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Title: Coexistence of alkaline-carbonatite complexes and high-MgO CFB in the Paranà-Etendeka province: Insights on plume-lithosphere interactions in the Gondwana realm

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perturbation of the conductive thermal regime and a crossing of volatilerich solidus (mostly P 2-3 GPa, Tp 1300-1400°C) with the generation of alkaline melts from the most fusible (hydrated and carbonated) mantle domains. Dear Editor

Thank you for your rapid decision and for the final corrections that have been promptly included in the manuscript.

Kind Regards,

Luigi Beccaluva and Co-authors.

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Research Highlights:

- High-MgO CFB and alkaline-carbonatite complexes coexist at the centre of Paranà-Etendeka LIP
- The two magma types are generated under distinct P-T-X conditions at the plume axial zone
- Similar tectonomagmatic occurrences are characteristic of other Gondwana LIPs
- High-MgO CFB magmas derived from plume-related sublithospheric mantle sources
- Alkaline magmas derived from metasomatized sources in the overlying lithosphere

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4	
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41 **1. Introduction**

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43 In the last decades, several authors have drawn attention to the close spatial-temporal 44 relationships between alkaline-carbonatite complexes and Continental Flood Basalts (CFB) in many 45 Large Igneous Provinces (LIP), giving rise to a longstanding debate on the role of lithosphere and 46 plume/asthenosphere components of their respective magma sources (Ellam and Cox, 1991; Bell, 47 2001; Gibson et al., 2006; Campbell, 2007; Ernst and Bell, 2010; Safonova and Santosh, 2014; 48 Pirajino, 2015). Specifically, in the Gondwana realm, alkaline-carbonatite complexes appear to be 49 closely related to high-MgO picrite-basalt rocks at the intersection of rift structures characterising 50 the inner part of plume-related CFB provinces, such as Karoo and Deccan (Natali et al., 2017) and, 51 particularly, Paranà-Etendeka, where these rock associations are well-documented (Comin52 Chiaramonti et al., 2011; Gomes et al., 2011). For the latter province, petrogenetic investigations 53 have relevant implications considering the debated origin of the Paranà-Etendeka, which is either 54 related to the Early Cretaceous activation of the proto-Tristan plume (Cordani et al., 1980; White 55 and McKenzie, 1989; 1995; Bizzi et al., 1995; Gibson et al., 1995; 2006; Thompson et al., 2001; 56 Campbell, 2001; Tuff et al., 2005; Campbell and Davies, 2006) or to passive rifting events that 57 mainly involved melting of the lithosphere (Peate et al., 1999; Hawkesworth et al., 2000; Ernesto et 58 al., 2002; Iacumin et al., 2003; Guarino et al., 2013; Rocha-Junior et al., 2013).

59 Therefore, the integrated study of nearly coeval high-MgO CFB and alkaline igneous rocks from this province represents a very convenient case study to investigate their genetic relationships, 60 61 including P-T conditions, source compositions and melting degree within a coherent 62 tectonomagmatic framework. For this purpose, more than three thousand analyses from the 63 literature, including major/trace and Sr-Nd-Pb isotopic data have been revisited, emphasising the 64 petrological and geochemical characteristics of CFB and the nearly coeval alkaline-carbonatite 65 complexes. In addition, the review includes the analogous occurrences from Deccan and Karoo with 66 the aim of evaluating, within a homogeneous scheme, whether these recurrent igneous associations 67 of west-central Gondwana may be attributed to a common tectono-magmatic scenario.

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69 **2. Methods**

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Bulk rock chemical analyses of CFB and associated alkaline-carbonatite complexes from the three investigated provinces were retrieved by the GEOROC database. For Paranà-Etendeka, CFB samples were reclassified as Low-Ti (LT) and High-Ti (HT1) suites, discriminating samples on the basis of Ti, Fe, Nb and Ce, as recently proposed by Natali et al. (2016; 2017) for the Deccan and Karoo provinces. Major element compositions of the investigated igneous associations are discussed and compared with the experimental petrology data in order to constrain phase equilibria and conditions of magma generation (Walter, 1998; Green and Fallon, 2005; Gudfinnsson and Presnall, 2005; Dasgupta et al., 2007; Pilet et al., 2008). Reconstruction of primary magmas and the relative thermobarometric conditions were estimated according to PRIMELT3MEGA (Herzberg and Asimow, 2015), FRACTIONATEPT (Lee et al., 2009) and MANTLEPT (Putirka, 2016) using major element compositions, whereas incompatible trace elements were used to constrain source enrichment and the degree of melting (Pilet et al., 2011). Moreover, the available Sr-Nd-Pb isotopic data were taken into consideration to constrain the nature of magma sources in terms of mantle components (Zindler and Hart, 1986; Hofmann, 1997; Stracke et al., 2012).

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3. High-MgO CFB and associated alkaline-carbonatite complexes in west-central Gondwana 87

- 88 3.1 Paranà-Etendeka igneous Province
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The Paranà-Etendeka CFB province of the south American and southwest African margins
mostly consists of LT and HT1 basalts, locally topped by rhyolitic volcanic rocks (Bellieni et al.,
1986; Piccirillo and Melfi, 1988; Piccirillo et al., 1989; 1990). The chronology of CFB spans from
139 to 128 Ma, with the oldest recorded ages occurring in some basalts of western Paranà. The
main magmatic phases are in the range of 135-130 Ma (Stewart et al., 1996; Renne et al., 1996a;
1996b; Gibson et al., 2006; Thiede and Vasconcelos, 2010; Janasi et al., 2011; Pinto et al., 2011).

