On the chemical markers of pyroxenite contributions in continental basalts in Eastern China: implications for source lithology and the origin of basalts

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Abstract

Although it is widely recognized that mantle peridotite is compositionally heterogeneous, primitive mantle-like peridotite is commonly considered the potential peridotite source when identifying the source lithology of oceanic and continental basalts. Here, a simple equation that compatible with thermodynamic result ($Ol(wt\%) = 144.27 \ln((Fe+Mg+Ca+2Na)/Si)$, mol ratio) is introduced to distinguish peridotitic and pyroxenitic rocks. Meanwhile, the basic assumptions, alternative interpretations and uncertainties of many of the literature markers for pyroxenite contributions in basalts are briefly summarized, and some detailed comments on these markers are presented using literature data on olivines and basalts from Eastern China. Both high-CaO (>0.1wt%) and low-CaO (<0.1wt%) olivines in these continental basalts are likely generated by a xenocryst diffusive re-equilibration process, suggesting that the olivine chemistry method may not be suitable for the source lithology identification of the basalts. For the Cenozoic and Mesozoic continental basalts in Eastern China, nearly all of the markers for pyroxenite sources based on whole-rock major and/or trace elements might be compromised by refertilized/metasomatized peridotite sources. These uncertainties also exist for many other oceanic and continental basalts. However, the FC3MS ($FeO^T/CaO-3*MgO/SiO_2$, all in wt%) value, combined with those of Na_2O+K_2O , $Mg^\#$, MgO and La/Yb , suggests that Cenozoic OIB-type basalts in Eastern China are mostly primary pyroxenite-derived products, and caution should be paid when interpreting the petrological and geological implications of these basalts, particularly that of the low-MgO pyroxenite-derived basalts. At the normal mantle potential temperature (~1,350-1,400°C), the pyroxenite source can produce melts that explain all of the compositional characteristics of the Cenozoic basalts, suggesting that the origin temperature and pressure of the Cenozoic basalts determined by the traditional peridotite model are overestimated. For the Mesozoic basalts, compositionally diverse peridotite sources can account for their petrogenesis, but the pyroxenite source cannot be excluded. We acknowledge that the pyroxenite source may represent crustal materials (continental or oceanic crust) recycling and/or mantle metasomatism, which have important geodynamic implications, but further work is needed to determine the origin of the pyroxenite.

Keywords: basalt, olivine, peridotite, pyroxenite, xenocrysts, primary magma

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1. Introduction

Identification of the source lithology of basalts plays a fundamental role in our understanding of the magmatic processes and origin of mantle-derived magmas, enabling us to gain insights into crustal material recycling and/or mantle metasomatism that contribute to mantle heterogeneity. Recently, a variety of markers have been experimentally and theoretically proposed to infer pyroxenite-derived melt contributions in both continental and oceanic basalts (Hauri, 1996; Herzberg, 2006, 2011; Lambart et al., 2013; Le Roux et al., 2010; Liu et al., 2008; Sobolev et al., 2007; Sobolev et al., 2005; Yang and Zhou, 2013), but the source lithology identification of basalts is still a debated issue because all of the markers are largely influenced by the compositional diversity of the source rocks and melting and crystallizing conditions of the basaltic magmas (Herzberg et al., 2014; Lambart et al., 2013; Matzen et al., 2013; Niu et al., 2011; Putirka et al., 2011; Yang and Zhou, 2013). Although mantle peridotite and pyroxenite from various locations around the world show large variations in bulk composition (Downes, 2007; Kogiso et al., 2004; Lambart et al., 2013; Yang and Zhou, 2013) and a wide range of compositions of peridotite and pyroxenite, such as the experimental peridotite and pyroxenite compiled in Table 1 that may be the potential source rocks of both oceanic and continental basalts, PM-like peridotite and olivine-free pyroxenite are frequently used to model the source lithology of basalts (Herzberg, 2011; Sobolev et al., 2007; Sobolev et al., 2005). Most of the samples compiled in Table 1 come from the lithosphere, but the convecting mantle, where basaltic melts can be produced, may have similar compositional characteristics through either crustal material recycling or mantle metasomatism. In providing support for the pyroxenite source, experimental melts from peridotite are typically not given sufficient attention, and many refertilized/metasomatized peridotite melts are excluded (Lambart et al., 2013; Putirka et al., 2011). However, for the classical peridotite model, many potential pyroxenite source signatures in bulk-rock major and/or trace elements of OIB-type magmas can be illustrated by refertilized/metasomatized peridotite (Davis et al., 2013; Niu et al., 2011; Putirka et al., 2011). In addition, olivines crystallized from refertilized peridotite melts exhibit many chemical similarities compared

with olivine-poor or olivine-free pyroxenite melts (Herzberg et al., 2014). Consequently, the major issue of the identification of the source lithology of basalts is to distinguish refertilized/metasomatized peridotite-derived basalts from pyroxenite-derived basalts.

Intraplate continental basalts are widely distributed in Eastern China, and numerous geochemical and petrological studies have been carried out over the years (Basu et al., 1991; Chen et al., 2007; Zhi et al., 1990; Zou et al., 2000), with particularly high-quality data accumulated in recent years (Liu et al., 2015; Liu et al., 2008; Qian et al., 2015; Wang et al., 2012; Xu, 2014; Zeng et al., 2011). However, there is some debate about the source lithology of these basalts, which in turn introduces significantly different interpretations of their petrogenesis and geological implications (Huang et al., 2013; Liu et al., 2008; Niu et al., 2011; Wang et al., 2012; Xu et al., 2014; Yang and Zhou, 2013; Zhang et al., 2009). The major evidence supporting the pyroxenite source of these basalts is largely taken from those that are used to infer the source lithology of oceanic basalts (Gao et al., 2008; Hong et al., 2013; Liu et al., 2015; Liu et al., 2008; Qian et al., 2015; Wang et al., 2012; Wang et al., 2011; Xu et al., 2014; Zeng et al., 2011; Zhang et al., 2009), which may also suggest a refertilized/metasomatized peridotite source (Niu et al., 2011; Yang and Zhou, 2013; Guo et al., 2014; Guo et al., 2016). Thus, to determine the source lithology of the basalts from Eastern China, special attention should be paid to test whether refertilized/metasomatized peridotite source can explain their geochemical and mineralogical characteristics. This work aims to briefly summarize the chemical markers of the pyroxenite contribution in basalts and discuss their prerequisites, alternative interpretations and uncertainties, as well as provide some detailed comments on the chemical markers for the pyroxenite contribution in basalts from Eastern China. Finally, we present a pyroxenite model for basalts from Eastern China.

Table 1 Compositions of typical experimental peridotites and pyroxenites that may be the source materials of basalts.

