ti-metasomatized G11 garnets (mostly 40– 212  $100 \,\mu g/g$  Ni). Besides these similarities in trace element concentrations between megacrystic and 213 metasomatized garnets, some subtle differences were observed. For example, the positive Ti-214 anomaly is observed only in megacrystic G1 and a few lherzolitic G9 garnets. Also, the garnet 215 'megacrysts' show relatively strong enrichment in Zr-Hf and lack the positive Sc anomaly (relative 216 to the HREE and V) compared to their Ti-metasomatized garnet counterparts (Fig. 7a, d).

217 Nearly half of the lherzolitic G9 garnets and all of the Ti-metasomatized G11 garnets, along 218 with the megacrystic G1 and harzburgitic G10 garnets, show 'normal' REE<sub>N</sub> patterns with strongly 219 depleted LREEs relative to the flat MREEs and HREEs (Fig. 6a-d). The remaining lherzolitic G9 220 garnets plus wehrlitic G12 garnets have 'sinusoidal' REE<sub>N</sub> patterns with a weaker depletion in 221 LREEs in addition to enriched Nd-Sm-Eu and progressive depletion from Gd-Tb-Dy to Dy-Ho-Er 222 (Fig. 6c). Overall, the REE<sub>N</sub> patterns (both normal and sinusoidal) and extended trace element 223 patterns of the lherzolitic G9 garnets exhibit a wide range of shapes (Fig. 6c, d), indicating 224 gradational changes in the compositions of these garnets. In contrast, the REE<sub>N</sub> and extended trace 225 element patterns of the other garnet types exhibit less scatter compared to the G9 garnets. In 226 extended trace element diagrams, all garnet types show prominent negative Sr, La and Th 227 anomalies and positive U anomalies (Fig. 7a-d). Positive Nb, Ta, Zr and Hf anomalies are also

common. Extended trace element diagrams of wehrlitic G12 garnets and a few lherzolitic G9 garnets are marked by depletion in Zr, Hf, Ti, Y and the MREEs to LREEs compared to other garnet types (Fig. 7c–d). Eclogitic G3, pyroxenitic/low-Ca eclogitic G4 and 'unclassified' G0 garnets show 'normal' REE<sub>N</sub> patterns and negative Th, La, Sr and Sm anomalies in extended trace element diagrams (Fig. 8a–f). Their REE<sub>N</sub> and extended trace element patterns are very similar to those of the megacrystic G1 garnets (Fig. 7, 8).

234

## 235 Thermobarometry

#### 236 *Clinopyroxene*

We applied the single-pyroxene thermobarometer of Nimis and Taylor (2000) to our samples, but out of 84 individual clinopyroxene xenocrysts, only 27 passed the compositional filter for suitability implemented by Nimis and Grütter (2010) and refined further by Ziberna et al. (2016). After additional pressure corrections on clinopyroxene grains that had equilibrated at >50 kbar (Nimis et al., 2020), our dataset yielded a range of temperatures and pressures between 525– 1100°C and 20–52 kbar (Fig. 9), which is consistent with a conductive geotherm of 38 mW/m<sup>2</sup> using the Hasterok and Chapman (2011) model.

Recently, Sudholz et al. (2021a) recalibrated the Cr-in-clinopyroxene barometer of Nimis and Taylor (2000) over an extended pressure range of up to 70 kbar. We calculated equilibrium pressures for our clinopyroxene xenocrysts applying this new calibration, and we obtained pressures in the range of 25 to 57 kbar, i.e., 3 to 4 kbar higher pressures relative to the original calibration. Data treatment using the Sudholz et al. (2021a) calibration yielded a slightly colder geotherm of 37 mW/m<sup>2</sup> compared with 38 mW/m<sup>2</sup> using the Nimis and Taylor (2000) method. Given that the difference between calibrations is very minor, we will work with the 38 mW/m<sup>2</sup> geotherm to enable more robust comparisons with the many literature datasets that used the
original Cr-in-clinopyroxene barometer (e.g., Ashchepkov et al., 2012; Hunt et al., 2012; Ziberna
et al., 2016; Tappe et al., 2021; Nkere et al., 2021; Shaikh et al., 2020).

254

255 Garnet

256 We calculated Ni-in-garnet temperatures  $(T_{Ni})$  for 386 peridotitic garnets using three 257 different thermometers (Ryan et al., 1996; Canil, 1999; Sudholz et al., 2021b). The calibrations of 258 Ryan et al. (1996) and Sudholz et al. (2021b) tend to give a broader range of temperatures than the 259 calibration by Canil (1999), with high-temperature outliers beyond the mantle adiabat that we 260 consider unrealistic (Supp. Fig. 1). Furthermore, the Sudholz et al. (2021b) calibration tends to 261 give higher temperatures for garnet grains with elevated Cr#, often resulting in a similar range of 262 temperatures for different Ni contents (Supp. Fig. 2). In contrast, the algorithm by Canil (1999) 263 does not produce such high-temperature outliers, and the results appear to be geologically 264 meaningful (Shaikh et al., 2020). Therefore, we only discuss T<sub>Ni</sub> results obtained with the Canil 265 (1999) method. This Ni-based thermometer gave a temperature range of 793-1329 °C for our garnet xenocrysts. The frequency distribution of T<sub>Ni</sub> shows two prominent peaks at around 975 °C 266 267 and 1100 °C (Fig. 10a). Whereas Iherzolitic (G9) garnets dominate the low-T peak, 'megacrystic' 268 (G1) and Ti-metasomatized (G11) garnets are more prominent at the high-T peak. Our T<sub>Ni</sub> results 269 have been projected onto the clinopyroxene-derived 38 mW/m<sup>2</sup> Nxau Nxau geotherm (Fig. 10b), 270 using the method discussed in Shaikh et al. (2019). The T<sub>Ni</sub> projections suggest that these garnets 271 were sampled from the lithospheric mantle by ascending kimberlite magmas between 210 and 105 272 km depths, with a possible thermal lithosphere-asthenosphere boundary at around 210 km depth 273 (based on the intersection between the conductive geotherm and the mantle adiabat). However,

