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Wehrlites from continental mantle monitor the passage and degassing of carbonated melts

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Supplementary Information

The Supplementary Information includes:

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Oxygen Fugacity Control on Redox Freezing vs. Wehrlitisation during Interaction with Carbonated Melts

The low proportion of wehrlite amongst garnet peridotites and inclusion-bearing diamonds is striking (Fig. S-1a,b) when compared against the proportion of wehrlite in spinel peridotite xenoliths in this study (Table S-3). This can be explained by the effect of oxygen fugacity (fO_2) on the stabilisation of carbonate/carbonated melt vs. graphite/diamond. Garnet and diamond are only stable at high pressures ≥~2 and ~3.5 GPa, depending on composition and the geothermal gradient to which their host rock equilibrated. The vast majority of garnet peridotite investigated to date have fO_2 relative to the Fayalite-Magnetite-Quartz (FMQ) buffer ≤ -1 (*e.g.*, Woodland and Koch, 2003; Yaxley *et al.*, 2017). However, solid carbonate or pure carbonatite melt are only stable at FMQ ≥ -1.5 to -1, depending on pressure (Stagno *et al.*, 2013). Carbonated silicate melts, which are stable to lower fO_2 precipitate graphite/diamond according to the reaction:

$$4\,FeO + CO_2 = 2\,Fe_2O_3 + C \quad \text{(Eq. S-1)}$$

which has been referred to as "redox freezing" (Rohrbach and Schmidt, 2011). Since fO_2 increases with decreasing pressure in the mantle lithosphere (*e.g.*, Woodland and Koch, 2003), pure carbonatite or carbonated silicate melt may be stable in the spinel peridotite facies. This may explain the higher abundances of wehrlite xenoliths at locations affected by extension.

Compositional Effects of Wehrlitisation and the Role of Lithosphere Thickness

The presence of a thick lithospheric lid, typically in intact cratons, impedes decompression melting, allowing no or only small-volume melts to be stabilised as a function of temperature, composition and fO_2 (*e.g.*, Aulbach, 2019). In contrast, thinner lithospheres allow longer melting columns to be established, with stronger dilution of the melt with silicate components (*e.g.*, Stamm and Schmidt, 2017). Consequently, disrupted cratons, the deep roots of which are thermally and compositionally strongly overprinted, and metacratons, the deep roots of which have been lost altogether, are typically associated with magmatism involving higher-volume melt (*e.g.*, Aulbach, 2019). Microstructures associated with garnet breakdown are described for some wehrlite-bearing xenolith suites (reviewed below), which suggests decompression to the spinel stability field in the context of extension and lithosphere thinning. Dilution with silicate components during sustained decompression melting entails higher contents of "basaltic" components in the melt, such as Al_2O_3 and FeO. Clinopyroxene in wehrlites from Greenland are characterised by low TiO_2 and FeO, but strongly elevated CaO/Al_2O_3 commonly ascribed to interaction with a carbonatite melt (Aulbach *et al.*, 2017), whereas those from Tok have high FeO and Al_2O_3 and in part TiO_2 contents, which are explained by reaction with silicate melt (Ionov *et al.*, 2005a) (Fig. S-2). Wehrlite-bearing xenolith suites from other localities show intermediate compositions (East African Rift, West Eifel Volcanic Field), which have been ascribed to carbonatites (Rudnick *et al.*, 1993) and alkaline mafic melts (Shaw *et al*., 2018), respectively. For the purpose of inferring the metasomatic agent based on reported clinopyroxene composition (Fig. S-2), we only consider garnet-free peridotites because the presence of garnet affects Al_2O_3 and Y-HREE partitioning into clinopyroxene, which we use to infer the metasomatic agent involved in wehrlitisation. This garnet effect is illustrated for trace elements in Figure S-3.

Interaction of mantle peridotite with carbonatite and silicate melt leads to a distinct minor- and trace-element relationships in the clinopyroxene: LREE tend to be strongly enriched over HREE in carbonatite-metasomatised peridotite while Ti is not enriched along with MREE, whereas the opposite results from interaction with silicate melt (*e.g.*, Yaxley *et al.*, 1991; Rudnick *et al.*, 1993; Coltorti *et al.*, 1999) (Fig. S-3). Elevated La/Gd, Ce/Yb and Pr and low Ti/Eu and Y result from interaction with carbonatite and/or with a melt that has percolated garnet-bearing mantle (Fig. S-3). Prior depletion related to partial melt extraction during stabilisation of the lithosphere is evident in low absolute Pr abundances. Thus, abundances of strongly incompatible elements decrease and of less incompatible elements increase with increasing volume of the metasomatic melt, and this signature is imposed on the metasomatised mantle volume. These systematics are also observed in clinopyroxene from peridotites other than wehrlite-group peridotites because cryptic metasomatism affects the incompatible element budget without changes in major element content and mineralogy (Dawson, 1984). In addition to this first-order effect of melt volume, the nature of the wall-rock with which the melt has interacted *en route* to the site of wehrlitisation strongly affects the compositional signature. For example, a kimberlite-like melt that extensively reacted with garnet-bearing mantle becomes increasingly Y- and HREE-poor and incompatible element-enriched (Aulbach *et al.*, 2013). When garnet-bearing mantle is absent, such melts have higher Y and La/Gd closer to the primitive mantle (Fig. S-3). Due to effects of lithosphere thickness, there is no single major- or trace-element characteristic associated with wehrlitisation.

Selection Criteria, and Assignation to Wehrlite-group Peridotites

Localities were chosen (1) that are considered representative with respect to tectonic settings (on- and off-craton, rift and basin), (2) for which the necessary data have been published that allow calculation of the CO_2 released during wehrlitisation (mineral modal abundances, clinopyroxene major-element compositions). Additional studies on wehrlite-bearing xenolith suites are available, but some are compositionally unequilibrated, precluding the use of average compositions or determination of mineral modes (*e.g.*, Eastern Australia: Yaxley *et al.*, 1991, 1998; Eger Graben: Loges *et al.*, 2019; Hoggar Swell: Kaczmarek *et al.*, 2016). Others are garnet-bearing (*e.g.*, Kimberley; Rehfeldt *et al.*, 2008), which complicates the interpretation of the metasomatic signatures as outlined in the previous section. Moreover, as ≤ 1% of garnets from the garnet and diamond-stable lithosphere are wehrlitic (Fig. S-2), they are not further considered here.