96 Geochemical data reviewed in this work are used for the paleogeographic reconstruction of 97 the Paranà-Etendeka province reported in Fig. 1. CFB are classified in LT (TiO₂ 0.5-2.5 wt%) and 98 HT1 (TiO₂ 2.0-4.5 wt%) suites, as shown in Fig. 2. In the Etendeka region, both suites include high-99 MgO (> 10 wt%) basaltic and picritic rocks typically represented by HT1 Doros picrites, olivine-100 rich gabbroic intrusives and LT Horingbaai dikes (Thompson et al., 2001; Jennings et al., 2017; 101 Owen-Smith et al., 2017). The significant systematic increase of incompatible elements (such as Nb 102 and Ce) from LT to HT1 at comparable MgO values is illustrated in Fig. 2. Chondrite-Normalized 103 Rare Earth Element (REE) patterns show a significant increase in the La_N/Yb_N ratio from LT to

104 HT1 (4.0-4.7 in LT, 7.0-8.5 in HT1; Fig. 3), indicating a systematic enrichment of incompatible 105 elements and/or a lower melting degree of HT mantle sources. In the petrogenetic grid of Fig. 4, the 106 two suites show distinct FeO enrichment at comparable MgO, particularly for the least fractionated 107 lavas, suggesting mantle sources with different iron content. Representative differentiation trends 108 modelled for the two suites (PETROLOG v.3, Danyushevsky and Pletchov, 2011) show that they 109 are both characterised by the early fractionation of olivine (Ol), followed by clinopyroxene (Cpx) 110 for HT1, and plagioclase (Pl) and Cpx for LT, which are consistent with the phenocryst 111 assemblages observed in the rocks.

The reconstruction of primary magmas and their potential temperature (T_p) have been 112 113 performed through the accumulated fractional melting algorithm PRIMELT3MEGA (Herzberg and Asimow, 2015), assuming the anhydrous lherzolite KR4003 (Walter, 1998) as a mantle source 114 115 having mg# of 0.90, which is suitable for tholeiitic magmas. The calculated primary melts have MgO 21.2 wt%, FeO 11.1 wt%, mantle potential temperature $(T_p) \sim 1590^{\circ}$ C for HT1, and MgO 116 18.1 wt%, FeO 9.6 wt%, $T_p \sim 1520^{\circ}$ C for LT. Phase equilibria constraints and the application of 117 118 geobarometers by Herzberg et al. (2007) and Guddfinnson and Presnall (2005) suggest that the 119 generation of these primary melts occurred at ~ 5 GPa for HT1 and 3-4 GPa for LT (Supplementary Table 1). Compared to the above thermobarometric estimates, T_p obtained by the model of Lee et al. 120 (2009) show a good agreement, and T_p by the model of Putirka (2016) are 70-80°C higher. 121 Conversely, the pressures obtained by Lee et al. (2009) and Putirka (2016) are always 122 123 underestimated with respect to those of Herzberg et al. (2007) and Guddfinnson and Presnall (2005; 124 Supplementary Table 1). As shown by the Primitive Mantle (PM) incompatible element distribution 125 (Fig. 5), the HT1 and LT primary magmas could be formed by 9% and 22% melting, respectively, from a mixed mantle source composed by 97% PM and 3% eclogite (recorded as xenoliths from 126 127 Angolan kimberlites; Shervais et al., 1988).

128 The most representative alkaline complexes coeval (133-128 Ma) with CFB occur in the 129 elliptical area depicted in Fig. 1, which corresponds to the centre of the restored Paranà-Etendeka 130 province. In this area, several extensional lineaments converge towards the Walvis Ridge and the 131 early track of the South Atlantic opening. On the south American side of the province, these 132 complexes are represented by Juquia, Jacupiranga and Anitapolis, located in the Ponta Grossa Arch 133 and along the Brazilian coast (Beccaluva et al., 1992; 2017; Gomes et al., 2011 and references 134 therein). In the south-western African margin, alkaline and alkaline-carbonatite complexes are 135 widespread in the Namibian Damara Belt (e.g., Erongo, Paresis, Okenyenya and Okurusu; Comin-136 Chiaramonti et al., 2011 and references therein) and in the Angolan Moçamedes Arch (e.g., 137 Chivira-Bonga; Coltorti et al., 1993; Alberti et al. 1999).

The magmatic associations of alkaline complexes from both south American and African 138 139 margins consist of mafic lithologies -such as alkali basalts, basanites/tephrites, nephelinites, and 140 ankaratrites- or their plutonic counterpart, coupled with ultramafic cumulates, nepheline-syenites 141 and carbonatites (Beccaluva et al., 1992; 2017; Morbidelli et al., 1995; Le Roex and Lanyon, 1998; 142 Trumbull et al., 2003; Comin-Chiaramonti and Gomes, 2005; Ruberti et al., 2005; Gibson et al., 143 2006; Gomes et al., 2011; Azzone et al., 2013). All of these lithologies are silica-undersaturated 144 with variable amounts of nepheline and the ubiquitous presence of hydrous phases, such as 145 amphibole and/or phlogopite. The latter is the dominant hydrous phase in the Brazilian alkaline 146 complexes, typically showing a more potassic character with respect to those from Namibia and 147 Angola (Fig. 6). Accordingly, the PM-normalized incompatible element patterns of Fig. 7 show that 148 the African alkaline magmas, unlike those from Brazil, are characterised by the presence of a 149 significant negative K anomaly. As demonstrated by experimental petrology, this indicates a major 150 role of amphibole with respect to phlogopite in mantle sources and a consequent K deficiency in the 151 generated melts (K< 20000 ppm; Späth et al., 2001; Rooney et al., 2017). Therefore, for a 152 comparable alkali/silica ratio, the potassic or sodic affinity of alkaline magmas depends on the 153 relative phlogopite/amphibole proportion in their mantle sources. Generally, the significant 154 presence of hydrous (and carbonated) phases is a necessary requirement in the genesis of alkaline melts, as invariably indicated by experimental petrology, either from 1) homogeneously 155