	KLB-1	KR4003	PM	DMM	HZ	HPY	PHN1611	HK-66	KG2	KG1	MIX1G	M5-40	Px-1
SiO ₂	44.48	44.9	45.00	44.71	46.62	45.2	43.7	48.02	46.22	46.97	45.60	48.53	52.67
TiO ₂	0.16	0.16	0.20	0.13	0.08	0.71	0.25	0.22	0.57	0.78	0.90	0.52	0.64
Al ₂ O ₃	3.59	4.26	4.45	3.98	0.65	3.54	2.75	4.88	7.69	9.75	15.20	12.37	11.26
Cr ₂ O ₃	0.31	0.41	0.38	0.57	0.57	0.43	0.28	0.25	0.22	0.17	0.11	0.12	0.25
FeOT	8.1	8.02	8.05	8.18	4.91	8.47	10.05	9.9	9.22	9.77	7.80	9.02	7.55
MnO	0.12	0.13	0.14	0.13	0.15	0.14	0.13	0.14	-	-	0.15	0.20	0.12
MgO	39.22	37.3	37.80	38.73	46.19	37.5	37.22	32.35	28.83	23.57	16.67	16.64	18.48
CaO	3.44	3.45	3.55	3.17	0.73	3.08	3.26	2.97	6.05	7.35	11.48	10.89	7.05
Na ₂ O	0.3	0.22	0.36	0.13	0.05	0.57	0.33	0.66	1.11	1.52	1.40	1.65	1.52
K ₂ O	0.02	0.09	0.03	0.01	0.03	0.13	0.14	0.07	0.09	0.12	0.04	0.06	0.06
NiO	0.25	0.24	0.25	0.24	-	0.2	0.25	0	-	-	-	-	0.13
P ₂ O ₅	0.03	-	0.02	0.02	-	0.04	-	0.07	-	-	0.01	-	-
Total	100.02	99.18	100.23	100.00	99.98	100.01	98.36	99.53	100.00	100.00	99.37	100.00	99.73
Ol(wt%)	66	57	60	62	67	60	65	37	41	28	11	5	-15
Mg#	89.6	89.2	89.3	89.4	94.4	88.8	86.8	85.3	84.8	81.1	79.2	76.7	81.4
Fe/Mn	67.8	61.9	59.9	63.2	32.9	60.7	77.6	71.0	-	-	52.1	45.3	63.7
Zn	-	-	55	-	-	-	81	-	-	-	-	-	-
Zn/Fe	-	-	8.79	-	-	-	10.37	-	-	-	-	-	-
La	-	-	0.69	0.192	-	-	0.89	-	-	-	-	-	-
Yb	-	-	0.49	0.365	-	-	0.33	-	-	-	-	-	-
La/Yb	-	-	1.39	0.53	-	-	2.70	-	-	-	-	-	-

KLB-1, Hirose and Kushiro (1993). KR4003, Walter (1998). PM, McDonough and Sun (1995). DMM, Workman and Hart (2005). HZ, Falloon and Danyushevsky (2000). HPY, Falloon et al. (1988). PHN1611, Kushiro(1996). HK-66, Hirose and Kushiro (1993). KG2 and KG1, Kogiso et al. (1998). MIX1G, Hirschmann et al. (2003). M5-40, Lambart et al. (2009). Px-1, Sobolev et al. (2007).

86 2. Relationship between the bulk composition and lithology of mafic and ultramafic 87 rocks

88 Refertilized/metasomatized peridotite may occasionally become olivine-poor or olivine-free
89 pyroxenite. Before identifying the source lithology of basalts, it is useful to find a relationship between the
90 bulk composition and lithology of mafic and ultramafic rocks. From a petrological perspective, peridotite
91 and pyroxenite are lithologically controlled by the relative phase proportions of pyroxene versus olivine,
92 and they are thermodynamically controlled by the bulk composition, temperature and pressure.
93 Pyroxene and olivine are the two major rock-forming minerals that commonly used to distinguish
94 peridotite from pyroxenite. Garnet could control the major-element composition of both peridotite- and
95 pyroxenite-derived melts. However, garnet-pyroxenite will become spinel-pyroxenite under low pressure,
96 thus the major difference between peridotite and pyroxenite should be pyroxene versus olivine.
97 Peridotite and pyroxenite can have the same mineral assemblage, but peridotite have >40% olivine in
98 volume and pyroxenite have <40% olivine in volume. Although it is better to distinguish peridotite from
99 pyroxenite based on the modal mineralogy when they equilibrate at a specific pressure and temperature,
100 it is generally believed that peridotite has more than 30-35 wt% MgO and that pyroxenite has less than
101 30-35 wt% MgO ([Downes, 2007](#); [Kogiso et al., 2004](#); [Lambart et al., 2013](#); [Yang and Zhou, 2013](#)) at the
102 typical upper mantle temperatures and pressures under which basaltic magmas are commonly produced.
103 However, the MgO content is not a good indicator of the lithology because pyroxenite may have a high
104 MgO content, such as orthopyroxenite, which will have 40.1 wt% MgO if it consists of enstenite, whereas
105 peridotite may have a low MgO content, such as dunite, which will have 23.4 wt% MgO if it consists of
106 Fo₅₀ olivine (FeMgSiO₄). The classical division between a pyroxenite and peridotite is based on the
107 relative proportions of olivine versus pyroxene; however, temperature and/or pressure, as well as other
108 phases, such as spinel, garnet and Fe-Ti oxide, could also affect the relative proportions of olivine
109 versus pyroxene. For typical pyroxenite, olivine-bearing spinel pyroxenite may become olivine-free
110 garnet pyroxenite at elevated pressures ([Kogiso et al., 2003](#)). Moreover, garnet peridotite will have an
111 elevated olivine/pyroxene ratio with increasing pressure because of the expanded stability field of garnet
112 at the expense of pyroxene ([Walter, 1998](#)). This suggests that the lithology can only be safely
113 determined for a given bulk composition at a specific temperature and pressure. However, the
114 temperature and pressure are generally not easily obtained, and basaltic magmas are typically produced
115 at a variety of temperatures and pressures. Thus, given that the bulk composition is typically used to

116 represent the source rocks of basalts, it is better to find a simple compositional parameter that has only
 117 slight temperature and/or pressure effects to distinguish olivine-poor/olivine-free pyroxenite from
 118 olivine-rich peridotite.