The wehrlites under consideration formed as a result of mantle metasomatism rather than accumulation from silica-undersaturated melt, such as those reported from the Wyoming Craton (Downes *et al*., 2004), and sometimes both types are present in a xenolith suite (*e.g.*, in the West Eifel Volcanic Field: Zinngrebe and Foley, 1995; East

African Rift: Davies and Lloyd, 1989). These two possibilities can be distinguished based on texture (sieved-textured clinopyroxene, absence of cumulate microstructures, textures showing replacement of orthopyroxene by clinopyroxene) and chemical composition (Na_2O in clinopyroxene >0.8 wt%, olivine Mg# >0.84; incompatible-element enrichment) (Lin et al., 2020).

A common classification scheme for ultramafic rocks is that by Streckeisen (1976), where peridotites with >40% olivine and <5% orthopyroxene are classified as wehrlites, whereas harzburgites with <5% clinopyroxene are produced by melt extraction from lherzolite. Melting reactions for spinel peridotite almost invariably indicate that clinopyroxene and orthopyroxene are consumed at a clinopyroxene/orthopyroxene ratio >1 (Walter, 2014). This produces olivine up to pressures of ~1.7 GPa, above which orthopyroxene may be produced, with estimates for clinopyroxene modes in the primitive mantle from 17 to 20% (Walter, 2014). Therefore, in melt residues, orthopyroxene/clinopyroxene ratios should be >1, whereas ratios <1 and clinopyroxene modes ≥20% may reflect wehrlitisation. Thus, clinopyroxene-rich, orthopyroxene-poor xenolith varieties that are classified as lherzolites according to Streckeisen (1976) were plausibly affected by the wehrlitisation process. In some xenolith suites, reaction dunites showing evidence for recent orthopyroxene-breakdown have major- and trace-element characteristics similar to wehrlites (*e.g.*, Mg# < olivine in primitive mantle) (*e.g.*, North Atlantic Craton in SW Greenland; Aulbach *et al.*, 2017). Such dunites are distinct from refractory melt extraction residues. They may form in channels preceded by reactive porous flow, as suggested for Fe-rich wehrlites (Raffone *et al.*, 2009), and they are here regarded as the (end) products of wehrlitisation under open-system conditions. As defined in the main text, wehrlite-group peridotites encompass Fe-rich "reaction" dunites, and orthopyroxene-poor lherzolites and harzburgites in addition to wehrlites.

Wehrlite-bearing Xenolith Suites and their Tectonic Setting

East African Rift (EAR), Tanzanian craton (disrupted)

Development of the EAR is associated with the Oligocene arrival of the Afar plume that caused widespread lithosphere thinning (Furman et al., 2016). The EAR stretches for some 6,000 km from north to south (Chorowicz, 2005). Samples from the Pleistocene Olmani and Labait cinder cones at and near the Tanzanian craton margin in the EAR comprise a total of 32 garnet-free peridotite xenoliths (Jones *et al.*, 1983; Rudnick *et al.*, 1993, 1994; Lee and Rudnick, 1999). 78 and 30%, respectively, of the garnet-free peridotite samples at these localities are wehrlites and reaction dunites (*i.e.* wehrlite-group peridotites; Table S-3). The latter show increased FeO, but inconspicuous Al_2O_3, CaO and TiO_2. Garnet-bearing peridotites also occur at Labait. Unusually low Al_2O_3 and high Ca/Al in clinopyroxene, the occurrence of monazite and apatite in some peridotites, as well as elevated Zr/Hf in peridotites from Olmani have been ascribed to carbonatite metasomatism (Rudnick *et al.*, 1993). Spinel peridotites from Labait, located at ~70 to 130 km depths record fO_2 of FMQ-0.5 to FMQ+0.4, *i.e.* they are oxidising relative to the Kaapvaal or Siberian craton at similar depth (Zhang *et al.*, 2017). A pre-entrainment enrichment event is recognised based on Li elemental and isotope systematics (Aulbach and Rudnick, 2009). Mid-lithospheric discontinuities related to melt metasomatism are detected at 60 to 100 km depth in the western craton (Wölbern *et al.*, 2012). The lithosphere-asthenosphere boundary (LAB) in the craton is at 150 to 200 km depth (Weeraratne *et al.*, 2003).

Tok, Aldan Shield, Siberian craton (disrupted)

Peridotite xenoliths from the Quaternary Tok volcanic field in the Aldan Shield at the SE Siberian craton margin were investigated by Ionov *et al.* (2005a,b, 2006). They comprise refractory, metasomatised lherzolites and wehrlites with elevated Ca, Fe and Ti contents (Fig. S-2), which have been ascribed to interaction with silica-undersaturated, alkali-rich silicate melt (Ionov *et al.*, 2005a). 21% of 48 xenoliths are wehrlites and orthopyroxene-poor lherzolites (Table S-1). The metasomatic agent ultimately formed from an underplated and fractionated basic melt, whereby carbonate-rich derivatives caused the strongest enrichments (Ionov *et al.*, 2006). Oxygen fugacities have not been reported for this mantle section. The Aldan shield was in an extensional regime in the early Cretaceous, with emplacement of alkaline rocks linked to the nearby Transbaikalian rift (Ivanov *et al.*, 2018). The Baikal rift, with a total of 2,000 km *en echelon* rift depressions (Tiberi *et al.*, 2003), records initial Late Cretaceous extension and increased activity in the Late Miocene to Early Pliocene, associated with subduction of the Pacific plate (Jolivet *et al.*, 2009). The associated lithospheric disruption resulted in reduced depth to the LAB (~140-190 km; Artemieva, 2006) compared to the intact Siberian craton (200 to 250 km; Priestley and Debayle, 2003).