156 metasomatised or 2) variably veined mantle sources. In the first case, experimental results (Green 157 and Fallon, 2005; Gudfinnsson and Presnall, 2005; Dasgupta et al., 2007) show that alkaline magmas could be generated by decreasing the degree of partial melting (< 10% alkali basalts and 158 159 basanites; < 5% nephelinites, melilitites and carbonatites) of hydrated and/or carbonated fertile 160 lherzolite at increasing pressure from ~ 2 to 3.5 GPa within the lithosphere (Morris and Pasteris, 1987; Nielson and Wilshire, 1993). In the second case, alkaline basic melts (e.g., basanites, 161 nephelinites) have been obtained by melting experiments on natural amphibole-rich veined 162 163 lherzolites at 1.5 GPa (Pilet et al., 2011).

164 From the above, the modelling of alkaline melts has to include the effect of metasomatism, 165 which decreases the mg# of the inferred source with respect to that of the unmetasomatised mantle. 166 In fact, olivine composition in alkaline magmas is generally \leq Fo₈₈ (Harris et al., 1999; Coltorti et 167 al., 1993; Beccaluva et al., 1992; 2017; Trumbull et al., 2003), a value that is significantly lower 168 than that recorded in CFB (Fo up to 89-90; Beccaluva et al., 2009; Natali et al., 2016; 2017; 169 Thompson and Gibson 2000), conforming to a source composition that does not exceed mg# 0.88. 170 Accordingly, the Paranà-Etendeka alkaline primary magmas and the related P-T estimates have 171 been modelled using the algorithms of Putirka (2016), which allows taking into proper account the 172 effect of metasomatism. The results, which are reported in Supplementary Table 1, indicate that the investigated alkaline magmas are compatible with generation in the P range 1.8-2.0 GPa and T_p 173 174 1300-1360°C, by melting of a mantle source having mg# 0.87. Notably, these thermobarometric 175 conditions approach those recorded by amphibole-bearing peridotite xenoliths from the Damara 176 Belt lithosphere (Baumgartner et al., 2000; Le Roex and Class, 2014). As reported in Fig. 8, the 177 incompatible element distribution of the Paranà-Etendeka alkaline magmas are satisfactorily 178 reproduced assuming mantle sources variably enriched by metasomatic veins, as proposed by Pilet 179 et al. (2008; 2011). The melting model of the African alkaline magmas indicates a best fit 180 calculation either by a low melting degree (F $\sim 2\%$) of a PM source hybridized by 20% amphibolerich metasomatic veins or by a higher melting degree (F \sim 10%) of highly veined (40%) PM source 181

182 (Fig. 8a). Conversely, the more potassic character of alkaline melts from south America necessarily 183 implies a significant presence of phlogopite, in addition to amphibole, in their mantle sources. In this case, best fit can be obtained either by $F \sim 2\%$ of a PM source hybridized by 40% amphibole-184 phlogopite veins, or by $F \sim 8\%$ of a PM source dominated by the amphibole-phlogopite metasome 185 186 (90%, Fig. 8b). It is important to note that conceptually, in these models the metasomatising veins 187 include variable proportions of cumulate hydrous minerals and residual liquids that could also be 188 carbonated (Pilet et al., 2011). As recently observed in various volcanic provinces, this phenomenon 189 conforms to the wide variability of lithospheric mantle sources, in terms of extent and composition 190 of metasomatism, configuration of the lowered solidus and melting degrees (Jung et al., 2011; 191 Rooney et al., 2017). Noteworthy, by a purely geochemical standpoint, the incompatible element 192 distribution could also be approached by < 1% melting of an unmetasomatised (volatile-poor) 193 mantle source, but this possibility is strongly discounted due to the ubiquitous evidence that alkaline 194 magmas are significantly hydrated and carbonated.

195 The distribution of Sr-Nd-Pb isotopes for Paranà-Etendeka CFB and associated alkaline 196 complexes is reported in Fig. 9. Taken as a whole, the two magmatic associations show distinct 197 isotopic signatures that imply significant differences in their mantle sources. In particular, most 198 high-MgO CFB from Etendeka (LT and HT1 picrites and some basalts) cluster in the upper left of the $\epsilon Nd^{-87}Sr/^{86}Sr_{(i)}$ diagram covering a restricted isotopic range (ϵNd 9.1-0.5 and $^{87}Sr/^{86}Sr_{(i)}$ 199 200 0.70326-0.70513). As observed by other authors (Thompson et al., 2001; Hoernle et al., 2015; 201 Owen-Smith et al., 2017) this isotopic signature is attributable to uncontaminated, sublithospheric 202 mantle components. Conversely, all of the Paranà and the remaining Etendeka CFB (MgO 9-5 wt%) extend in the lower right quadrant towards low ϵ Nd (down to -10) and extremely high 87 Sr/ 86 Sr_(i) 203 204 (up to 0.7142) values, reflecting the variable involvement of lithospheric mantle components and/or 205 variable continental crust contamination at progressive differentiation. In particular, HT1 from Paranà display a relatively restricted isotopic range (⁸⁷Sr/⁸⁶Sr_(i) 0.70495-0.70620 and ɛNd from -206 207 1.27 to -4.32) that could be related to a significant role of lithospheric components in their mantle