274 geochemical proxies such as Y- and Ti-depletion in garnet (Griffin and Ryan, 1995) suggest that 275 the base of the lithospheric mantle (i.e., the geochemical LAB) is located at shallower depth of 276 around 145 km (Fig. 11a, d), which is consistent with geophysical data that cover the study region 277 (e.g., Evans et al., 2019; White-Gaynor et al., 2020). A cratonic geotherm is assumed to be close 278 to the conductive model up to the temperatures calculated for the lithosphere base, however, it may 279 show a kinked nature at higher temperatures (Finnerty and Boyd, 1987; Griffin et al., 2003). This 280 assumption may cause an underestimation of the depths of origin for the high-temperature garnets, 281 but this minor caveat does not alter the here developed model for the CLM beneath the 282 southernmost portion of the Congo craton in a significant way.

283 Data treatment shows that all Ti-metasomatized G11 garnets and most of the 'megacrystic' 284 G1 garnets were likely sampled from the deepest levels of the CLM (Fig. 10a, b). The accuracy of 285 our temperature estimates for the 'megacrystic' garnet population relies on the assumption that 286 these grains equilibrated with ordinary mantle olivine containing approximately 3000  $\mu$ g/g Ni 287 (Canil, 1999). Because all of the olivine macrocrysts in the Nxau Nxau kimberlites studied are 288 altered (Farr et al., 2018), it is impossible to test a megacrystic origin using mineral compositions 289 (e.g., Moore and Costin, 2016). However, olivine is an integral part of the discrete low-Cr 290 megacryst suite in kimberlites (Nixon and Boyd, 1973) so that the assumption of coexistence 291 between garnet and olivine (among other phases) in the megacryst source is well founded. We note 292 further that genetic links between low-Cr megacrysts and sheared peridotites from the lowermost 293 CLM have been suggested, and in these plastically deformed peridotites ultra-coarse garnet clasts 294 and other megacryst phases are typically embedded in an olivine-dominated matrix containing 295 between 3000 and 4000 µg/g Ni (e.g., Tappe et al., 2021). Nkere et al. (2021) showed that the Nibased garnet thermometers can be applied to megacrysts, because independent constraints yieldedsimilar equilibration temperatures.

298 Our dataset reveals some prominent depth-specific compositional features. For example, 299 there is a higher variance in the TiO<sub>2</sub> and Zr concentrations, with more garnets having higher Mg# 300 in the deeper CLM portions compared to the shallower mantle (Fig. 11a, b, c). In the  $(Nd/Dy)_N$ 301 versus (Nd/Ce)<sub>N</sub> diagram (Supp. Fig. 3a), the 'megacrystic' G1, Ti-metasomatized G11 and 302 harzburgitic G10 garnets fall into the NW-quadrant with high (Nd/Ce)<sub>N</sub> and low (Nd/Dy)<sub>N</sub>, which 303 is consistent with their strongly LREE depleted 'normal' REE patterns. Lherzolitic G9 garnets show 304 a wide compositional range in the (Nd/Dy)<sub>N</sub> versus (Nd/Ce)<sub>N</sub> diagram consistent with their 'normal' 305 and 'sinusoidal' REE<sub>N</sub> patterns. When projected down to depth, there is a substantial increase in 306 (Nd/Dy)<sub>N</sub> at ~140 km with low to moderate (Nd/Ce)<sub>N</sub> ratios (Supp. Fig. 3b, c). This indicates that 307 garnets from this depth exhibit the strongest sinuosity in their REE patterns.

308

#### 309 **DISCUSSION**

## 310 Architecture and thermal state of the lithospheric mantle beneath NW Botswana

311 Clinopyroxene and garnet xenocrysts studied here define a relatively cold conductive geotherm of 312  $\sim$ 38 mW/m<sup>2</sup> for the CLM beneath NW Botswana, with a lithospheric thickness of  $\sim$ 145 km (Fig. 313 9; 10b). Our data indicate some distinct lithological and compositional variations within the 314 studied lithospheric mantle section. HREE depletion in clinopyroxene xenocrysts indicates their 315 derivation from a garnet-bearing lithology (Fig. 4a). Most garnet grains studied were derived from 316 lherzolites (G9, 38%) and metasomatized lherzolites (G11, 41%), which testifies to a relatively 317 fertile nature of the CLM beneath NW Botswana. The reconstructed vertical distribution of garnet 318 xenocrysts shows compositionally distinctive CLM layers, with a shallow less metasomatized layer (<145 km depth) dominated by lherzolites, and a deeper more extensively metasomatized layer (145–210 km depths) dominated by metasomatized lherzolites and megacrystic garnets (Fig. 10a, b). The shallow CLM layer also contains rare harzburgites and wehrlites, two lithologies that are seemingly absent from the deeper layer, implying an original difference between CLM layers, or lack of sampling, or metasomatic overprinting. An increasing metasomatic overprint with depth is supported by the observed increase in metasomatic alteration of the rare harzburgite and wehrlite derived crystals from within the shallow CLM layer.