North Atlantic Craton in SW Greenland (GNAC) (disrupted)

Forty-two metasomatised peridotite xenoliths from the Mesozoic Pyramidefjeld and Midternaes kimberlite comprise garnet-free lherzolites, harzburgites and reaction dunites from 100 to 170 km depth, and texturally equilibrated reaction dunites and olivine-rich phlogopite-bearing wehrlites from 90 to 110 km depth (Aulbach *et al.*, 2017) (Table S-1). Based on microstructural evidence for garnet break-down, the metasomatic overprint may be linked to the latest, Mesozoic, rifting event, which was preceded by multiple failed rifting episodes and accompanied by partial destruction of the lithospheric mantle (Tappe *et al.*, 2012). The intense metasomatism is proposed to have been oxidising based on the less compatible behaviour of V in wehrlites compared to other peridotites (Aulbach *et al.*, 2017). The high olivine modes also in the wehrlites (>80%) suggest an open-system process that precludes calculation of CO_2 fluxes through the lithosphere. The 20 km depth interval at 90 to 110 km appears to be nearly completely converted to wehrlite and related rocks (only one of 17 samples is lherzolite). Wehrlitic clinopyroxenes are characterised by very high CaO/Al_2O_3 (Fig. S-2), whereas trace elements in part show the influence of the garnet-bearing deeper lithosphere (Figs. S-3) (Aulbach *et al.*, 2017). Wehrlites and related rocks spatially overlap a seismically detected discontinuity with a negative phase (Kumar *et al.*, 2005). Garnet peridotites with equilibration pressures of up to 5.5 GPa (Nielsen *et al.*, 2008) indicate a minimum depth to the LAB of ~170 km, and a depth of ~210 km is seismically determined (Artemieva, 2019).

Tan Lu Fault Belt (TLFB), NE China (including decratonised North China Craton)

The TLFB experienced sinistral strike-slip in the earliest Cretaceous, followed by a brief period of compression and a long period of extension in the Early Cretaceous, related to subduction, slab roll-back and back-arc extension of the Palaeo-Pacific plate (Zhu *et al.*, 2018). Its total length and width are 5,000 × 800 to 1,000 km (Xu *et al.*, 1987). Numerous well-studied mantle xenolith suites comprising a high proportion of wehrlites were entrained during Late Cretaceous (Liaoyuan) to Cenozoic (Beiyan, Liaoyuan, Nushan, Shanwang, Yitong) alkaline magmatism (Table S-1). The wehrlites formed by interaction with silica-undersaturated CO_2-H_2O-bearing melts, based also on the presence, or evidence for the former presence, of amphibole (Lin *et al.*, 2020, and references therein). Signatures of wehrlitisation along the fault vary. Based on CaO-Al_2O_3-FeO relationships observed in clinopyroxene (Fig. S-2) 15 of 60 samples are inferred to have reacted with a carbonatite, nine with a carbonated silicate melt and the majority (n = 36) with a silica-undersaturated silicate melt (Table S-3). The proportion of wehrlites at each locality varies from none at Nushan to 75% at Liaoyuan (36 ± 30 1σ). Peridotites along the TLFB record oxygen fugacities of FMQ-2 to FMQ+0.8 at pressures of 1 to 2 GPa (Lin *et al*., 2020, and references therein). Regional oxidation and hydration, via influx of fluids sourced from the Palaeo-Pacific plate, may have remobilised CO_2 stored in the lithosphere by lowering the peridotite solidus (Geng et al., 2019). Development of the TLFB temporally overlaps the Mesozoic loss of the diamondiferous mantle root beneath the eastern North China craton (NCC) (Zhu *et al.*, 2018). Decratonization of the eastern NCC has been linked to seismic velocity reductions at 80 to 120 depth in the intact western NCC, which are interpreted as weak zones that were present also beneath the eastern NCC before the Mesozoic decratonisation (Chen *et al.*, 2014).

Middle Atlas, Morocco, NW Africa (reactivated Pan-African basement)

Late Pliocene to Quarternary alkaline volcanism (alkali basalts, basanites, nephelinites) in the Pan-African basement of the Middle Atlas is associated with the Trans-Moroccan fault system and entrained a diverse xenolith suite with variable amphibole contents (Raffone *et al.*, 2009). This suite, which records fO_2 of FMQ-0.1 to FMQ+1.8, comprises ~24% wehrlites that formed through interaction with alkaline melts (Fe-wehrlites) and subordinately carbonatite or highly evolved melts (Mg-wehrlites) (Raffone *et al.*, 2009) (Table S-1). One wehrlite and two clinopyroxene-rich lherzolites with clinopyroxene/orthopyroxene ratios of 1.7 (43%) out of seven peridotite xenoliths from the Azrou-Timahdite region are reported in Chanouan *et al*. (2017), confirming a high proportion of mantle affected by silica-undersaturated melt-metasomatism. Metasomatism is thought to have occurred in the Late Cretaceous or Eocene during tectonic reactivation (Raffone *et al.*, 2009; Wittig *et al.*, 2010). Reaction patches with secondary clinopyroxene and olivine, ascribed to interaction with asthenospheric alkali silicate melts, are reported for spinel lherzolites entrained in basalts some 1,500 km southeast, in Gharyan, Libya (Beccaluva *et al.*, 2008). These authors suggest that the Cenozoic volcanism in NW Africa reflects lithosphere rejuvenation and rifting within the Pan-African (or older) basement in reaction to the collision of the African and European plates.