119 The major end-member components of pyroxene, i.e., Wo($\text{Ca}_2\text{Si}_2\text{O}_6$), En($\text{Mg}_2\text{Si}_2\text{O}_6$), Fs($\text{Fe}_2\text{Si}_2\text{O}_6$),
 120 Di($\text{CaMgSi}_2\text{O}_6$), Jd($\text{NaAlSi}_2\text{O}_6$), CATS($\text{CaAl}_2\text{SiO}_6$), and garnet (Ca, Mg, Fe) $_3\text{Al}_2\text{Si}_3\text{O}_{12}$, all have
 121 $(\text{Fe}+\text{Mg}+\text{Ca}+2\text{Na})/\text{Si}=1$ (mol ratio). Olivine Fo (Mg_2SiO_4) and Fa (Fe_2SiO_4) have
 122 $(\text{Fe}+\text{Mg}+\text{Ca}+2\text{Na})/\text{Si}=2$ (mol ratio). Therefore, the relative proportions of olivine versus pyroxene and
 123 garnet for a given bulk composition can largely be determined by the $(\text{Fe}+\text{Mg}+\text{Ca}+2\text{Na})/\text{Si}$ ratio. Here,
 124 we propose a simple empirical equation that $\text{Ol}(\text{wt}\%)=144.27\ln((\text{Fe}+\text{Mg}+\text{Ca}+2\text{Na})/\text{Si})$, which can be
 125 used to discriminate peridotite from pyroxenite for a given bulk composition (Fig. 1a). The olivine
 126 contents calculated by this simple equation are in good agreement with those estimates of abyssal
 127 peridotite based on whole-rock CIPW norm calculation (Niu, 1997), mass balance calculation (Chu et al.,
 128 2009) and of experimental peridotite and pyroxenite based on pMELTS calculations (Ghiorso et al., 2002)
 129 (Fig. 1a and Supplementary Fig. 1), although our model olivine phase involves spinel and Fe-Ti oxides.
 130 Our model olivine content for a given bulk composition is generally slightly higher than its real content
 131 (supplementary Fig. 1). Silica-excess and -deficient pyroxenite have $(\text{Fe}+\text{Mg}+\text{Ca}+2\text{Na})/\text{Si}<1$ and >1 ,
 132 respectively, and they can all become olivine-bearing pyroxenite at low pressure and olivine-free
 133 pyroxenite at high pressure.\

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135

136 **Fig. 1** Olivine modal abundance (wt%) as a function of $(\text{Fe}+\text{Mg}+\text{Ca}+2\text{Na})/\text{Si}$ (mol) in (a) and relationship between
 137 calculated olivine modal abundance and MgO (wt%) in (b). Abyssal peridotite from Niu (1997). Xenoliths (peridotite and
 138 pyroxenite) in basalts from Chu et al. (2009). Experimental peridotite and pyroxenite from the data compiled by Yang and

139 [Zhou \(2013\)](#). Detailed phase proportions of the representative experimental peridotite and pyroxenite shown in (a), see
140 Table S7 of [Yang and Zhou \(2013\)](#). The DMM+MORB compositions are the same as those from [Kogiso et al. \(1998\)](#). The
141 KR4003+dacite melt compositions are the same as those from [Herzberg et al. \(2014\)](#). Negative olivine contents in (b)
142 indicate $(\text{Fe}+\text{Mg}+\text{Ca}+2\text{Na})/\text{Si}<1$

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144 The relationship between the olivine modal abundance calculated by
145 $\text{Ol}(\text{wt}\%) = 144.27 \ln((\text{Fe}+\text{Mg}+\text{Ca}+2\text{Na})/\text{Si})$ and the bulk MgO content suggests that both peridotite and
146 pyroxenite show large variations in the MgO content with a constant olivine content ([Fig. 1b](#)). Our model
147 suggests that if less than 30 wt% MORB is mixed with DMM peridotite, the mixed product will have ~40%
148 olivine and can be called peridotite ([Fig. 1b](#)). TiO_2 enrichment in ocean island basalts requires only
149 ~1-10% MORB mixed with DMM peridotite ([Prytulak and Elliott, 2007](#); [Putirka et al., 2011](#)), suggesting
150 that TiO_2 enrichment in basalts does not exclusively suggest a pyroxenite source from the lithological
151 perspective. When the average amount of MORB added to the peridotite reaches approximately 70%,
152 the mixture becomes nearly olivine-free pyroxenite, similar to the experimental pyroxenites MIX1G and
153 M5-40 ([Fig. 1b](#)). Pyroxenite may also be produced if acid melts are mixed with peridotite, and as shown
154 in [Fig. 1b](#), ~10% dacite A200K ([Pertermann and Hirschmann, 2003](#)) added to peridotite will produce an
155 olivine pyroxenite. When the amount of dacite added to peridotite reaches ~40%, the mixture becomes
156 olivine-free pyroxenite, consistent with the conclusion of [Herzberg et al. \(2014\)](#). It worth noting that the
157 mafic pyroxenite is often confused with the ultramafic pyroxenite in discussion on the origin of OIB-like
158 basalts. The former is commonly in excess of silica and thus a kind of mafic eclogites, whereas the latter
159 is deficient in silica and often represents ultramafic metasomatites or cumulates. The silica-excess mafic
160 pyroxenite such as MORB typically depleted in melt-mobile incompatible elements and produce acid
161 melts that cannot be source materials of OIB-like basalts. In contrast, ultramafic metasomatites in the
162 forms of either refertilized peridotite or pyroxenite would have trace elements enriched source that can
163 be a better candidate of the source of OIB-like basalts ([Niu and O'Hara, 2003](#)). Our current simple
164 method is still arbitrary from a strict petrological perspective, but to our knowledge, it provides the
165 simplest direct estimate of the olivine modal abundance of any given mafic and ultramafic bulk
166 compositions and can thus help us to understand the lithological variation of the source materials of
167 basalts.

168 **3. Uncertainties and alternative interpretations of literature markers for the** 169 **contribution of pyroxenite in basalts**

170 Nearly all of the commonly used markers for pyroxenite contribution in basalts and their basic
 171 assumptions, alternative interpretations and uncertainties are briefly summarized in [Table 2](#), and some
 172 detailed rigorous comments on most of these markers are presented below based on literature data for
 173 olivines and basalts from Eastern China. Some new alternative interpretations of pyroxenite source
 174 signatures of these continental basalts are explored, which may have important implications for source
 175 lithology and the origin of basalts.