sources, whereas the wide 87 Sr/ 86 Sr_(i) variation recorded in LT certainly reflects continental crust 208 209 contamination. The Pb isotopic composition of the entire CFB population ranges between ²⁰⁷Pb/²⁰⁴Pb 15.4-15.8 and ²⁰⁶Pb/²⁰⁴Pb 17.1-19.7, with the abovementioned high-MgO Etendeka CFB 210 plotting in the middle of the distribution. On the whole, the Sr-Nd-Pb isotope distributions of 211 212 uncontaminated CFB from Etendeka shows a good agreement with the "Gough" component, which 213 has been recently identified as the marker of the initial (proto-Tristan) plume activity since 133-132 214 Ma (Hoernle et al., 2015). In diagrams of Fig. 9 the Paranà-Etendeka alkaline complexes show 215 distinct isotopic compositions that plausibly reflect a derivation from independent and different 216 mantle sources: the Brazilian complexes extend from near FOZO (Prevalent mantle) to EM1 (ENd from +4.5 to -7.8, 87 Sr/ 86 Sr_(i) 0.70410-0.70632, 206 Pb/ 204 Pb 17.1-18.0), whereas those from Namibia 217 and Angola display affinity with the HIMU component (ϵ Nd from +5.0 to -1.8, 87 Sr/ 86 Sr_(i) 0.70375-218 0.70487, ²⁰⁶Pb/²⁰⁴Pb 18.2-19.7). These different isotopic signatures appear to be correlated with the 219 220 potassic vs sodic affinity, testifying for a different history of metasomatic enrichment in the 221 lithospheric mantle of the south American and African margins.

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223 3.2 Comparison with Deccan and Karoo igneous provinces

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Deccan is a LIP where CFB cover an area of ~ $600,000 \text{ km}^2$ and mostly consist of tholeiitic 225 226 basalts -locally overlain by rhyolites- that were emplaced from 63 to 68 Ma, with the main 227 magmatic phase at 65-66 Ma (Cox and Hawkesworth, 1985; Melluso et al., 1995; 2004; 2006; 228 Chenet et al., 2007; Sheth and Melluso, 2008; Sheth et al., 2014; Chatterjee and Bhattacharji, 2008; 229 Keller et al., 2009; Manikyamba et al., 2015; Richards et al., 2015). The Deccan province is widely 230 considered linked to the activation of the Reunion hot spot and was originally close to the 231 Seychelles plateau before the opening of the Central Indian Ocean (Cox, 1989; White and McKenzie, 1989; Courtillot et al., 2003), although this view is contended by some authors (e.g., 232 233 Sheth, 2005). In the paleogeographic reconstruction reported in Fig. 10 (modified after Natali et al.,

234 2017), the Deccan traps represent part of a wider magmatic province originally extending for \sim 235 2000 km maximum diameter. The spatial distribution of Deccan lavas is zonally arranged with most 236 CFB represented by LT basalts, whereas the occurrence of HT1 basaltic and picritic lavas is 237 restricted to the NW sector. This area is located at the intersection of major NS-EW rift systems and is also characterised by the occurrence of nearly coeval alkaline-carbonatite complexes. 238 239 Northeastward of the main rift systems, the CFB outcrops decrease in relation to the rapid increase 240 of lithosphere thickness below the Bastar craton (Paul et al., 2008; Sen et al., 2009; 2012; 241 Chalapathi Rao and Lehmann, 2011 and references therein). Reconstruction of primary CFB 242 magmas and thermobarometric conditions using the Guddfinnsonn and Presnall (2005), Herzberg et 243 al. (2007) and Herzberg and Asimow (2015) algorithms suggest that HT1 could be generated by T_p up to 1560°C at pressures 4-5 GPa, whereas LT were formed at lower temperature ($T_p \sim 1500^{\circ}$ C) 244 and pressures (3-4 GPa). The thermobarometric models by Lee et al. (2009) and Putirka (2016) give 245 246 temperature estimates in reasonable agreement (T_p 1500-1600°C), whereas pressures are 247 underestimated with respect to those obtained by the Guddfinnsonn and Presnall (2005) and 248 Herzberg et al. (2007) models (2017; Supplementary Table 1). Based on the incompatible element 249 distribution of primary melts, Natali et al. (2017) estimated that HT1 and LT were generated by \sim 250 9% and 17% melting degree of a PM source, respectively.

251 Geochemical features of alkaline basic rocks s.l. coeval with CFB and related to the main 252 rift structures (Kutch, Cambay and Narmada rifts) are reported in Fig 6 and 7, where they display a 253 sodic affinity and variable LREE enrichment (La_N/Yb_N 13-34). As modelled for Paranà-Etendeka, 254 Deccan alkaline primary magmas have been calculated by the Putirka (2016) model, assuming a 255 metasomatised mantle source having mg# 0.87. Accordingly, mantle potential temperature (T_p) and pressure (P) of generation are in the range of 1370-1350°C and 2.1-2.0 GPa, respectively. Fig. 7 256 257 shows that the incompatible element patterns of Deccan alkaline magmas are analogous to those of 258 Namibia and could be generated by similar metasomatised mantle sources, as modelled in Fig. 8.

259 The isotope systematics of the Deccan magmatic province is reported in Fig. 11. Similar to what 260 was observed in the Paranà-Etendeka, HT1 picrites and the least differentiated (high-MgO) LT basalts display a restricted isotopic range (ENd from 6.8 to 2.5, ⁸⁷Sr/⁸⁶Sr_(i) 0.70386-0.70491) 261 262 plotting in the upper-left quadrant of the diagram, which can be attributed to uncontaminated sublithospheric mantle components. The rest of the Deccan CFB record ⁸⁷Sr/⁸⁶Sr_(i) up to 0.71756, 263 264 clearly in relation to the variable extent of crust contamination. Alkaline-carbonatite complexes 265 show a distinctly different Sr-Nd-Pb composition with respect to CFB and a tendency towards the 266 HIMU geochemical component, similar to those from Namibia and Angola.