Hoggar Swell, Algeria, NW Africa (reactivated Pan-African basement)

Some 1,000 km to the SSE of the Middle Atlas, in the Hoggar Swell, xenoliths from Neogene to Quarternary basanites and nephelinites show evidence for metasomatism by highly alkaline silicate melts to carbonate melts derived from the asthenosphere or rejuvenated lithosphere (Dautria *et al.*, 1992; Beccaluva *et al.*, 2007; Kourim *et al.*, 2014; Kaczmarek *et al.*, 2016). Kourim *et al.* (2014) report petrographic data for 28 peridotite xenoliths from the Tahalgha District, of which 7 are wehrlites (5 amphibole-bearing) (25%). Of 22 texturally heterogeneous samples from In Teria (Kaczmarek *et al.*, 2016), two are wehrlites and one is an olivine clinopyroxenite, as grouped together by the authors (14%) (Table S-1). There is evidence for the re-equilibration of spinel lherzolites from garnet-bearing mantle domains, which testifies to decompression (Beccaluva *et al.*, 2007). Rare garnet peridotites reported from other localities are interpreted as the relics of an original thicker lithosphere (Kaczmarek *et al.*, 2016). The Hoggar Swell adjoins the Saharan “metacraton” (500,000 km^2), which is transected by multiple megafaults and has a lithosphere thickness of ~100 to 150 km, compared to the adjacent West African craton with up to 250 km thickness (Liégeois *et al.*, 2003; Abdelsalam *et al.*, 2011). The intervening decratonised lithosphere underlying the Pan-African Swell, with a diameter of ~1,000 km (Liégeois *et al.*, 2005), is similarly vast.

West Eifel Volcanic Field (WEVF), Germany (off-craton)

The <1 Myr WEVF is located between the Upper and Lower Rhine Graben (part of the European Cenozoic Rift System (ECRS), with a total length of ~1,100 km; Ziegler and Dèzes, 2005). It belongs to the Central European Volcanic Province (Trieloff and Altherr, 2007). The underlying mantle is heterogeneous, with an abundance of wehrlites and orthopyroxene-poor harzburgites (Table S-1). Micaceous hornblendite veins in composite xenoliths attest to the action of hydrous melts before entrainment, whereas strongly LREE-enriched clinopyroxenes in the vein-hosting peridotite require earlier metasomatism (Witt-Eickschen *et al.*, 1998). Wehrlite and orthopyroxene-poor harzburgites (n = 4; 12%), of which one adjacent to a hornblendite vein, are described amongst 33 xenoliths from Dreiser Weiher, Meerfelder Maar and other localities, including two samples from the East Eifel (Witt-Eikschen *et al.*, 1998; 2003; Witt-Eickschen and O’Neill, 2005). Zinngrebe and Foley (1995) describe a suite of xenoliths from Gees with an abundance of wehrlites and clinopyroxene-dunites (n = 13; 68%) relative to other peridotites (n = 6). Abundant glass and unequilibrated textures preclude modal estimates. Twenty spinel peridotites from the Rockeskyllerkopf comprise 55% wehrlites and reaction dunites with abundant phlogopite and minor amphibole (not counting phlogopite-clinopyroxene veins), which were metasomatised by a silica-undersaturated alkaline melt before Quaternary magmatism (Shaw *et al.*, 2018). Witt-Eikschen and O’Neill (2005) obtain FMQ-0.2 ± 0.5 based on mineral equilibria. Diffusion modelling indicates that metasomatism occurred within <1 Myr of entrainment (Shaw *et al.*, 2018). Noble gas isotope systematics in mantle xenoliths from the WEVF and Pannonian Basin suggest that CO_2-rich fluids were trapped during mantle metasomatism from lithospheric and plume-derived sources (Trieloff and Altherr, 2007). Regional lithospheric thickness ranges from 50 to 120 km, after reworking of an originally thicker Variscan root (Ziegler and Dèzes, 2005). The ECRS was activated in the Late Eocene, by compression resulting from Alpine Pyrenean collision zones (Dèzes *et al.*, 2004).

Pannonian Basin, Hungary (off-craton)

The Pannonian Basin, wedged between the Alpine, Dinaride and Carpathian orogenic belts, experienced Early Miocene passive rifting with the formation of an extensional back-arc basin, followed by Late Miocene asthenospheric upwelling and active thinning in the central basin (Szabó *et al.*, 2004). Mantle xenoliths entrained in Neogene alkali basalts provide evidence for the reaction of the lithospheric mantle with carbonated melts. This includes direct observations of CO_2-bearing glass pockets in a wehrlite-bearing suite dominated by lherzolite and harzburgite, part of which show evidence for orthopyroxene break-down (Créon *et al.*, 2017). Modal abundances are not reported. Furthermore, CO_2-rich fluid inclusions with negative crystal shapes in clinopyroxene occur in lherzolites and harzburgites described by Berkesi *et al.* (2012). Two of nine xenoliths (22%) in their study are olivine-rich (>81.7% olivine) and orthopyroxene-poor (≤11.1 % orthopyroxene), and are assigned here to the wehrlite-group peridotites. Abundant wehrlite xenoliths (24% of 63 samples) entrained from ~1.2 to 1.6 GPa are found in the central part of the Nógrád-Gömör Volcanic Field in the northern Pannonian Basin (Patkó *et al.*, 2013, 2020; Liptai *et al.*, 2017) (Table S-1). Wehrlitisation occurred via interaction with a silicate melt similar to the host basalt, leading to increased Fe, Ti, Ca and Al contents and formation of amphibole (Patkó *et al.*, 2020). Lherzolites record fO_2 of FMQ-0.8 ± 0.7 (Patkó *et al.*, 2019). Of 22 samples entrained in Pliocene basalt from Balaton, some 200 km southwest (Ntaflos *et al.*, 2017), one clinopyroxene-rich dunite and one orthopyroxene-poor lherzolite (9%) appear to have interacted with silica-undersaturated melt. In the eastern basin, refertilisation by Carpathian-Pannonian-type subduction-related silicic melts preceded an alkaline event (Faccini et al., 2020), making it difficult to disentangle the effects of the latter, and this locality is therefore not considered here. Garnet breakdown products observed in some mantle xenoliths (Szabó *et al.*, 2004) suggest extension-related decompression of originally thicker lithosphere. The depth of the LAB is estimated at 70 to 100 km (Alasonati Tasárova *et al.*, 2016).