Table 2 Major markers of pyroxenite contribution in basalts and their alternative interpretations

Marker	Assumed source	peridotite	Alternative interpretations and/or potential uncertainties ^l	Potential peridotite source ^k
Melts with Low CaO at a given MgO content ^a	PM-like (KR4003)	peridotite	1, Alkaline-rich melts. 2. Metasomatized/refertilized peridotite partial melts. 3, Harzburgite melts. 4, Clinopyroxene fractionation.	PHN1611, HPY, HK-66, MORB+DMM, MORB melts+PM, HZ, PS6.1, KPS2.1, KPS3.2, KPS4, MBK, MBK+
Olivine with low Ca, Mn and high Ni, Fe/Mn at a given Fo ^b	PM and DM		1, Temperature effect. 2, Magma mixing. 3, Xenocryst diffusive re-equilibration. 4, Olivine crystallized from low CaO peridotite melts or from metasomatized/refertilized peridotite melts. 5, Clinopyroxene fractionation.	PHN1611, HPY, MORB+DMM, MORB melts+PM, PS6.1, KPS2.1, KPS3.2, KPS4, MBK, MBK+
Melts with high Fe/Mn ^c	PM-like (KR4003)	peridotite	1, Temperature and pressure effect. 2, High Fe/Mn (iron-rich) peridotite source. 3, Clinopyroxene fractionation.	PHN1611, HK-66, KLB-1, MORB melts+PM
Melts with high Zn/Fe ^d	PM-like peridotite		1, Pressure effect. 2, High Zn/Fe peridotite source. 3, Clinopyroxene fractionation.	PHN1611, MORB melts+PM
Melts with high TiO ₂ ^e	PM and DM		TiO ₂ -rich peridotite	PHN1611, HPY, MORB+DMM, MORB melts+PM
Melts with low SiO ₂ and high FeO ^f	Volatile-free DMM1		1, Carbonated peridotite. 2, Iron-rich peridotite source	PHN1611, HK-66, KLB-1+2.5 wt% CO ₂ , PERC, PERC3,
Melts with high CaO/Al ₂ O ₃ ^g	Volatile-free peridotite		Carbonated peridotite	KLB-1+2.5 wt% CO ₂ , PERC, PERC3,
Melts with high SiO ₂ ^h	PM-like peridotite		Harzburgite melts	HZ
Melts with high FC3MS value ⁱ	MgO>30wt%		1, Extremely low CaO harzburgite melts. 2, Clinopyroxene fractionation.	HZ

176 a Herzberg (2006)
 177 b Sobolev et al. (2005, 2007), Herzberg (2011)
 178 c Herzberg (2011)
 179 d Le Roux et al. (2010)
 180 e Prytulak and Elliott (2007)
 181 f, g Lambart et al. (2013)

h Hauri (1996)
i Yang and Zhou (2013)
^J For detailed discussions of the alternative interpretations and/or potential uncertainties, see the text and Yang and Zhou (2013).
^k Carbonated peridotite PERC and PERC3 from (Dasgupta et al., 2007), KLB-1+2.5 wt% CO₂ from (Hirose, 1997). K₂O-rich peridotite PS6.1, KPS2.1, KPS3.2 and KPS4 from (Davis and Hirschmann, 2013), and MBK and MBK+ from (Laporte et al., 2014). Other peridotite sources are the same as in Table 1.

3.1. Whole-rock CaO-MgO relations

It has been argued that at a given MgO content, pyroxenite can produce a melt with a lower CaO content than a peridotite melt (Herzberg, 2006; Herzberg and Asimow, 2008), which has been widely used to identify the source lithology of basalts (Fig. 2) (Gao et al., 2008; Hong et al., 2013; Liu et al., 2015; Liu et al., 2008; Nikulin et al., 2012; Timm et al., 2009; Zeng et al., 2011). However, this argument holds only for peridotite KR4003; KLB-1 melts if the solidus is at or above 3 GPa, which can be easily compromised by peridotite compositional diversity and magma mixing (Putirka et al., 2011; Yang and Zhou, 2013). The CaO-MgO relation was originally used by Herzberg (2006) to argue for a pyroxenite source for Hawaiian tholeiite. For alkalic melts, peridotite can produce low-CaO melts, as emphasized by Herzberg (2006) in his supplementary materials. However, this alternative interpretation was not considered in many subsequent studies, and numerous alkaline basalts are inappropriately plotted in the CaO-MgO diagram to argue for a pyroxenite source (Gao et al., 2008; Hong et al., 2013; Liu et al., 2015; Liu et al., 2008; Nikulin et al., 2012; Timm et al., 2009; Zeng et al., 2011). More importantly, because CaO content in peridotite melts has a significantly negative correlation with Na₂O+K₂O content (Supplementary Fig. 2), this simple correlation results in a significantly negative correlation between CaO-13.81+0.274*MgO and Na₂O+K₂O (Fig. 2). The experimental melts of peridotite KR4003 (Walter, 1998) and near-solidus melt of peridotite KLB-1 (Davis et al., 2011) with <3.5% Na₂O+K₂O and their CaO-13.81+0.274*MgO>0 are consistent with the argument that peridotite produces high-CaO melts. However, low-Na₂O+K₂O harzburgite melts and high-Na₂O+K₂O lherzolite melts, including tholeiitic and alkaline basalts, can have low CaO contents and CaO-13.81+0.274*MgO<0 (Fig. 2). Recent experimental petrology suggests that K₂O-rich metasomatized garnet peridotite at 3 GPa produces melts with Na₂O+K₂O=4.3-7.1wt% and CaO-13.81+0.274*MgO=-0.13~-2.08 (Davis and Hirschmann, 2013). This seems suggest that only K₂O rich low CaO alkaline basalt can be considered as the production of peridotite melting, but Na₂O rich basaltic melts can be produced by peridotite melting at low pressure (Falloon et al., 1988; Hirose and Kushiro, 1993; Laporte et al., 2014) which may mixed with high pressure peridotite melts to produce Na₂O rich low CaO basaltic melts (Zeng et al., 2013). Moreover,

Small fraction MORB-eclogite melts reacted with normal peridotite, when equilibrium with peridotitic mineral assemblage, can produce Na₂O rich low CaO basaltic melts at garnet stable pressures (Mallik and Dasgupta, 2012). Thus, Na₂O rich low CaO basaltic melts are likely produced by Na₂O-rich metasomatized garnet peridotite melting at high pressure. Therefore, the vast majority of basalts that have more than 3 wt% Na₂O+K₂O that have been considered as pyroxenite melts using the CaO-MgO relation also can be considered peridotite melts. Before using the CaO-MgO relation to infer the source lithology of basalts, the Na₂O+K₂O content must be considered. Additionally, olivine phenocrysts crystallized from these low-CaO and high-Na₂O+K₂O melts will inherit their parental magma characteristics and will have lower CaO contents relative to the typical high-CaO peridotite melts (Supplementary Fig. 3).

Fig. 2 CaO-MgO relation and $\text{CaO}-13.81+0.274*\text{MgO}$ (Herzberg and Asimow, 2008) versus Na₂O+K₂O of experimental peridotite melts and natural basalts. Peridotite melts with MgO < 16 wt% and C-OIB (Ocean island basalt-like basalts in Eastern China and Mongolian) with MgO > 7.5% from data compiled by Yang and Zhou (2013). Peridotite KLB-1 3 GPa near-solidus melts from (Davis et al., 2011). Peridotite KR4003 3-7 GPa melts from (Walter, 1998). Harzburgite 1.5-2.5 GPa melts from (Falloon and Danyushevsky, 2000). Peridotite+1% and +2% CO₂ melts from (Dasgupta et al., 2007; Dasgupta et al., 2013). Cenozoic and Mesozoic basalts from Eastern China that have been considered as pyroxenite-derived low-CaO melts from (Gao et al., 2008; Hong et al., 2013; Liu et al., 2015; Zeng et al., 2011). C-OIB (MgO > 7.5 wt%), Continental OIB-type basalts from Eastern China and Mongolia compiled by Yang and Zhou (2013). As CaO in olivine is highly incompatible, CaO-MgO relation of natural basalts would not significantly change when olivine-addition method is applied for primary magma correction. Thus, for basalts with MgO > 7.5~10%, olivine fractionation and accumulation would not change the results in identification of source lithology of natural basalts using the CaO-MgO relation.