326 The extensive metasomatism of the deeper layer, possibly representing the lithosphere-327 asthenosphere transition, is evident from the larger variance in TiO<sub>2</sub> contents of garnets with 328 increasing depth (Fig. 11a). This observation is consistent with bottom-up refertilization of the 329 CLM noticed for many cratons worldwide (e.g., Griffin and Ryan, 1995; Griffin et al., 1999a; Hunt 330 et al., 2012; Ziberna et al., 2013; Aulbach et al., 2017a; Shaikh et al., 2020; de Freitas Rodrigues 331 et al., 2023). Garnet Mg# is increasing with depth (Fig. 11b), which may be a function of increasing 332 temperature (O'Neill and Wood, 1979), or an increasing degree of depletion in basaltic melt 333 component over the course of lithosphere evolution (e.g., Lehtonen and O'Brien, 2009). The latter 334 option is less likely because these high-Mg# garnets near the lithosphere base show trace element 335 evidence for melt/fluid related fertilization. Furthermore, there is no clear trend in the garnet Cr# 336 versus depth systematics (Supp. Fig. 4a), which argues against compositionally induced changes 337 of garnet Mg#. A pronounced metasomatic character of the lower CLM layer is also evident from 338 the V/Sc, Zr/Y and Ti/Eu systematics of garnet (Fig. 12b, c, d; Supp. Fig. 4b), with an apparent 339 increase in these trace element ratios with depth. This is also consistent with the common 340 occurrence of peridotitic garnet with 'normal' REE<sub>N</sub> patterns in the lower layer compared to the 341 upper CLM layer (Supp. Fig. 3b, c). It is also important to note that many garnet xenocrysts from

342 the lower layer show transitional compositions between 'normal' and 'sinusoidal' end-member REE 343 patterns (Fig. 6a-d), suggesting gradational trace element enrichment through the interaction of 344 continuously evolving melt with the CLM wall rocks (e.g., Ziberna et al., 2013), or increasing 345 levels of interaction between mantle peridotite and infiltrating melt. The nearly constant Mg# of 346 'megacrystic' G1 garnets suggests that the metasomatic melt was buffered with respect to its Fe-347 Mg content. The apparent confinement of 'megacrystic' G1 garnets to the lower CLM layer (Fig. 348 10a, b) stands in contrast to the distribution of megacrysts in other cratonic mantle sections 349 worldwide. For example, in the Slave (Kopylova et al., 2009) and Eastern Dharwar cratons (Shaikh 350 et al., 2020), 'megacrystic' G1 garnets and other members of the megacryst suite appear to be 351 dispersed throughout the CLM column, suggesting their crystallization from kimberlitic precursor 352 melts at various depth levels (e.g., Shaikh et al., 2021). Unlike these examples, the relatively 353 restricted distribution of 'megacrystic' G1 garnets beneath NW Botswana may be explained by 354 their formation as part of stockwork-like bodies of proto-kimberlite melt that stalled at the 355 lithosphere base (Nkere et al., 2021). The strongly metasomatized and megacryst-rich lower CLM 356 layer identified beneath NW Botswana resembles what has been recently described as a thick 357 lithosphere-asthenosphere transition zone beneath the central Kaapvaal craton, a layer that is 358 dominated by deformed fertile lherzolites and associated megacrysts (Tappe et al., 2021). 359 Similarly, the fertilized deeper layer between 145 and 210 km depth beneath NW Botswana may 360 represent a lithosphere-asthenosphere transition zone at the southern margin of the Congo craton. 361 An alternative explanation for the relatively high temperatures recorded by the majority of 362 'megacrystic' G1 garnets (and the Ti-metasomatized G11 garnets) could be heating during 363 interactions with 'hot' metasomatic melt, as has been suggested to explain the abundance of Ti-364 rich CLM lithologies beneath the Chidliak kimberlite field in NE Canada (Kopylova et al., 2019).

365 Such high-temperature mantle xenoliths appear to be common in craton margin positions and have 366 also been reported from off-craton kimberlites along the Proterozoic mobile belts that border 367 against the western and southern Kaapvaal craton margins (Janney et al., 2010). In the context of 368 the southern margin of the Congo craton examined here, the deeper and hotter garnets could also 369 stem from underthrust CLM portions of the Damara orogen. A similar notion was put forward by 370 Griffin et al. (2009) for high-temperature garnets recovered from kimberlites in southern India, 371 speculating on the presence of a large-scale tectonic suture zone at mantle depth. However, mantle-372 derived crystal cargo from many more kimberlite localities, spread along and across the southern 373 Congo craton margin, would need to be studied to substantiate this idea in the African context.

374 The trace element concentrations of hypothetical melts in equilibrium with garnet and 375 clinopyroxene xenocrysts were calculated using the partition coefficients for carbonated silicate 376 melts at deep upper mantle conditions between 6 and 12 GPa (Girnis et al., 2013). The 377 reconstructed trace element compositions were compared with those of the bulk hypabyssal 378 kimberlites from Nxau Nxau as reported in Tappe et al. (2020). In general, we note similarities 379 between the bulk kimberlite and metasomatized garnet trace element compositions. However, the 380 strong depletions in Th, U and La for the melts that crystallized the megacrystic G1 and Ti-381 metasomatized G11 garnets at depth, relative to bulk kimberlite at surface, suggest fractionation 382 of these elements by other common megacryst phases such as clinopyroxene and ilmenite, or even 383 zircon (Fig. 13). Overall, the trace element modelling results for garnet are consistent with the 384 notion of megacryst formation through the interaction of carbonate-rich proto-kimberlite melts 385 with cratonic mantle lithosphere (Kopylova et al., 2009; Tappe et al., 2021). In contrast, the trace 386 element patterns of the bulk kimberlites and the inverted melts using clinopyroxene xenocrysts do not match, which suggests a lack of a direct genetic relationship or distinctly different evolutionary
paths of the kimberlitic liquids involved.