Southeastern Australia (off-craton)

Wehrlite xenoliths from SE Australia are compositionally unequilibrated (*e.g.*, Yaxley *et al.* 1991, 1998), precluding their use to estimate CO_2 flux described in the next section. Nevertheless, a more detailed description is provided here because the wehrlitisation reaction involving carbonatite was originally described in xenoliths from Victoria in southeastern Australia (Green and Wallace, 1988). Late Cretaceous to Holocene intraplate basaltic volcanism was widespread in SE Australia, and has been linked to the breakup of Gondwana and rifting of the Australian plate, followed by edge-driven convection that facilitated decompression melting (Oostingh *et al.*, 2016). The volcanics were emplaced into the Lachlan and Delamerian Fold Belts, which are crossed by a series of north-south-trending faults (Oostingh *et al.*, 2016), but a continent-scale rift or basin are absent. The faults currently provide pathways for predominantly mantle-derived CO_2 in SE Australian gas fields (Karolyte *et al.*, 2019). This is sampled in CO_2 springs, which are abundant in the Central Victorian Highlands, and in gas wells in the Otway Basin to the south (Karolyte *et al.*, 2019). The mantle CO_2 has been related to Pliocene to Recent basalts of the Newer Volcanic Province (Cartwright *et al.*, 2002), in which wehrlite-bearing xenolith suites occur.

CO_2 Liberation during Wehrlitisation: Modelling and Rationale

For the purpose of modelling the mass of CO_2 liberated during wehrlitisation, we consider only garnet-free and spinel peridotites, which sample the shallow mantle lithosphere where the decarbonation reaction takes place (Wallace and Green, 1988). Individual xenoliths with key characteristics are listed in Table S-1, key median compositions are shown in Table S-3. Based on the decarbonation reaction (main text), we calculate the mass of enstatite required to generate the additional diopside formed due to wehrlitisation (*i.e.* the difference between median clinopyroxene modes in wehrlite-group and “other peridotites”, weighted by median diopside component). No adjustment is made for modal abundances reported as weight vs. volume fractions, but for rocks dominated by mantle olivine and pyroxenes with similar densities (see, *e.g.*, Lee, 2003), the effect is minor (*e.g.*, 10.1 wt% clinopyroxene correspond to 10.0 vol.%) relative to uncertainties in the CO_2 degassing modelling. According to the reaction, the molar abundance of CO_2 liberated corresponds to ½ that of enstatite. This is then converted to mass of CO_2 liberated per 100 kg of wehrlitised peridotite and finally weighted by the proportion of wehrlite-group peridotites to calculate the mass of CO_2 liberated per 100 kg of peridotite in the lithosphere column. This result is independent of the CO_2 content in the metasomatic agent. For increasingly SiO_2-rich and correspondingly CO_2-poor liquids (corresponding to increasing melt fractions) higher volumes of melt, hence melt-rock ratios, are required to convert orthopyroxene to clinopyroxene. The likely metasomatic agent is inferred from the combined FeO and CaO/Al_2O_3 characteristics of wehrlites, which is evaluated on a suite-by-suite basis (Fig. S-2). As an example, for the TLFB, wehrlites with CaO/Al_2O_3 >6 and FeO <3.5 wt% are assigned to the carbonatite-metasomatised suite, those with CaO/Al_2O_3 ≤6 and FeO ≥ 3.5 wt% to the silicate melt-metasomatised suite, and the remainder to the carbonated silicate melt-metasomatised suite.

To calculate the total mass of CO_2 liberated, it is necessary to estimate the area and depth interval of lithosphere that was affected. Pressures for spinel peridotites are difficult to estimate accurately. We here conservatively assume that a 10 km lithosphere depth interval has been converted to wehrlite. Furthermore, we assume that xenolith localities sampling a portion of a rift system, continental-scale fault system or basin are representative of the entire system. Areal estimates are available for the Hoggar Swell (~785,000 km^2; Liégeois *et al.*, 2005), the Pannonian Basin (133,000 km^2) and the TLFB (4,500,000 km^2; Xu *et al.*, 1987, assuming a median width of 900 km). Brune *et al.* (2017) suggest, as a minimum, that rifts are 50 km wide. We use double this estimate because small-volume melt magmatism occurs on the shoulders of currently active rifts, such as the EAR, whereas this type of magmatism has been superseded by higher-volume basaltic melts in the rift itself (Foley and Fischer, 2017). For the 3250 km long Eastern Rift (Hunt *et al.*, 2017), this yields an area of 325,000 km^2, for the 2000 km long greater Baikal and Transbaikal rifted region 200,000 km^2 and for the 1100 km long ECRS (Ziegler and Dèzes, 2005) 110,000 km^2. Based on Figure 1 in Liégeois et al. (2005), the entire length and width of the area encompassing the Middle Atlas and

Gharyan in NW Africa are estimated at 2000 × 200 km, respectively.

To estimate the carbon flux, the duration of CO_2 liberation must be known. For the TLFB, Zhu *et al.* (2018) suggest a major extensional period in the Early Cretaceous following compression in the earliest Cretaceous and followed again by compression before the end of the Early Cretaceous, which is here taken to correspond to ca. 30 Ma. Although xenolith-bearing basalts along the TLFB are mainly Cenozoic, abundant wehrlites in the Late Cretaceous Liaoyuan basalts suggest a temporal link of wehrlitisation to main rift activity. Miocene passive to active rifting in the Pannonian Basin (Szabó *et al.*, 2004) translates to some 20 Ma of activity. Foley and Fischer (2017) estimate a 40 Ma lifespan for continental rifts, which is applied to the EAR. Activation of the ECRS in the Late Eocene (Dèzes *et al.*, 2004) may imply some 40 Ma of activity until today. Taking increased activity in the Late Miocene to Early Pliocene in the greater Baikal and Transbaikal rifted region (Jolivet *et al.*, 2009), we estimate a duration of 10 Ma. Based on isotopic compositions of xenoliths from the Hoggar Swell, metasomatism has been estimated to have occurred no earlier than 40 Ma prior to eruption. This timespan is adopted here for the Hoggar Swell and also the Middle Atlas, where magmatism and pre-entrainment metasomatic evolution has been linked to rifting of the Pan-African basement (Raffone *et al.*, 2009).