239

240 **3.2. Ni and Ca in olivine**

241 Recently, the olivine phenocryst composition has been widely used to detect the source lithology of
242 basalts (Herzberg, 2011; Herzberg et al., 2014; Sobolev et al., 2007; Sobolev et al., 2005). Many
243 olivine/liquid distribution models were obtained from parameterizations of experimental data (Beattie et
244 al., 1991; Hart and Davis, 1978; Li and Ripley, 2010; Matzen et al., 2013; Niu et al., 2011; Putirka et al.,
245 2011). However, the Beattie (Beattie et al., 1991) model provides the best equation for describing the Ni
246 partition between the olivine and liquid, with the lowest prediction error when considering all
247 experimental data (Herzberg et al., 2013; Herzberg et al., 2014). This suggests that the Ni partition
248 coefficient of olivine is only controlled by the melt composition and has no temperature or pressure effect;
249 however, recent high-quality experimental results on a liquid with a MgO content of ~18% suggest that
250 the temperature effect can significantly influence olivine Ni partitioning, which would successfully explain
251 the Hawaiian high-Ni olivine phenocrysts (Matzen et al., 2013). This suggests that the temperature effect
252 is still an alternative interpretation of high-Ni olivine. However, more high-quality experimental work is
253 needed to quantify the effect of temperature on olivine Ni partitioning for liquids with various MgO
254 contents.

255 The chemical features of olivine crystals occurring in basalts from Eastern China have also been
256 used to infer a pyroxenite source (Gao et al., 2008; Hong et al., 2013; Liu et al., 2015; Qian et al., 2015;
257 Wang et al., 2012; Wang et al., 2011). However, whether the chemical signals result from the melt
258 composition or are simply characteristics of mantle olivine was not adequately resolved, as discussed
259 below. Many Mesozoic and Cenozoic basalts from Eastern China contain varying amounts of olivine
260 xenocryst that disaggregated from mantle peridotite (Chu et al., 2013; Fan and Hooper, 1991; Gao et al.,
261 2008; Qian et al., 2015; Sakuyama et al., 2013; Wang et al., 2012), which were identified mainly based
262 on their CaO content of <0.1 wt%. However, olivine from magmatic cumulates and low-CaO melts may
263 also contain <0.1 wt% CaO (Kamenetsky et al., 2006; Li et al., 2012). Mantle olivines can also be
264 distinguished from magmatic olivines by other minor elements such as Ti and Cr (Foley et al., 2013), but
265 Ti and Cr contents are too low to precisely determine, which may be used to identify origin of olivine
266 when high quality data obtained for basalts in Eastern China. Moreover, textural characteristics, such as
267 resorption and kink-bands, that have been taken to imply a xenocrystic origin may be produced from
268 magmatic processes (Erdmann et al., 2014; Welsch et al., 2013). Thus, it is generally difficult to

269 determine the origins of the olivine crystals that commonly occur in basalts. Here, we do not intend to
 270 determine the origins of the olivine that occur in basalts from Eastern China; instead, we present an
 271 alternative interpretation of their chemical characteristics.

272 Chemical zoning, typically representing elemental diffusion and re-equilibration processes, is a
 273 common feature of olivine. The diffusion patterns of olivine from natural samples and experimental work
 274 suggest that olivine cores generally have high Fo and Ni contents and low Ca and Mn contents. The
 275 Mg-Fe diffusion zone is typically comparable with that of Ni but is generally wider than those of Ca and
 276 Mn because of their different diffusion coefficients (Costa et al., 2008; Costa and Dungan, 2005; Qian et
 277 al., 2010). For mantle olivine xenocrysts present in typical basaltic rocks, the diffusion characteristics of
 278 Fo, Ni and Ca can be simply illustrated as in Fig. 3a. Accordingly, several CaO-Fo and NiO-Fo
 279 co-variation trends can be easily obtained from a qualitative perspective (Fig. 3b, c).

280

281

282 **Fig. 3** Mantle olivine xenocryst diffusive re-equilibration model compared with olivine literature data from Eastern China. (a)
 283 Compositional distribution patterns of Fo, Ni and Ca and of Ni-Fo and Ca-Fo for a typical olivine xenocryst that has been
 284 re-equilibrated with its hosting melt. (b) and (c) CaO versus Fo and NiO versus Fo for olivine from Eastern China. Jiangsu
 285 and Anhui olivine from (Wang et al., 2011). Hannuoba olivine from (Qian et al., 2015). Hainan olivine from (Liu et al., 2015;

286 Wang et al., 2012). Xindian olivine from (Hong et al., 2013). Feixian and Sihetun olivine from (Gao et al., 2008). Peridotite
287 olivine and calculated peridotite KLB-1 (Hirose and Kushiro, 1993) olivine was equilibrated at 2 GPa at various
288 temperatures using the pMELTS algorithm (Ghiorso et al., 2002). Olivine crystallized from peridotite primary magmas (8
289 wt%<MgO<38 wt%) and L+Ol, L+Ol+Cpx and L+Ol+Cpx+Pl trends adopted from (Herzberg et al., 2014).