267 The Karoo CFB province of southern Africa (and its extension in the Dronning Maud Land 268 of Antarctica) is classically considered a LIP originated by the breakup of southern Gondwana and 269 the opening of the south western Indian Ocean (White and McKenzie, 1989; Storey and Kyle, 1997; 270 Elliot and Fleming, 2000; Storey et al., 2013; Riley et al., 2005; Heinonnen et al., 2014). The Karoo CFB activity mostly occurred from 174 and 184 Ma, (Hastie et al., 2014) and mainly consist of 271 272 tholeiitic basalts-picrites (locally topped by rhyolites), whose variable geochemical and isotopic 273 characteristics have been attributed either to the Sub Continental Lithospheric Mantle (SCLM, 274 Duncan et al., 1984; Hawkesworth et al., 1984 Jourdan et al., 2007) or to mantle sources modified 275 by plume-related asthenospheric components (Ellam and Cox, 1991; Ellam et al., 1992; Sweeney et al., 1991; 1994). As reported in Fig. 12, Karoo CFB are zonally arranged, with very high-TiO₂ 276 277 (HT2) picrite-basalt lavas in the central area (Mwenetzi) and progressively lower-TiO₂ (HT1 and 278 LT) basalts towards the periphery (Natali et al., 2017). The Guddfinnsonn and Presnall (2005), 279 Herzberg et al. (2007) and Herzberg and Asimow (2015) algorithms applied to Karoo CFB yield T_p 280 of 1580°C at 5GPa and 1490°C at 3-4 GPa for HT2 and LT primary magmas, respectively. 281 Thermobarometric models by Lee et al. (2009) and Putirka (2016) give the same discrepancies 282 observed for Paranà-Etendeka and Deccan CFB, providing comparable temperatures but 283 systematically lower pressures (see Supplementary Table 1). Based on the incompatible element 284 distribution of primary melts, Natali et al. (2017) estimated that LT were generated by a ~ 14%

melting degree of a PM source, whereas HT2 were generated by 8% batch melting of a PM garnet
peridotite source hybridized with 15% eclogite.

Coexistent alkaline complexes occur at a triple junction located at the convergence of huge 287 288 dike swarms in the Mwenetzi region. These complexes are represented by the Mashikiri nephelinites outcropping below the HT2 lavas and the ijolite-nephelinite-carbonatite complexes of 289 290 Dorowa and Shawa, northward (Ellam and Cox, 1991; Harmer et al., 1998; Jourdan et al., 2007). 291 Rocks from these complexes are the most sodic (Fig. 6) and silica undersaturated with respect to 292 those from Paranà-Etendeka and Deccan, and according to the thermobarometric model of Putirka (2016), their primary magmas were generated at T_p of 1350-1410°C in the pressure range of 2.4-2.7 293 294 GPa (Supplementary Table 1). The PM-normalized incompatible element and the chondrite-295 normalized REE distribution (Fig. 7) reveal the most depressed patterns with respect to those of the 296 other investigated provinces, the lowest LREE enrichment ($La_N/Yb_N \sim 13$ in Mashikiri nephelinites and ~ 5 in ijolites), as well as unusual positive anomalies in Sr and Ti. These features suggest a 297 298 peculiar metasomatic enrichment of previously depleted mantle sources, quite different with respect 299 to that envisaged for other provinces.

300 Sr-Nd isotopes of Karoo HT and LT, plotted in Fig. 13, display divergent trends that are 301 partially overlapped in proximity to the Bulk Solid Earth composition, which could be attributed to 302 the uncontaminated sublithospheric mantle of this region. The HT picrite-basalt suites display 303 variation towards lower ENd (down to -9.7), plausibly in relation to the involvement of lithospheric components in their magma genesis. The LT suite shows a trend towards very high 87 Sr/ 86 Sr_(i) 304 305 values, indicating a variable extent of crustal contamination. Pb isotopes also show systematic 306 differences between LT and HT suites. Conversely, alkaline magmas are distinct in that they show 307 an extremely unradiogenic Nd isotopic composition (ENd down to -19.9), which has been interpreted as the signature of the enriched SCLM (cf. Harmer et al., 1998). 308

309

4. Discussion and conclusions

312 The critical review and new elaboration of literature data on high-MgO CFB and coeval 313 alkaline complexes from the Paranà-Etendeka LIP provide constraints on the P-T-X conditions of 314 their mantle sources. The inception of the CFB magmatism (approximately 139 Ma) occurred in the 315 northwestern portion of the Paranà basin and migrated southeastward towards the Etendeka region, 316 likely in connection with a generalized northwestward lithospheric drift of this region of Gondwana 317 over an active plume (Turner et al., 1994; Gibson et al., 2006). Whatever the extent of the 318 lithospheric drift before the South Atlantic opening, the focus of the tectonomagmatic activity at 319 135-130 Ma was well-established in the Etendeka region, which comprises the oldest parts of the 320 Walvis Ridge and several extensional lineaments that intersect the early track of the south Atlantic 321 opening. Paleogeographic restoration shows that this region, at the centre of the CFB province, is 322 characterised by the exclusive occurrence of high-MgO basalt-picrite rocks (belonging to both LT 323 and HT1 suites) and is spatially/temporally (133-128 Ma) associated with alkaline-carbonatite 324 complexes from the Ponta Grossa arch (Brazil) and Damara belt (Namibia) extensional structures.