Supplementary Tables

Table S-1 Rock types, clinopyroxene modes and compositions for wehrlite-bearing xenolith suites from various localities, and clinopyroxene associated with carbonatitic high-density fluids in diamond (Excel file available for download from the online version of the article at http://www.geochemicalperspectivesletters.org/article2031).

Table S-2 List and salient compositions (wt. %) of high-density fluids in diamond and of experimentally produced liquids (Excel file available for download from the online version of the article at http://www.geochemicalperspectivesletters.org/article2031).

Table S-3 List and salient median compositions of clinopyroxene in wehrlite-bearing xenolith suites.

Locality/lithology	n	% wehr (1σ)	cpx mode %	FeO^{total} wt. %	CaO/Al_2O_3	Di mol
Labait and Olmani, Tanzanian Craton margin (disrupted), East African Rift						
Wehrlite-group	14	54 (34)	14	3.9	15.3	0.80
Other peridotites	18		3	2.7	9.2	0.76
Tok, Siberian Craton margin (disrupted), Aldan Shield						
Wehrlite-group	10	21 (20)	16	3.6	3.8	0.67
Other peridotites	37		5	2.7	4.1	0.68
Pyramidefjeld and Midternaes, North Atlantic Craton (NAC, disrupted) in SW Greenland						
Wehrlite-group	9	na	na	2.5	83.1	0.87
Other peridotites	15	na	na	3.1	9.5	0.78
Tan Lu Fault Belt, NE China (including decratonised North China Craton)						
Wehrlite - type 1	15		20	2.7	12.2	0.81
Wehrlite - type 2	9	36 (30)	25	4.9	9.2	0.78
Wehrlite - type 3	36		23	3.9	3.3	0.71
Other peridotites	156		11	2.8	3.1	0.69
Middle Atlas, Morocco, rifted Pan-African basement						
Wehrlite-group	7	34 (8)	24	3.6	4.3	0.70
Other peridotites	23		9	3.3	3.5	0.69
Hoggar Swell, Algeria, rifted Pan-African basement						
Wehrlite-group	7	16 (9)	18	4.9	3.3	0.66
Other peridotites	21		12	3.5	3.3	0.70
West Eifel Volcanic Field, Shoulder Rhine Graben, European Cenozoic Rift System (off-craton)						
Wehrlite-group	15	45 (29)	18	3.1	7.3	0.78
Other peridotites	38		5	2.7	4.4	0.75
Nógrád- Gömör+Bakony-Balaton, Pannonian Basin, Cenozoic European Basin System (off-craton)						
Wehrlite-group	19	18 (8)	21	4.0	4.1	0.70
Other peridotites	76		8	2.8	3.8	0.71

n number of samples (Table S-1), *% wehr (1σ)* average percentage of wehrlite-group peridotites in multiple xenolith suites and one standard deviation; average standard deviation from all other estimates is used for Tok, where wehrlite percentage is available from only one locality; *cpx mode* clinopyroxene modal abundance, *Di mole* diopside mole fraction in clinopyroxene; wehrlite-group includes wehrlite, orthopyroxene-poor lherzolite and harzburgite and reaction dunite, other peridotites includes harzburgites and lherzolites; type refers to wehrlitising agent as defined in Table 1; individual samples with references in Table S-1, further details in Supplementary Information.