389

## **Diamond prospectivity at the southern margin of the Congo craton**

391 Whereas the eclogitic diamond potential of kimberlites cannot be assessed with reasonable 392 confidence using xenocrystic garnet compositions, it is common practice to utilize such 393 mineralogical data to evaluate the peridotitic diamond potential of kimberlite bodies and clusters 394 (e.g., Gurney and Moore, 1993). Our data suggest that the CLM beneath NW Botswana exhibits a 395 suitable thickness and thermal state that would promote diamond formation and preservation 396 within a thin interval between ~120 and 145 km depth (i.e., 25 km thick; Fig. 10a, b). This narrow 397 diamond window is significantly thinner than beneath major diamond mining districts in sub-398 Saharan Africa, from where >50 km thick diamond windows have been reported (e.g., Griffin et 399 al., 1999a, 2004; Ashchepkov et al., 2012; Nimis et al., 2020; Tappe et al., 2021).

400 The majority of peridotitic diamonds in a global database are linked to harzburgites and 401 depleted lherzolites (Stachel et al., 2004; Stachel and Harris, 2008); however, at Nxau Nxau the 402 major and trace element compositions of the most deeply derived garnet xenocrysts indicate that 403 harzburgitic and depleted lherzolitic potential diamond source rocks are absent or scant below 145 404 km depth (Fig. 14a-c). The deeper garnet populations are dominated by Ti-metasomatized G11 405 and 'megacrystic' G1 compositions, which fall outside the compositional field for garnets found 406 as inclusions in diamonds from cratons worldwide (Stachel and Harris, 2008). Mineral 407 compositions dissimilar from those of inclusions in diamond, as recorded in the global database, 408 suggest a low diamond prospectivity in NW Botswana. This is consistent with first previous

409 exploration results that reported a lack of macro-diamonds and a general paucity of micro-410 diamonds for the Xaudum kimberlite province (de Wit et al., 2017).

411 The diamond potential of continental shields may be affected by poor diamond preservation 412 in their mantle source rocks, including the complete destruction of the diamond endowment, 413 possibly triggered by heating, decompression and oxidation (e.g., Stagno et al., 2015; Howarth et 414 al., 2023). In addition, kimberlites and related carbonated magma types are known to be able to 415 resorb diamond (Fedortchouk et al., 2019). For example, it has been argued that diamonds beneath 416 the metacratonic Bangweulu Block in the D.R. Congo (Fig. 1) were destroyed by melt interaction 417 in the mantle source, explaining the barren nature of the ca. 30 Ma Kundelungu kimberlite field 418 (Batumike et al., 2009). Importantly, the same block – inferred to be an extension of the Congo 419 craton – contained lithospheric diamonds at ca. 180 Ma as evident from the diamondiferous nature 420 of Karoo-age lamproites that erupted on its southern margin (Tappe et al., 2023). Also, a barren 421 suite of mantle-derived eclogite xenoliths from the highly diamondiferous Orapa kimberlites in 422 NE Botswana (Kalahari craton) was interpreted to have interacted with proto-kimberlite melt to 423 the extent of complete diamond destruction (Aulbach et al., 2017b; 2020a). For the southern 424 margin of the Congo craton in NW Botswana, our data reveal the extensive nature of mantle 425 metasomatism that overprinted the lower CLM and may have diminished the diamond potential of 426 this poorly exposed cratonic region.

427

## 428 Comparisons of lithospheric mantle sections across sub-Saharan Africa

429 Southernmost (Nxau Nxau) versus central Congo craton (Mbuji Mayi and Catoca)

430 Lithospheric mantle sections from different kimberlite locations on or around cratons provide

431 valuable insights into CLM architecture and thermal evolution (e.g., Griffin et al., 2004; Sand et

432 al., 2009; Ashchepkov et al., 2012; Hunt et al., 2012; Kopylova et al., 2019; Shaikh et al., 2020; 433 de Freitas Rodrigues et al., 2023). Recently, Ozaydin et al. (2021) presented magnetotelluric 434 evidence showing that lithospheric mantle sections reconstructed from kimberlite-borne mantle 435 cargo appear to be representative of CLM architecture. Such a combined geophysical and 436 petrological approach is lending new confidence to the interpretations of lithosphere-scale 437 structures based on mantle xenolith/xenocryst studies alone. However, studies based only on 438 petrology have the additional advantage of exploring craton architecture and thermal state through 439 time.

440 Below, we compare lithospheric mantle sections reported from the more central parts of 441 the Congo craton at Mbuji Mayi (and Tshibwe) in the D.R. Congo and Catoca in Angola with the 442 CLM architecture at the southern margin of the Congo craton in NW Botswana (this study). We 443 note that these reconstructed CLM sections were probed by relatively young kimberlite 444 magmatism during the Cretaceous between ca. 120 and 70 Ma (Robles-Cruz et al., 2012; Farr et 445 al., 2018; Tappe et al., 2018b). Whereas the lithosphere was in excess of 200 km thick beneath the 446 central Congo craton, with extensive metasomatic overprinting at its base (Batumike et al., 2009; 447 Ashchepkov et al., 2012; Kosman et al., 2016; Korolev et al., 2021), our findings from the southern 448 craton margin suggest the presence of a 145 km thick lithosphere, with a thick and heavily 449 overprinted lithosphere-asthenosphere transition zone down to 210 km depth (Fig. 9, 10). The 450 obtained 37-38 mW/m<sup>2</sup> geotherm for the southern margin of the Congo craton is similar to those 451 reported for the locations at the centre of the craton, ranging between 35 and 40  $mW/m^2$ 452 (Ashchepkov et al., 2012; Batumike et al., 2009). It must be noted, however, that Kosman et al. (2016) reported geotherm estimates of >40 mW/m<sup>2</sup> for the Kasai block, obtained by garnet and 453 454 clinopyroxene thermobarometry on inclusions in alluvial diamonds. Given that the geological