290

291 The model CaO-Fo and NiO-Fo co-variation trends of typical mantle olivine xenocrysts that have
292 been re-equilibrated with their host low-MgO melts can successfully explain the chemical characteristics
293 of most low-CaO olivines in basalts from Eastern China (Fig. 3b). The olivine data provided by Wang et
294 al. (2011) and Qian et al. (2015) show consistent results, and they are in good agreement with the
295 chemical diffusion trends between typical mantle olivine xenocrysts and the low-MgO basaltic melts.
296 Because the CaO diffusion rate is commonly slower than those of Ni and Fe-Mg (Costa et al., 2008;
297 Costa and Dungan, 2005; Qian et al., 2010), mantle olivine will typically have <0.1 wt% CaO even when
298 Fo decreases to 85. This diffusive re-equilibration model can explain why some slightly high-CaO
299 olivines (>0.1 wt%), which have been considered as magmatic olivines from a pyroxenite source, have a
300 low Fo and high Ni content. However, this alternative explanation is underestimated, and only olivines
301 with <0.1 wt% CaO are considered mantle xenocrysts (Gao et al., 2008; Liu et al., 2015; Qian et al.,
302 2015; Wang et al., 2012). In contrast, olivines from Hainan, Sihetun and Feixian basalts exhibit a large
303 variations in CaO content, ranging from ~0% to 0.2%, even when Fo>90 (Fig. 3a). Although olivine with
304 <0.1 wt% CaO has been excluded in these case studies when using olivine chemical data to infer source
305 lithology (Gao et al., 2008; Wang et al., 2012), high-CaO olivine may also derive from mantle peridotite,
306 as discussed below. The mantle olivine CaO content is mainly controlled by the equilibration
307 temperature and pressure of mantle peridotite, and high-CaO olivine (>0.1 wt%) commonly occurs in
308 high-temperature peridotite at a given pressure (De Hoog et al., 2010; Köhler and Brey, 1990;
309 Witt-Eickschen and O'Neill, 2005) (Supplementary Fig. 4). pMELTS calculations (Ghiorso et al., 2002)
310 for normal peridotite KLB-1 suggest that mantle olivine will have a CaO content of >0.1 wt% when the
311 peridotite mineral assemblage re-equilibrates at 2 GPa with temperature >~1,200°C, and they will
312 have >0.2% CaO when the temperature is >~1,400°C (Fig. 3b and Supplementary Fig. 4). Typical
313 mantle olivine and olivine xenocrysts carried by basalts, mainly with <0.1 wt%, may originate from a
314 random sampling process, and spinel peridotite mainly equilibrates at temperatures between 800 and
315 1,200°C (Rudnick et al., 2004; Wang et al., 2015; Witt-Eickschen and O'Neill, 2005). However, basaltic
316 magmas may accidentally pick up some mantle olivine that has been re-equilibrated at higher

temperatures in the vicinity of the source melting region, such as the high-temperature spinel peridotite reported by Witt-Eickschen and O'Neill (2005). Therefore, if both low- and high-temperature mantle olivine were picked up by basaltic magmas when in transit in the upper mantle, typical low-CaO and slightly higher-CaO mantle xenocrysts would be observed in basaltic rocks. In contrast, the NiO content of mantle olivine remains nearly constant during the peridotite re-equilibration processes at the near-solidus temperature, which is consistent with the NiO-Fo pattern of olivine in Cenozoic basalts (Fig. 3c). It worth noting that CaO and Ni distribution patterns shown in typical olivine xenocryst may not occur in every xenocryst because diffusive re-equilibration, depending on the olivine size and residence time, can decrease or even erase elemental distribution patterns of olivine xenocrysts. Moreover, cut section effect of granular olivine may also affect their geochemical patterns observed in thin section.

For Sihetun and Feixian olivines in Mesozoic picrites, the large variation in NiO content appears to be inconsistent with the mantle olivine diffusive re-equilibration model. However, a closer examination of the olivine data reported by Gao et al.(2008) suggests that the olivine in peridotite also exhibits a large variation in NiO content, and some have a very high NiO contents at a given Fo (Supplementary Fig. 5), displaying chemical features similar to those high-CaO olivines that have been considered magmatic olivine. In addition, a mantle olivine xenocryst included in a hybridized hydrous dioritic magma suggests that the NiO content can vary from ~0.4% to ~0.65% with Fo~90, even when CaO<0.1% wt (Qian et al., 2010). This suggests that the hosting melt composition may significantly influence the chemical diffusion patterns of the olivine xenocryst. Chromite separates of Gao et al. (2008) have significantly lower $^{187}\text{Os}/^{188}\text{Os}$ than wholerock and one chromite separate has sub-chondritic Os isotope composition ($^{187}\text{Os}/^{188}\text{Os} = 0.112$). This also imply that most olivines of Gao et al. (2008) with a xenocrystic origin. The Cenozoic basalts and Mesozoic picrites have significantly different bulk compositions, which might explain the different diffusive re-equilibration patterns of CaO-Fo and NiO-Fo in olivine. Therefore, CaO and NiO in olivine are likely inappropriately used in the source lithology identification of basalts in Eastern China, although this method has been successfully used in oceanic basalts.

3.3. Fe/Mn ratio and Mn content

High Fe/Mn ratios have frequently been used as an indicator of pyroxenite-derived melt contributions for basalts in Eastern China (Gao et al., 2008; Liu et al., 2008; Wang et al., 2012; Wang et al., 2011; Zhang et al., 2009), mainly based on the modeling and interpretations of normal peridotite melts (Herzberg, 2011; Sobolev et al., 2007). However, it is unrealistic to assume that a homogeneous

peridotite is the potential source rock of any basalt because many observations suggest that the Fe/Mn ratio in peridotite varies considerably (Hirose and Kushiro, 1993; Kushiro, 1996; Le Roux et al., 2010; Lee, 2004; McDonough and Sun, 1995). In contrast, the Fe/Mn ratio in basaltic rocks has been considered a useful indicator of melting pressure (Niu et al., 2011). However, using the same partition coefficient of Fe and Mn and the peridotite melting equations as (Niu et al., 2011), our calculation suggests that the pressure effect is not sufficient to account for the large variations in the Fe/Mn ratio of C-OIB (Fig. 4). Moreover, recent experimental determinations of the partition coefficients for Mn and Fe between basaltic melt and olivine, garnet, pyroxenes, and spinel at 3 GPa also suggest that the pressure effect cannot explain the high Fe/Mn ratios in OIB (Davis et al., 2013). Thus, the high Fe/Mn ratio in basalts should be mainly used as a compositional indicator of their source materials (Fig. 4a). The higher Fe/Mn ratios of basalts in Eastern China compared to typical MORB are mainly due to iron enrichment (Liu et al., 2008). This iron enrichment and high Fe/Mn can be achieved by iron-rich and high Fe/Mn peridotite (Davis et al., 2013; Humayun et al., 2004; Putirka et al., 2011) (Fig. 4b), such as peridotite KLB-1 and PHN1611, which does not necessarily require a mantle plume model or pyroxenite source.

Fig. 4 (a) Calculated Fe/Mn versus melting degree (F%) of peridotites PHN1611 (Kushiro, 1996), KLB-1 (Hirose and Kushiro, 1993) and KR4003 (Walter, 1998) at garnet and spinel stability pressure, Fe and Mn partition coefficients of peridotite minerals are the same as those compiled by Liu et al. (2008), and the non-modal batch melting phase relations for garnet peridotite and spinel peridotite are the same as those used by Niu et al. (2011). (b) Fe/Mn versus MgO for Mesozoic and Cenozoic basalts from Eastern China. High-quality Fe/Mn data of Cenozoic and Mesozoic basalts from (Gao et al., 2008; Liu et al., 2008), Hainan basalts from (Liu et al., 2015). Jiangsu and Anhui-1 basalts from (Zhang et al., 2009). Jiangsu and Anhui-2 basalts from (Wang et al., 2011). MORBs from (Le Roux et al., 2010). Note that high Fe/Mn Cenozoic basalts are generally with low CaO contents, but this does not due to clinopyroxene fractionation because

371 Fe/Mn ratios are negatively correlated with Yb, a moderate incompatible element in clinopyroxene (not shown) (Liu et al.,
372 2008; Wang et al., 2012).