Supplementary Figures

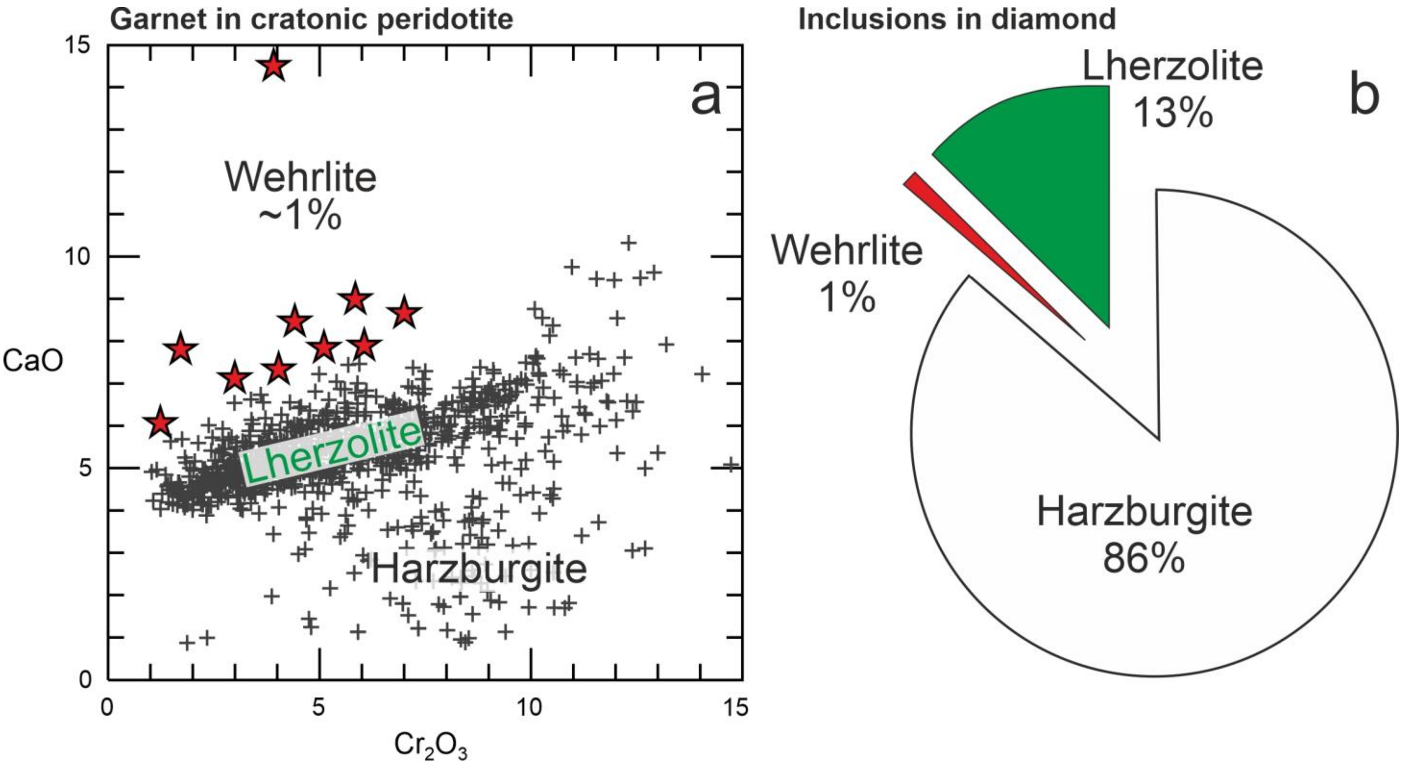

Figure S-1 **(a)** Wehrlitic garnet (red stars) and garnet in other peridotites (crosses) in >950 cratonic peridotite xenoliths (Boyd, 1974; Ehrenberg, 1982; Mitchell, 1984; Danchin and Boyd, 1976; Sobolev *et al.*, 1984; Hervig *et al.*, 1986; Winterburn *et al.*, 1990; Viljoen, 1994; Franz *et al.*, 1996; Boyd *et al.*, 1997; Stachel *et al.*, 1998; Kopylova *et al.*, 1999; MacKenzie and Canil, 1999; Schmidberger and Francis, 2001; Hearn, 2004; Kopylova and Caro, 2004; Menzies *et al.*, 2004; Grégoire *et al.*, 2005; Westerlund *et al.*, 2006; Aulbach *et al.*, 2007; Simon *et al.*, 2007; Creighton *et al.*, 2008, 2010; Hin *et al.*, 2009; Ionov *et al.*, 2010; Ivanic *et al.*, 2012), using the classification scheme of Grütter *et al*. (2004). **(b)** Parageneses of inclusions in diamond (Stachel and Harris, 2008).