455 meaning of such information from alluvial diamonds is far from straightforward, our discussion 456 will focus on more conventional approaches to heat-flux estimations for continental shields. These 457 thermal constraints suggest that the CLM beneath NW Botswana is truly cratonic, albeit 458 compositionally strongly overprinted, possibly due to tectonomagmatic activity during Gondwana 459 assembly and breakup since around 600 Ma. Although metasomatic overprinting is also prominent 460 for the deep mantle root beneath the central regions of the Congo craton (150-240 km depths; 461 Batumike et al., 2009; Ashchepkov et al., 2012), it was apparently less intensive compared to the 462 southern margin, where no harzburgitic components appear to have 'survived' at depth (Fig. 10). 463 Congo craton architecture, as broadly reconstructed here, is consistent with the global model of 464 craton formation and evolution, which identifies thick lithospheric roots beneath the central 465 regions of cratons surrounded by thinning margins that accommodate most of the tectonic stresses 466 and act as pathways to plume-related magmatism (e.g., intensive mantle metasomatism) over the 467 course of Earth history (Shirey et al., 2002; Begg et al., 2009; Pearson et al., 2021). More recent 468 mantle plume activity across sub-Saharan Africa appears to have further complicated the 469 distribution of thick versus thinned and overprinted cratonic mantle lithosphere (e.g., Evans et al., 470 2019; Celli et al., 2020), but these geophysical models cannot be tested using mantle petrology 471 studies because of the extremely rare occurrence of Cenozoic kimberlite magmatism (Tappe et al., 472 2018b).

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## 474 Southern Congo craton margin versus Kalahari craton

The Kalahari craton experienced episodic kimberlite magmatism since at least Early Proterozoic times (e.g., Tappe et al., 2018b), which has allowed petrologists to constrain the evolution of the CLM beneath southern Africa. The highly refractory peridotites of the Kaapvaal mantle

478 lithosphere were formed by extensive Archean melt depletion (e.g., Pearson et al., 1995), with 479 numerous subsequent events of metasomatic enrichment involving different styles and melt/fluid 480 compositions (Griffin et al., 2003; Grégoire et al., 2003; Bell et al., 2005; Simon et al., 2007; 481 Rehfeldt et al., 2008; Kobussen et al., 2009). Nevertheless, as evidenced by the main kimberlite 482 magmatic phase during the Mesozoic between 160 and 80 Ma, the Kaapvaal lithosphere has 483 remained relatively thick (~190–220 km) and slightly warmer (~40 mW/m<sup>2</sup>) (e.g., Kobussen et al., 484 2009; Bell et al., 2003 Muller et al., 2013; Ozavdin et al., 2021) compared to the Nxau Nxau 485 lithospheric mantle section in NW Botswana (~145 km thick lithosphere at 38 mW/m<sup>2</sup>; this study). 486 Three distinct layers have been recognized beneath the Kaapvaal craton: (1) a fertile 487 lherzolite-dominated shallow layer (~90-125 km depth) with high orthopyroxene/olivine ratios 488  $(\sim 0.5)$ , (2) a middle layer dominated by depleted lherzolite and harzburgite (125–170 km depth) 489 with lower orthopyroxene/olivine ratios ( $\sim 0.25$ ), and (3) an extensively metasomatized lower layer 490 at >170 km depth (e.g., Kobussen et al., 2009; Griffin et al., 1999b, 2003; Özaydin et al., 2021). 491 The shallower layer beneath the Kaapvaal craton was strongly affected by potassic metasomatism 492 resulting in phlogopite enrichment (e.g., Fitzpayne et al., 2018) that coincides with a seismically 493 detected mid-lithospheric discontinuity (Smart et al., 2021). Such phlogopite-rich mantle 494 metasomatism is not known from the southern margin of the Congo craton (e.g., Ashchepkov et 495 al., 2012). Instead, a distinctive basaltic melt-related metasomatic style, possibly linked to Karoo-496 age LIP magmatism (i.e., the Okavango mafic dyke swarm), is prevalent beneath the Nxau Nxau 497 kimberlite cluster in NW Botswana (this study). This metasomatized layer beneath the Nxau Nxau 498 kimberlite cluster is thicker than the metasomatic portion of the Kaapvaal lithosphere (~145-210 499 km versus 180-200 km depth). In general, the metasomatic history of the Kaapvaal craton is better 500 studied and thus better understood, and several major magmatic events can be held responsible for 501 reworking of the wider Kalahari CLM; for example, the Bushveld LIP at ca. 2 Ga (Richardson and 502 Shirey, 2008; Zhang et al., 2022), the Umkondo LIP at ca. 1.1 Ga (Koornneef et al., 2017), the 503 Karoo LIP at ca. 180 Ma (Rehfeldt et al., 2008; Burness et al., 2020), and Mesozoic 504 orangeite/lamproite and subsequent kimberlite magmatism (e.g., Giuliani et al., 2015; Tappe et al., 505 2018b). Importantly, the off-craton mantle lithosphere beneath the Proterozoic mobile belts that 506 surround the Kaapvaal craton is relatively thin and notably warmer than the central portion of the 507 craton (e.g., Janney et al., 2010; Muller et al., 2013). An example for this pattern is the Gibeon 508 kimberlite field on the Paleoproterozoic Rehoboth block in Namibia (Fig. 1), where the lithosphere is  $\sim 180$  km thick and characterized by a warm cratonic geotherm of 40–45 mW/m<sup>2</sup> (Bell et al., 509 510 2003; Muller et al., 2013). Furthermore, a variably thinned (as thin as 180 km) and warmer off-511 craton lithosphere has been identified beneath the Mesoproterozoic Namaqua-Natal mobile belt 512 (Janney et al., 2010). Mantle-derived peridotite xenoliths from these off-craton regions are 513 relatively depleted and have experienced at least two stages of melt extraction, followed by 514 variable metasomatic re-enrichment. These off-craton CLM sections also experienced a Mesozoic 515 heating episode possibly related to continental breakup that might have caused up to 30 km of 516 lithosphere thinning due to thermal erosion. Recently, Pearson et al. (2021) extended the boundary 517 of the Kalahari craton to include all Proterozoic domains such as the Rehoboth block as well as 518 the Kheis and Namaqua-Natal belts, because the formation of these mobile belts during the 519 Proterozoic marks the stabilization of thick CLM roots.