325 The new petrogenetic modelling and thermobarometric estimates obtained in this work indicate that the primary magmas of the HT1 suite are the hottest and deepest CFB ($T_p \sim 1590^{\circ}$ C, P 326 327 \sim 5 GPa) of the entire province. This finding conforms to the interpretation that magma generation 328 was triggered by thermal and tectonic effects related to the impingement of a hot plume on a 329 relatively thick continental lithosphere. The maximum temperature excess (T_{ex}) , both with respect to 330 notional MORB and local mantle xenoliths thermobarometry, is estimated as 250-300°C, in 331 agreement with what was obtained for Deccan and Karoo (this work and Natali et al., 2017). 332 Moreover, most high-MgO CFB of the Etendeka region show a Sr-Nd-Pb isotopic range 333 corresponding to prevalent mantle compositions uncontaminated by lithospheric signatures, and 334 share the "Gough" geochemical component recently recognized as the initial proto-Tristan plume 335 activity (Hoernle et al., 2015). All of these features agree with a rapid ascent of high-MgO Etendeka 336 magmas that, in our opinion, effectively represent the most genuine proxies of plume-related 337 sublithospheric melts, virtually unaffected by lithospheric contamination. By contrast, out of the 338 focal zone, most basalts of the Paranà-Etendeka CFB province are variably differentiated (MgO 9-5 wt%) and display variable increases of 87 Sr/ 86 Sr_(i), reflecting either lithosphere/asthenosphere mixed 339 sources (HT1 from Paranà) and/or remarkable shallow level crustal contamination. This 340 341 interpretation can reconcile the contrasting views on the role of "lithosphere" vs "plume" in the 342 genesis of CFB (Turner et al., 1996; Hawkesworth et al., 2000; Gibson et al., 2000; 2006), since 343 both views appear to be appropriate for the different CFB sections, reflecting different extents of 344 plume-lithosphere interaction.

345 The Paranà-Etendeka alkaline-carbonatite complexes coeval with CFB show petrological 346 and isotopic signatures that agree with melts from lithospheric mantle sources. The results from 347 modelling favour the genesis of the studied alkaline magmas by moderate to low melting degrees of 348 lithospheric mantle sources that were significantly enriched (veined?) by metasomatic phases 349 (amphibole and phlogopite). Alkaline rocks display regional geochemical differences, suggesting 350 distinct metasomatising events with a more relevant role of phlogopite in magma sources from south America with respect to those from southern Africa. Accordingly, the Brazilian complexes 351 352 have a more potassic character and isotopic tendency to the EM1 mantle component, whereas those 353 from Namibia and Angola display sodic affinity coupled with a signature approaching the HIMU 354 mantle component.

355 On a regional scale, the main tectono magmatic characteristics of Paranà-Etendeka are also 356 shared by Deccan and Karoo, where superheated picrite-basalt (mostly HT tholeiitic suites) and 357 alkaline-carbonatite complexes occurred at the intersection of multiple extensional lineaments -such 358 as faulting, rifting and dike swarms- radiating from the central area of each province (cfr Natali et 359 al., 2017). As already observed for Paranà-Etendeka, the isotopic data of alkaline-carbonatite 360 complexes of Deccan and Karoo invariably show significant differences with respect to associated 361 high-MgO CFB, and mostly record continental lithospheric signatures. In our opinion, a satisfactory explanation for the spatial-temporal association of such contrasting, and isotopically distinct, 362

363 magma types cannot be ascribed to common mantle sources, but requires the nearly contemporaneous generation of high-MgO CFB and alkaline melts from distinct mantle systems, 364 namely the convective asthenosphere and the subcontinental lithosphere in the focal zone of LIPs. 365 366 Accordingly, in the generalized tectonomagmatic model proposed in Fig. 14, the same thermal and tectonic events that characterised the axial zone of the impinging hot plume underneath the 367 368 Gondwana lithosphere triggered melting of both asthenospheric and lithospheric mantle sources 369 with magma rising through a nearly open feeding system. The results from modelling indicate that 370 the asthenospheric peridotite solidus is crossed mostly in the range 4-5 GPa and T_p 1500-1600°C 371 with the generation of high-MgO CFB magmas whose sources plausibly experienced adiabatic 372 decompression and melting over a large mantle column (Gibson et al., 2000; Thompson et al., 2001; 373 Herzberg and Asimov, 2015; Natali et al., 2016; 2017; Jennings et al., 20017). The same plume 374 effects favoured the generation of alkaline melts by moderate to low melting degrees of the most 375 fusible lithospheric domains (P 2-3 GPa, T 1300-1400°C), where the solidus is variably depressed 376 due to occurrences of hydrated and/or carbonated phases; despite the small volume, alkaline melts 377 ascended to upper crustal levels, favoured by their intrinsic low density and viscosity.

It should be noted that for LIP that do not present anomalous and focalized thermomechanical input on thick continental lithosphere-as in the case of the cooler CAMP and Ferrar CFB provinces-other models could be more appropriate as alternative (or complementary) to the hot plume hypothesis (Coltice et al., 2009; Hole, 2015; Natali et al., 2016).

382

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784 **Figure captions**

785

Figure 1 – Paleogeographic reconstruction of Parana-Etendeka CFB at 135-130 Ma (modified after White and McKenzie, 1989). Low-Ti (LT) and high-Ti (HT1) spatial distribution was reviewed after data from Piccirillo and Melfi (1988), Piccirillo et al. (1990), Hawkesworth et al. (1992), Peate et al. (1999), Marzoli et al. (1999), Thompson et al. (2001), Lustrino et al. (2005), Cernuschi et al. (2015), Marsh and Swart (2016), Rämö et al. (2016). Locations of Early Cretaceous alkalinecarbonatite complexes after Beccaluva et al. (1992; 2017), Coltorti et al. (1993), Trumbull et al. (2003), Comin-Chiaramonti and Gomes (2005), Gibson et al. (2006), Comin-Chiaramonti et al. (2011; 2014) and references therein, Gomes et al. (2011) and references therein. The hot spot focal zone includes the oldest part of the Walvis volcanic ridge and the Etendeka high-MgO rocks (max T_p 1590°C) and is considered the axis of the Proto-Tristan mantle plume during the main magmatic phase of the Paranà-Etendeka province; it also includes the majority of coeval alkaline-carbonatite complexes, along the extensional structures that intersect the early track of the south Atlantic opening.