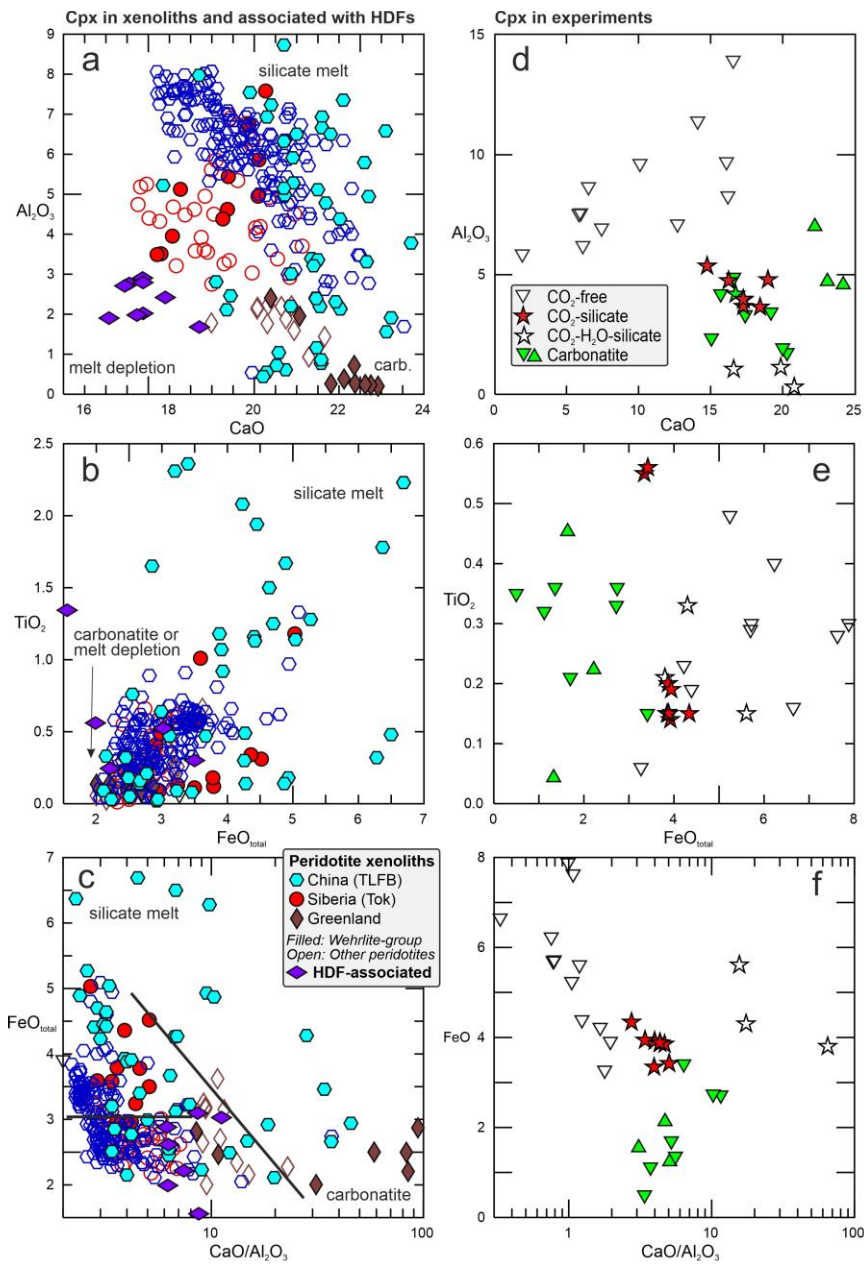

Figure S-2 Major-element relationships in clinopyroxene (cpx) from garnet-free xenolith suites (Table S-1) including wehrlite-group peridotites and other peridotites, as well as clinopyroxene occurring with diamond-hosted high-density fluids (HDF) **(a-c);** clinopyroxene or high-Ca pyroxene produced in experiments with variable amounts of $CO_2 \pm H_2O$ **(d-f)**. These relationships are used to illustrate varied effects of wehrlitisation depending on the nature of the metasomatic melt (silicate melt *vs.* carbonatite). For clarity, not all xenolith suites used in this study are shown. Depletion refers to melt extraction from peridotite. References: Tan Lu Fault Belt (TLFB): Xu *et al.* (1996, 1997, 1998), Zheng *et al.* (1998), Xu and Bodinier (2004), Hao *et al.* (2006, 2016), Yang *et al.* (2008), Liu *et al.* (2010), Xia *et al.* (2010), Xiao *et al.* (2010, 2013), Zhou *et al.* (2010), Lu *et al.* (2012), Wang *et al.* (2014), Lin *et al.* (2020); Aldan Shield in Siberia: Ionov *et al.* (2005a,b); North Atlantic craton in Greenland: Aulbach *et al.* (2017); clinopyroxene associated with HDF: Klein-BenDavid *et al.* (2009), Weiss *et al.* (2009, 2011, 2013, 2014), Weiss and Goldstein (2018). Experimental studies: Salters and Longhi (1999: open inverted triangles), Klemme *et al.* (1995: green triangles), Dasgupta *et al.* (2007: red stars, 2009: green inverted triangles), Girnis *et al*. (2013: open stars). Filled symbols denote wehrlite-group peridotites, open symbols denote lherzolites and harzburgites.

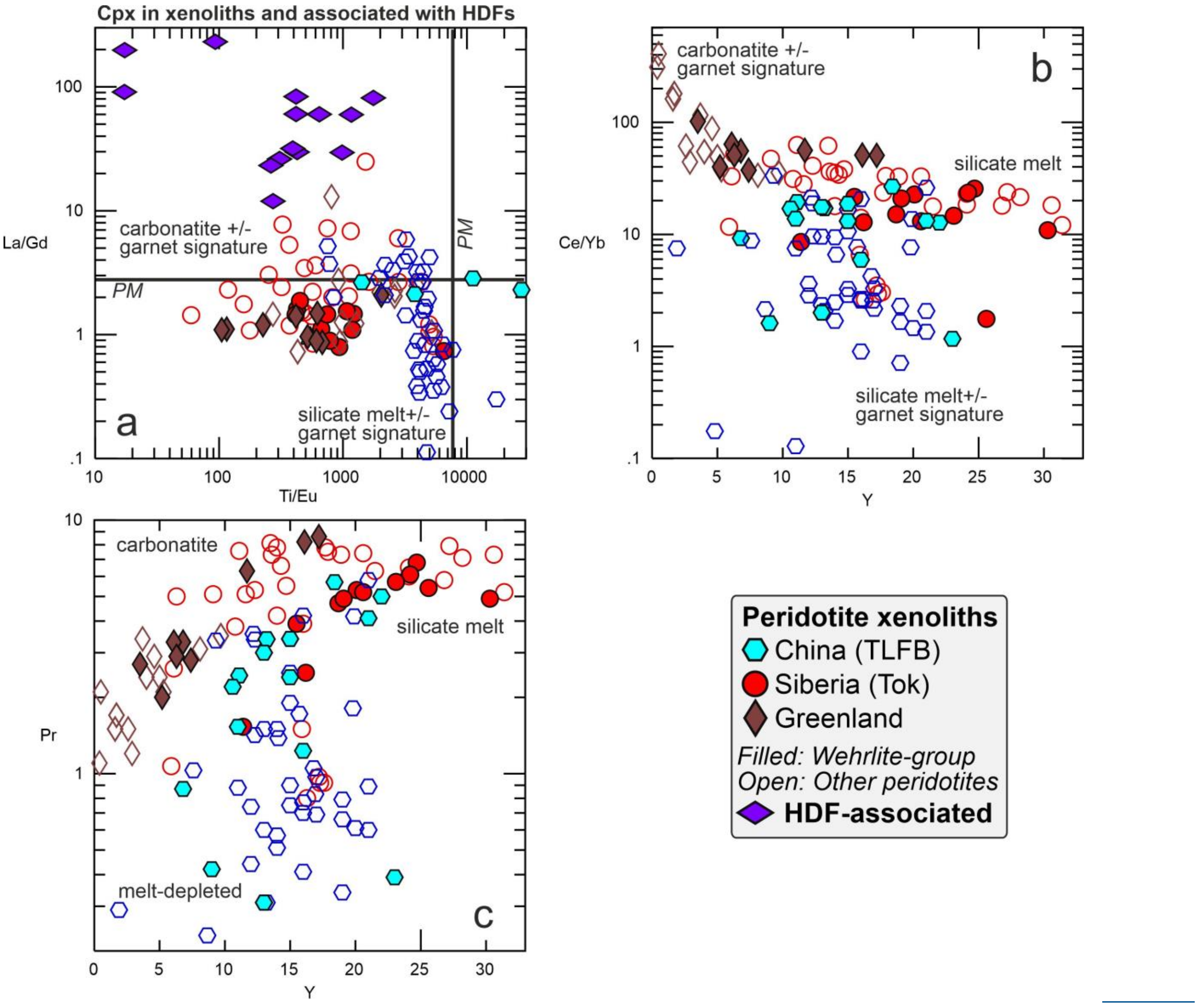

Figure S-3 **(a-c)** Trace-element relationships of clinopyroxene (cpx) in selected wehrlite-bearing peridotite xenolith suites reflecting processes and metasomatic agents as indicated in the panels, plus clinopyroxene associated with high-density fluids (HDFs) in diamond. References as in Figure S-2, Primitive Mantle PM of McDonough and Sun (1995).

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