The above scenario is different from the southern margin of the Congo craton, where cold cratonic geotherms prevail despite strong metasomatic overprinting (Fig. 9, 10). It is therefore permissible to speculate that the Congo and Kalahari cratons may be contiguous at lithospheric mantle depth (see also the interpretation of magnetotelluric data in Evans et al., 2019), and that 524 Neoproterozoic to Early Paleozoic tectonism in the region (i.e., Gondwana assembly) was largely 525 confined to the upper continental crust (i.e., thin-skinned tectonics). Similar ideas have been 526 expressed for Late Proterozoic tectonic processes that operated along the eastern margins of the 527 Congo craton, where contiguous cratonic mantle lithosphere has been proposed to link the central 528 African cratonic assemblage to the Tanzania craton in the east (Link et al., 2010) and to the 529 Kalahari craton (Zimbabwe cratonic block) in the southeast (Ngwenya and Tappe, 2021). This 530 could potentially mean that a significant part of the Gondwana assembly was shaped by intraplate 531 deformation, with crustal shortening and strike-slip movements hundreds of kilometres away from 532 plate margins and *bona fide* collision zones.

533

# 534 Remarks on non-peridotitic mantle-derived garnets

535 Major and minor element contents of eclogitic G3 and pyroxenitic/low-Ca eclogitic G4 garnets 536 can be used to identify their source rock characteristics. For example, Na<sub>2</sub>O contents of garnets 537 were used to classify kimberlite-borne eclogite xenoliths from Roberts Victor mine in South Africa 538 into Group I (with large-sized rounded garnets that have on average >0.09 wt.% Na<sub>2</sub>O that are set 539 in a clinopyroxene matrix) and Group II (with Na<sub>2</sub>O <0.09 wt.% in garnets interlocking with 540 clinopyroxene) varieties (McCandless and Gurney, 1989). Similarly, garnet Na<sub>2</sub>O contents of 0.04 541 and 0.07 wt.% were arbitrarily taken as cut-off values for diamond-indicator eclogitic garnets by 542 Gurney (1984). Using pyrope-almandine-grossular proportions of garnet, mantle-derived eclogite 543 xenoliths can be divided into Type A, Type B and Type C (Taylor and Neal, 1989). More recently, 544 trace element signatures such as Eu/Eu\* anomalies, LREE<sub>N</sub> depletion patterns, and Sr-Zr-Hf 545 anomalies were included into the classification of eclogites (Jacob et al., 2009; Viljoen et al., 2005; 546 Aulbach and Jacob, 2016; Smart et al., 2017a; Radu et al., 2019). Based on mineral and

reconstructed bulk compositions, Aulbach and Jacob (2016) described eclogites as three main types – eclogites, pyroxenites and gabbroic counterparts. The eclogites were divided further into high-Ca (Ca# for garnet = molar Ca/(Ca + Fe + Mg + Mn) > 0.2), high-Mg (Ca#  $\leq$ 0.2 and Mg# = molar Mg/(Mg + Fe<sub>total</sub>) > 0.6) and low-Mg (Ca#  $\leq$  0.2 and Mg#  $\leq$ 0.6) types by these authors. We apply some of these major and trace element proxies to eclogitic (G3) and pyroxenitic/low-Ca eclogitic (G4) garnets (40 grains) recovered from the Nxau Nxau kimberlites to constrain their protoliths in more detail.

Most of the eclogitic (G3) and pyroxenitic/low-Ca eclogitic (G4) garnet grains are marked by Na<sub>2</sub>O contents below 0.1 wt.% with high Mg# values (>0.6), which is indicative of a high pyrope content typical for Type A bimineralic eclogites (Taylor and Neal, 1989). These authors attributed Type A garnet compositions with high REE contents to fractional crystallization processes. Furthermore, Na<sub>2</sub>O and TiO<sub>2</sub> (0.2–0.6 wt.%) contents of eclogitic (G3) and pyroxenitic/low-Ca eclogitic (G4) garnet from the Nxau Nxau kimberlites are comparable with the Group II eclogites identified by McCandless and Gurney (1989).