799

Figure 2 – FeO_t, TiO₂, Nb and Ce *vs* MgO variation diagrams for the Paranà-Etendeka CFB. Data from the GEOROC database (http://georoc.mpch-mainz.gwdg.de/georoc/) and by Owen-Smith et al. (2017). LT type localities are Esmeralda, Gramado in Paranà and Horingbaai, Tafelberg in Etendeka. HT1 type localities are Paranapanema, Pitanga, Urubici in Paranà and Khumib, Doros in Etendeka. Empirical boundaries between LT and HT1 CFB are drawn in order to minimise the misclassified samples (generally less than 5%).

806

Figure 3 – Chondrite-normalized Rare Earth Element (REE) patterns for LT and HT1 Paranà-Etendeka CFB. Average La_N/Yb_N for each group are also reported. Normalizing factors are from Sun and McDonough (1989). Data are from the GEOROC database and from Owen-Smith et al. (2017).

811

Figure 4 – MgO *vs* FeO diagram for the Paranà-Etendeka CFB. HT1 and LT primary magmas were modelled according to Herzberg and Asimow (2015). Liquid lines of descent were modelled according to Petrolog software v.3 (Danyuschevsky and Pletchov, 2011). Data are from the GEOROC database and from Owen-Smith et al. (2017). Abbreviations: Ol = olivine, Cpx =clinopyroxene; Pl = plagioclase.

817

Figure 5 – Incompatible element distribution of calculated LT and HT1 primary melts (data from
Ewart et al., 1998; Gibson et al., 2000) and modelled composition obtained by batch melting of a
PM source hybridized with 3% eclogite. Source mode and melting proportions conform to
experimental data by Walter (1998); partition coefficients (*Kd*) from the GERM database.
Normalizing factors are from Sun and McDonough (1989). See text for further explanation.

823

824 Figure 6 – Na₂O vs K₂O (wt%) binary diagram for mafic rocks from alkaline-carbonatite complexes 825 coeval with CFB from the a) Paranà-Etendeka, b) Deccan and c) Karoo igneous provinces. 826 Subdivision among various sodic and potassic affinities are from Middlemost (1975). Paranà-827 Etendeka data are from Beccaluva et al. (1992; 2017), Coltorti et al. (1993), Trumbull et al. (2003), 828 Comin-Chiaramonti et al. (2002; 2011; 2014), Comin-Chiaramonti and Gomes (2005), Gibson et al. 829 (2006), Gomes et al. (2011) and references therein. Deccan data are from Simonetti et al. (1998) 830 and Sen et al. (2009); alkaline rocks locally interbedded within CFB (e.g., Melluso et al., 1995) are 831 not considered. Karoo data are from Harmer et al. (1998) and de Bruiyn et al. (2005).

832

833 Figure 7 - Chondrite-normalized Rare Earth Element (REE) and Primitive Mantle-normalized 834 incompatible element patterns for mafic rocks from alkaline-carbonatite complexes coeval with 835 CFB from a) Paranà-Etendeka, b) Deccan and c) Karoo igneous provinces. Paranà-Etendeka data 836 are from Beccaluva et al. (1992; 2017); Coltorti et al. (1993); Trumbull et al. (2003), Comin-837 Chiaramonti et al. (2002; 2011; 2014); Comin-Chiaramonti and Gomes (2005); Gibson et al. 838 (2006); Gomes et al. (2011) and references therein. Deccan data are from Sen et al. (2009), whereas 839 Karoo data are from Harmer et al. (1998) and de Bruiyn et al. (2005). Normalizing factors after Sun 840 and McDonough (1989).

841

Figure 8 – Incompatible element distribution of alkaline basic melts coeval with CFB in the ParanàEtendeka province. a) modelling indicates that Etendeka alkaline basic melts could be generated

844 either by 2% or 10% batch melting of a PM source hybridized with 20% and 40% amphibole-rich 845 metasomatic veins, respectively. Composition of metasomatic veins, mineral modes and melting 846 coefficients are after Pilet et al. (2011). b) Modelling for Brazilian alkaline basic magmas requires 847 the significant presence of phlogopite (in addition to amphibole) in the source. Best fit is obtained either by 2% or 8% batch melting of a PM source hybridized with 40% and 90% of metasomatic 848 849 veins, respectively. In this case, amphibole-rich metasomatic veins (Pilet et al., 2011) contain up to 850 30% of phlogopite. Data source as in Fig. 7. Partition coefficients (Kd) from the GERM database. 851 Normalizing factors are from Sun and McDonough (1989).

852

Figure 9 - (a) Sr-Nd and (b) Pb isotopic composition of CFB and coeval alkaline-carbonatite 853 854 complexes for the Paranà-Etendeka igneous province. Data from GEOROC and from Huang et al. 855 (1995); Milner and Le Roex (1996); Le Roex and Lanyon (1998); Harris et al., (1999); Alberti et al. 856 (1999); Trumbull et al. (2003; 2007); Comin-Chiaramonti et al. (2007; 2011); Gomes et al. (2011); 857 Beccaluva et al. (2017); Owen-Smith et al. (2017). Isotopic composition of Gough and Tristan hot 858 spot tracks are from Hoernle et al. (2015). Reference mantle end-members (DM, EM1, EM2, HIMU 859 and FOZO) are also reported for comparison (Zindler and Hart, 1986; Hofmann, 1997; Stracke, 860 2012). Sr-Nd initial isotopic values have been calculated at 132 Ma.