373

374 Similar to the effect of temperature on Ni, the Mn content in olivine may be greatly influenced by the
375 temperature effect on Mn partitioning between the olivine and melt. The temperature-dependent
376 equation 2d of Putirka and co-workers (Putirka et al., 2011) has a lower prediction error than the
377 composition-dependent equation of (Herzberg and O'Hara, 2002) in describing Mn partitioning between
378 the olivine and melt even though the former captured 93 data points and the latter captured 143 data
379 points. Moreover, the Mn partition coefficient of olivine calculated by the temperature-dependent
380 equation 2d proposed by Putirka and co-workers (Putirka et al., 2011) is in good agreement with the
381 recent high-precision experimental results (Davis et al., 2013; Le Roux et al., 2011). For olivines in
382 basalts from Eastern China, Mn depletion and the high Fe/Mn ratio have not been widely used to infer
383 the source lithology, partially because of the lack of high-precision data (Xu et al., 2012). Nevertheless,
384 before using this method, the mantle olivine xenocryst diffusion model, as discussed in the previous
385 section, should be carefully considered because mantle olivine generally has a low Mn content, and the
386 chemical re-equilibration process may produce a low Mn content similar to that of olivine from
387 pyroxenite-derived melts. Therefore, the high Fe/Mn in basalts and olivines cannot be safely used to
388 identify the source lithology of basalts from Eastern China.

389 **3.4. Zn/Fe ratio**

390 The high Zn/Fe ratio in basaltic rocks from Eastern China has also been used to support
391 pyroxenite-derived melts (Liu et al., 2015; Qian et al., 2015; Wang et al., 2012; Xu et al., 2012). However,
392 as PM-like ($\text{Zn/Fe}=8.8\times10^{-4}$) peridotite produces low-Zn/Fe ($\sim10\times10^{-4}$ - 12×10^{-4}) melts, high-Zn/Fe melts
393 can be produced from a peridotite source if the peridotite has been metasomatized and enriched in Zn
394 (Le Roux et al., 2011)(Fig. 5). Many trace element studies suggest that OIB-type basalts from Eastern
395 China are mainly derived from enriched mantle, not a PM-like mantle source (Basu et al., 1991; Chen et
396 al., 2007; Zou et al., 2000). Thus, it is reasonable to assume that the potential source peridotite has a
397 high Zn content and high Zn/Fe ratio. Importantly, some high-precision data provided by Le Roux and
398 co-workers suggests that mantle peridotite has Zn/Fe values from $\sim6\times10^{-4}$ to $\sim11\times10^{-4}$ (Le Roux et al.,
399 2010). This wide range may explain why OIB has a large variation in the Zn/Fe ratio for the peridotite
400 model (Davis et al., 2013). Interestingly, high-Fe/Mn peridotite PHN1611 that has been metasomatized

by undersaturated silicate melts has a high Zn/Fe ratio (10.4×10^{-4}) (Smith et al., 1993), which can produce both high-Fe/Mn and high-Zn/Fe melts (Fig. 5). However, high-Zn/Fe peridotite can have either high Fe/Mn or low Fe/Mn ratios, and vice versa (Le Roux et al., 2010). This suggests that peridotite melts can have large variations in both Fe/Mn and Zn/Fe, as shown in Fig. 5b. Therefore, as suggested by Davis and co-workers, the Zn/Fe ratio in basalts is not a sensitive indicator of non-peridotite source lithology (Davis et al., 2013).

Fig. 5 Zn/Fe versus MgO and Fe/Mn versus Zn/Fe for peridotite melts and Mesozoic and Cenozoic basalts from Eastern China. Zn/Fe ratios of melts derived from primitive mantle peridotite (PM) (McDonough and Sun, 1995), peridotite PHN1611 (Smith et al., 1993) and other peridotite with different Zn/Fe ratios (6-11) (dashed square) are calculated using the same method as (Le Roux et al., 2010). Hainan basalts from (Liu et al., 2015; Wang et al., 2012). Shandong basalts from (Xu et al., 2012). Cenozoic and Mesozoic basalts from (Liu et al., 2008). The data in (b) only include basalts with MgO > 7.5 wt%. MORBs from (Le Roux et al., 2011).

4. Pyroxenite model of basalts from Eastern China based on the FC3MS value

As discussed above, when identifying the source lithology of basalt from Eastern China, the olivine chemistry method may be compromised by xenocryst diffusive re-equilibration processes. High Fe/Mn, Zn/Fe and low CaO may be characteristics of refertilized or metasomatized peridotite melts (Table 2). Moreover, in the commonly used Harker diagrams and phase projections, partial melts from a number of peridotitic rocks collectively show indistinguishable major element signatures compared with pyroxenite melts when olivine-dominated fractional crystallization occurs (Lambart et al., 2013; Putirka et al., 2011; Yang and Zhou, 2013). As shown in Fig. 6 for diverse peridotite melts and two representative pyroxenite

424 melts, the most noticeable signature of pyroxenite melts is that they generally have lower MgO contents
425 than peridotite melts, but olivine fractional crystallization can easily decrease the MgO content of
426 peridotite melts, producing major element features that are collectively indistinguishable from those of
427 pyroxenite melts. Moreover, if magma mixing occurs for both peridotite- and pyroxenite-derived melts,
428 these melts will become more complex in their major element patterns. Consequently, it seems
429 impossible to identify the source lithology of the Cenozoic and Mesozoic basalts from Eastern China
430 based on their major elements (Fig. 6). Nevertheless, using a parameterization of multi-decadal melting
431 experiments on a variety of peridotite and pyroxenite, Yang and Zhou (2013) proposed a parameter
432 called the FC3MS value ($\text{FeO}^T/\text{CaO}-3*\text{MgO}/\text{SiO}_2$, all in wt%), which can identify most pyroxenite-derived
433 basalts. This marker was obtained based on the assumption that any peridotitic and pyroxenitic rocks
434 that commonly occur in the upper mantle in both volatile-free and volatile-present conditions may
435 produce melts contributing to basaltic magmas. The FC3MS value, which is mainly controlled by the
436 source bulk composition and melting degree, has nearly no temperature or pressure effects, suggesting
437 that the OIB-type magmas from Eastern China and Mongolia are mainly derived from pyroxenite sources
438 (Yang and Zhou, 2013).