561 Such low Na<sub>2</sub>O contents were considered indicative of last equilibration at relatively low 562 pressures (e.g., Sobolev and Lavrent'ev, 1971; Gurney and Moore, 1993). However, Grütter and 563 Quadling (1999) demonstrated that elevated Na in eclogitic garnet is not an ideal indicator of an 564 association with diamond because garnet compositions from many graphite-bearing eclogite 565 xenoliths overlap with those of garnets found as inclusions in diamond. Accordingly, high-Na 566 eclogitic garnets can also occur at pressures lower than those that define the graphite-diamond 567 phase transition. It is possible that high Na<sub>2</sub>O contents (>0.07 wt.%) in eclogitic garnets reflect 568 derivation from diamond-facies eclogites, or eclogite assemblages that equilibrated at graphite-569 facies P-T conditions but with high bulk Na contents, permitting elevated solubility of Na in the

570 garnet structure at relatively low pressures (Grütter and Quadling, 1999). Conversely, those 571 eclogitic garnets with <0.07 wt.% Na<sub>2</sub>O may represent graphite-facies eclogites, or diamond-facies 572 eclogites with low bulk Na contents. Our eclogitic garnet data do not allow a clear assignment to 573 either graphite- or diamond-facies equilibration conditions of their eclogitic source rocks. The 574 eclogitic (G3) and pyroxenitic/low-Ca eclogitic (G4) garnet xenocrysts studied do not show 575 Eu/Eu\* anomalies (Fig. 8a-f), which suggests a non-gabbroic (basaltic) nature of the protolith 576 (Aulbach and Jacob, 2016; Smart et al., 2017b). The high Mg# values coupled with low Ca# values 577 for these garnets suggest that they are derived from disaggregated high-Mg eclogite xenoliths (Fig. 578 15b), commonly interpreted to have metasomatic imprints from their protracted journey through 579 Earth's crust and mantle (De Stefano et al., 2009; Smart et al., 2009; Tappe et al., 2011; Smart et 580 al., 2012; Aulbach et al., 2017b). The eclogitic garnets studied have very similar trace element 581 systematics to those from Mg-rich metasomatized eclogite xenoliths from the Orapa kimberlite 582 field in NE Botswana (Aulbach et al., 2017b) and the Catoca kimberlite cluster in NE Angola 583 (Korolev et al., 2021), with the exception of slightly higher Zr-Hf concentrations compared to 584 peridotitic garnets (Fig. 8d). However, the extended trace element patterns of eclogitic (G3) and 585 pyroxenitic/low-Ca eclogitic (G4) garnets from Nxau Nxau in NW Botswana do not show negative 586 Zr-Hf anomalies relative to neighbouring MREEs (Fig. 8b, d). Very similar trace element patterns 587 are shown by garnets from Type IIA bimineralic eclogites from Roberts Victor (Hardman et al., 588 2001; Radu et al., 2019). We suggest that the subtle Zr-Hf enrichment in garnet relative to the 589 REEs may be indicative of mantle metasomatism, especially when coupled to notably higher Ti 590 contents relative to the global eclogite database (Fig. 15a). This observation has been corroborated 591 frequently by other proxies for metasomatic overprinting of mantle-derived eclogite xenolith suites, such as the presence of hydrous phases and incompatible trace element enrichment inclinopyroxene (e.g., Aulbach et al., 2020b; Smart et al., 2021).

594

## 595 SUMMARY AND CONCLUSIONS

596 This study investigated the major and trace element geochemistry of mantle-derived garnet and 597 clinopyroxene xenocrysts from the Nxau Nxau kimberlites in NW Botswana. The EPMA and LA-598 ICP-MS datasets for these lithospheric mantle materials are the first of their kind for this poorly 599 exposed region at the southern margin of the Congo craton. The following conclusions can be 600 drawn.

601 1. Our data reveal the existence of a relatively thin cratonic lithosphere (~145 km thick) beneath
 602 the southernmost margin of the Congo craton, characterized by a cold conductive geotherm of
 603 ~38 mW/m<sup>2</sup> at ca. 85 Ma.

2. The lithospheric mantle section at the southern margin of the Congo craton is dominated by
depleted lherzolites to around 145 km depth, and this unit is underlain by a ~65 km thick
strongly overprinted zone dominated by metasomatized lherzolites and megacrysts. This layer
possibly represents a lithosphere–asthenosphere transition zone that developed from the bottom
third of once ~200 km thick cratonic lithosphere (i.e., progressive asthenospherization).
Harzburgite appears to be a very rare component at the southern margin of the Congo craton,
probably due to the strong mantle metasomatic imprint.

3. The major and trace element compositions of mantle-derived eclogitic garnet xenocrysts
indicate the presence of a high-Mg eclogite component within the cratonic lithosphere. Such a
lithology provides yet another record of the protracted metasomatic overprinting of the
lithospheric mantle at the southern margin of the Congo craton.

4. The compositional dissimilarity between the deepest-derived garnets from the Nxau Nxau
kimberlites and garnet inclusions in diamond from cratons worldwide, together with the very
low diamond grades of the kimberlites from the Xaudum province explored so far, suggest a
diminished diamond potential of the 'metacratonic' lithosphere at the southern margin of the
Congo craton.

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## 955 FIGURE CAPTIONS

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Fig. 1. Map of sub-Saharan Africa showing the location of the Nxau Nxau kimberlite cluster at the
inferred southern margin of the Congo craton. Other kimberlite localities and cratonic domains
referred to in the main text are shown for orientation (modified after Jelsma et al., 2018).
Geographic coordinates and emplacement ages for the kimberlite clusters shown are summarized
in Tappe et al. (2018b).

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Fig. 2. Location map of the Nxau Nxau kimberlite cluster in NW Botswana. Sample material for
this study was recovered from kimberlite pipes K20 and K21, which are shown with the star
symbol relative to the other unstudied pipes. Modified after Farr et al. (2018).