861

Figure 10 – Paleogeographic reconstruction of the Deccan igneous province at ca. 65 Ma reporting the spatial distribution of HT1 and LT suites (modified after Natali et al., 2017 and references therein). Note that the centre of the encircled equidimensional area (Reunion hot spot) corresponds to the distribution of HT1 picrite-basalt, the maximum concentration of nearly coeval alkaline (carbonatite) complexes and the intersection of the main rift structures.

867

Figure 11 – (a) Sr-Nd and (b) Pb isotopic composition of CFB and coeval alkaline-carbonatite
complexes for the Deccan igneous province. Data from GEOROC and from Simonetti et al. (1995;

1998) and Sen et al. (2009). Reference mantle end-members (DM, EM1, EM2, HIMU and FOZO)
are also reported for comparison (Zindler and Hart, 1986; Hofmann, 1997; Stracke, 2012). Sr-Nd
initial isotopic values have been calculated at 65 Ma.

873

Figure 12 –Paleogeographic reconstruction of the Karoo igneous province at ca. 170 Ma, reporting the spatial distribution of HT2, HT1 and LT suites (modified after Natali et al., 2017 and references therein). Note that the central CFB area correspond to a triple junction defined by the convergence of dike swarms, the distribution of HT2 picrite-basalt and the location of nearly coeval alkalinecarbonatite complexes.

879

Figure 13 – (a) Sr-Nd and (b) Pb isotopic composition of CFB and coeval alkaline-carbonatite
complexes for the Karoo igneous province. Data from GEOROC and from Hawkesworth et al.
(1984) and Harmer et al. (1998). Reference mantle end-members (DM, EM1, EM2, HIMU and
FOZO) are also reported for comparison (Zindler and Hart, 1986; Hofmann, 1997; Stracke, 2012).
Sr-Nd initial isotopic values have been calculated at 180 Ma.

885

886 Figure 14 – Impinging of the mantle plume head on the pre-existing lithosphere caused a dramatic thermal anomaly (T_{ex} 250-300°C) coupled with bulging, thinning and development of extensional 887 888 lineaments that intersected and radiated from the plume axial zone. The combined tectonic and 889 thermal effects at the plume axis could explain the contemporaneous generation, virtually on the 890 same plumbing system, of high-MgO CFB and alkaline magmas from the convective asthenosphere 891 and the overlying lithosphere, respectively, under distinct P-T-X conditions (see inset): 1) high-892 MgO CFB could derive from volatile-poor sublithospheric mantle sources where the solidus is 893 crossed at T_p 1500-1600°C, 4-5 GPa by moderate-high melting degree under nearly-adiabatic 894 conditions; 2) parental melts of the alkaline-carbonatite complexes formed by a generally low 895 degree of melting in the lower portion of the lithosphere (T_p 1300-1400°C, P 2-3 GPa), where the 896 solidus is variably depressed owing to the common occurrence of hydrated and carbonated 897 components. Data for volatile-rich (hydrated and carbonated lherzolite) and volatile-poor 898 (anhydrous lherzolite) solidi are taken from the literature (Turner et al., 1996; Walter, 1998; 899 Thompson et al., 2001; Green and Fallon, 2005; Gudfinnsson and Presnall, 2005). The thermal 900 regime and composition of the lithosphere is based on mantle xenoliths from the Gondwana realm 901 that consist of Sp- to Gt-lherzolite/harzburgite variably enriched by metasomatic phases (amphibole, phlogopite and carbonates; Rivalenti et al., 2000; Fodor et al., 2002; Dessai et al., 902 903 2004; Griffin et al., 2003; Beccaluva et al., 2007; 2008; 2011; Karmalkar et al., 2009; Natali et al., 904 2013; Bianchini et al., 2014; Sgualdo et al., 2015; Stanley et al., 2015). The hatched area 905 corresponds to the asthenosphere-lithosphere transition.

906

907 **Table captions:**

908

909 Supplementary Table 1 – Major element composition of calculated primary melts for representative 910 high-MgO CFB and coeval alkaline basic melts Paranà-Etendeka, Deccan Karoo LIPs. 911 Reconstruction of CFB primary melts has been obtained by the Herzberg and Asimow (2015) 912 model assuming a mantle source with mg# 0.90, whereas alkaline primary melt have been obtained 913 using the Putirka (2016) model assuming mg# 0.87 for the metasomatized source. 914 Thermobarometric estimates have been obtained using the models of Guddfinnsonn and Presnall 915 (2005), Herzberg et al. (2007), Lee et al., (2009), Herzberg and Asimow (2015), Putirka (2016) for 916 CFB, whereas for alkaline magmas only the Putirka (2016) model could be applied.

917

Paragraphs at lines 90-126, 127-192 and 310-342 have been broken up.

The problem with horizontal axis of Fig. 3 has been fixed.

Arial font has been used for labels in Fig. 4.

The problem with vertical axis of Fig. 7a has been fixed.

Acronyms of mantle end-members in the isotopic diagrams of Figs. 9, 11 and 13 have been homogeneised.



Figure 2



Figure 3





Figure 4









Figure 8



Figure 10

²⁰⁶Pb/²⁰⁴Pb

²⁰⁶Pb/²⁰⁴Pb

20.0

Supplementary Table 1 Click here to download Background dataset for online publication only: Supplementary Table 1_rev2.xls