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Fig. 3. Mg# values of mantle-derived clinopyroxene xenocrysts plotted against their Na<sub>2</sub>O (a),
Al<sub>2</sub>O<sub>3</sub> (b) and Cr<sub>2</sub>O<sub>3</sub> (c) contents. Fields for Kalahari megacrysts are from Nkere et al. (2021) and
Tappe et al. (2021). Clinopyroxenes suitable for P-T calculations have passed the compositional
filters given by Nimis and Grütter (2010) and Ziberna et al. (2016).

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Fig. 4. Primitive-mantle normalized trace element patterns (values for normalization after Palme
and O'Neill, 2003) for mantle-derived clinopyroxene xenocrysts from the K20 and K21 pipes of
the Nxau Nxau kimberlite cluster in NW Botswana at the southern margin of the Congo craton.
Fields for Kalahari clinopyroxene megacrysts are after Nkere et al. (2021) and Tappe et al. (2021).

976 Clinopyroxenes suitable for P-T calculations passed the compositional filters given by Nimis and
977 Grütter (2010) and Ziberna et al. (2016).

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Fig. 5. (a) Cr<sub>2</sub>O<sub>3</sub> (wt.%) versus CaO (wt.%) G-type classification diagram (Grütter et al., 2004) for
mantle-derived garnet xenocrysts from the K20 and K21 pipes of the Nxau Nxau kimberlite cluster
in NW Botswana. The radar plot (b) illustrates the relative frequency of each G-type garnet
population identified at the southern margin of the Congo craton.

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Fig. 6. Chondrite-normalized REE diagrams (normalization after McDonough and Sun, 1995) for
megacrystic G1 (a), Ti-metasomatized G11 plus harzburgitic G10 and wehrlitic G12 (b),
lherzolitic G9 'sinusoidal' (c) and lherzolitic G9 'normal' garnet xenocrysts from the Nxau Nxau
kimberlite cluster in NW Botswana.

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Fig. 7. Primitive-mantle-normalized trace element patterns (normalization after Palme and
O'Neill, 2003) for megacrystic G1 (a), lherzolitic G9 (b), harzburgitic G10 and wehrlitic G12 (c),
and Ti-metasomatized G11 garnets (d) from the Nxau Nxau kimberlite cluster in NW Botswana.

Fig. 8. Chondrite-normalized REE diagrams (normalization after McDonough and Sun, 1995) and
primitive-mantle-normalized extended trace element patterns (normalization after Palme and
O'Neill, 2003) for 'unclassified' G0, eclogitic G3 and pyroxenitic/low-Ca eclogitic G4 garnets
from the Nxau Nxau kimberlite cluster in NW Botswana.

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Fig. 9. Pressure-temperature plot for mantle-derived clinopyroxene xenocrysts from the Nxau
Nxau kimberlites in NW Botswana. Conductive model geotherms (35 and 38 mW/m<sup>2</sup>) are after

1000 Hasterok and Chapman (2011), and the diamond–graphite transition curve is after Day (2012).

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1002 Fig. 10. (a) Frequency distribution of Ni-in-garnet temperatures (T<sub>Ni</sub>) (after Canil, 1999) obtained

1004 Nxau kimberlites in NW Botswana. (b) Projection of  $T_{Ni}$  onto a 38 mW/m<sup>2</sup> geotherm (Hasterok

for various classes of mantle-derived garnet xenocrysts (after Grütter et al., 2004) from the Nxau

and Chapman, 2011). The diamond–graphite transition curve is after Day (2012).

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Fig. 11. Ni-in-garnet temperatures ( $T_{Ni}$ ) plotted against TiO<sub>2</sub> (a), Mg# (b), and Zr (c) for mantlederived garnet xenocrysts from the Nxau Nxau kimberlites in NW Botswana. The garnet classes are after Grütter et al. (2004).

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1011 Fig. 12. (a) Y ( $\mu$ g/g) versus Zr ( $\mu$ g/g) plot after Griffin et al. (1999b) and (b) Zr/Hf versus Ti/Eu 1012 plot after Shu and Brey (2015) for mantle-derived garnet xenocrysts from the Nxau Nxau 1013 kimberlites in NW Botswana.

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Fig. 13. Primitive-mantle-normalized trace element patterns (normalization values after Palme and
O'Neill, 2003) for melts calculated to have beeen in equilibrium with Nxau Nxau clinopyroxene
and garnet (G1, G9, G11) under deep upper mantle conditions (6-12 GPa). The whole-rock trace
element pattern for hypabyssal kimberlite from Nxau Nxau is taken from Tappe et al. (2020).
Partition coefficients for clinopyroxene/melt and garnet/melt are after Girnis et al. (2013).

Fig. 14. (a) Nd/Y versus Sc/Y, (b) Nd/Y versus T<sub>Ni</sub> (°C), and (c) Mg# versus Cr<sub>2</sub>O<sub>3</sub> (wt.%)
diagrams for mantle-derived garnet xenocrysts from the Nxau Nxau kimberlites in NW Botswana.
Diamond inclusion fields (garnet) after Stachel et al. (2004) and Batumike et al. (2009). The
"deeper garnets" mentioned in the figures are derived from >145 km depth.

Fig. 15. (a) TiO<sub>2</sub> (wt.%) versus Na<sub>2</sub>O (wt.%) and (b) Ca# versus Mg# diagrams for eclogitic G3 and pyroxenitic/low-Ca eclogitic G4 garnets from the Nxau Nxau kimberlite cluster in NW Botswana. Fields for high-Ca, low-Mg and high-Mg eclogite xenolith suites and the eclogite xenoliths database are after Aulbach and Jacob (2016) and Aulbach et al. (2020b).