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441

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Fig. 6 Major element characteristics of experimental melts of peridotite and pyroxenite and representative basalts from Eastern China. Data for Cenozoic basalts from (Huang et al., 2013; Liu et al., 2015; Qian et al., 2015; Wang et al., 2012; Wang et al., 2011; Xu et al., 2012; Zhang et al., 2009) (Supplementary Table 1). Data for Mesozoic basalts from (Gao et al.,

2008; Liu et al., 2008). Experimental pyroxenite melts including pyroxenites KG1 (1.5-3 GPa) (Kogiso et al., 1998) and M5-40 (1-2.5 GPa) (Lambart et al., 2012; Lambart et al., 2009; Lambart et al., 2013) melts (Supplementary Table 2). Experimental peridotite melts including PM-like peridotite KLB-1 (1-3 GPa) (Davis et al., 2011; Hirose and Kushiro, 1993) and KR4003 (3-7 GPa) (Walter, 1998), iron-rich peridotite HK-66 (1-3 GPa) (Hirose and Kushiro, 1993), iron-rich and K₂O-rich PHN1611 (0.5-3 GPa) (Hirose and Kawamura, 1994; Hirose and Kushiro, 1998; Kushiro, 1996), Hawaiian peridotite HPY (1-3 GPa) (Falloon et al., 1988), depleted harzburgite HZ (1.5-2.5 GPa) (Falloon and Danyushevsky, 2000) and K₂O-rich peridotite (1 and 3 GPa) (Davis and Hirschmann, 2013; Laporte et al., 2014) melts (Na₂O+K₂O>12% melts are not shown) (Supplementary Table 3). Peridotite HK-66 has less than 40 wt% olivine in both our model (Fig. 1) and the natural sample (Hirose and Kushiro, 1993) and should be considered as a transitional lithology between pyroxenite and peridotite. L-OI, typical olivine fractionation trend. Only basalts with MgO>7.5 wt% from Eastern China are shown in plots of FeO versus SiO₂ and CaO-13.81+0.274*MgO versus Na₂O+K₂O.

Here, we firstly pay more attention to the source lithology and petrological implications of the Mesozoic and Cenozoic continental basalts with SiO₂=45-53 wt% from Eastern China because this type of basalt is very similar as both peridotite melts and pyroxenite melts (Fig. 6), and is widely distributed in not only Eastern China but also other countries and has been used to deduce many petrological and geological implications based on different assumptions of source lithology (Chen et al., 2007; Fan and Hooper, 1991; Liu et al., 2015; Timm et al., 2010; Zhang et al., 2009; Zhi et al., 1990). As discussed above, all of the geochemical characteristics of these basalts can be accurately explained by the peridotite model under the assumption that mantle peridotite is heterogeneous in bulk composition (Fig. 6). Although both the Cenozoic and Mesozoic basalts are geometrically similar as peridotite melts in every major element (Fig. 6), the high FC3MS value of the Cenozoic basalts at given MgO, Na₂O+K₂O, Mg[#] (MgO/(MgO+FeO^T), molar fraction) and Fe/Mn contents cannot be explained by any peridotite melts if olivine-dominated fractionation occurs (Fig. 7). Hydrous and carbonated peridotite-derived basaltic melts, not shown in Fig. 7, have identical and lower FC3MS values compared with volatile-free peridotite-derived melts, respectively (Yang and Zhou, 2013). Recent experimental petrology suggests that garnet peridotite and metasomatized peridotite-derived melts (Davis and Hirschmann, 2013; Grove et al., 2013; Laporte et al., 2014; Pilet et al., 2008) do not have a sufficiently high FC3MS value to explain the Cenozoic basalts from Eastern China. We noticed that some basalts from Hainan and Wudalianchi have sub-chondritic Os isotopes (¹⁸⁷Os/¹⁸⁸Os = 0.11869~0.12954) (Chu et al., 2013; Wang et al., 2013), but their high FC3MS (0.64~1.11) values suggest that their source lithology is pyroxenite. This seems inconsistent with their peridotite-like sub-chondritic Os isotopes, however, refertilized

475 peridotite may become pyroxenite by infiltration of large amount of asthenosphere-derived melts, and
476 preserve the sub-chondritic Os isotopes.

477 One may argue that the high FC3MS value of Cenozoic basalts is due to large amount of
478 clinopyroxene fractionation, which appears to be consistent with the co-variation of FC3MS versus
479 $\text{Na}_2\text{O}+\text{K}_2\text{O}$, $\text{Mg}^\#$ and MgO (Fig. 7a-c). However, this interpretation is inconsistent with the typical
480 evolution processes of peridotite-derived melts because the ascent and decompression of peridotite
481 melts would result in olivine oversaturation and pyroxene undersaturation, and clinopyroxene
482 crystallization commonly occurs when $\text{MgO}<6\text{-}10\text{ wt\%}$. In addition, a number of previous studies suggest
483 that the geochemical features of Cenozoic basalts from Eastern China are mainly controlled by source
484 materials, partial melting and olivine-dominated fractionation when the MgO content $>6\text{-}10\text{ wt\%}$ (Fan and
485 Hooper, 1991; Huang et al., 2013; Liu et al., 2015; Liu et al., 2008; Wang et al., 2012; Zeng et al., 2011;
486 Zhang et al., 2009). Moreover, most of these basalts carry a variety of mantle-derived xenoliths,
487 indicating that magma evolution is limited during the ascent in crustal conduits. Alternatively, pyroxenite
488 melts can reasonably explain all of the co-variations of FC3MS versus $\text{Na}_2\text{O}+\text{K}_2\text{O}$, $\text{Mg}^\#$, MgO and Fe/Mn
489 (Fig. 7a-c). Large analytical errors in the MnO content of both peridotite and pyroxenite melts results in
490 large variations in Fe/Mn , and peridotite melts can have high Fe/Mn ratios but low FC3MS values (Fig.
491 7d). Importantly, the pyroxenite M5-40 model melts that equilibrium with garnet and clinopyroxene can
492 more reasonably account for the overall positive correlation of FC3MS and La/Yb than simple
493 clinopyroxene fractional crystallization trends (Fig. 7e). In some cases, the pyroxenite M5-40 model
494 melting trends are in good agreement with the correlation of FC3MS and La/Yb of continental OIB-type
495 basalts when $\text{MgO}=5\text{-}12\%$ (see Fig. 3c in (Yang and Zhou, 2013)) and the Hannuoba basalts (Liu et al.,
496 2015; Zhi et al., 1990) when $\text{MgO}=5\text{-}11\%$ (see Fig. 7e). Note that low- MgO ($<6\text{-}10\text{ wt\%}$) basalts are
497 commonly considered as highly evolved products and excluded in discussing their petrogenesis, but the
498 low- MgO basalts shown here and our previous work (Yang and Zhou, 2013) with higher FC3MS value
499 and higher La/Yb than cogenetic high- MgO basalts, which consistent with low degree partial melts of
500 pyroxenite source. Alternatively, peridotite-derived melts can also produce comparable geochemical
501 trends between FC3MS and La/Yb if garnet and clinopyroxene-dominated high pressure fractional
502 crystallization occur, but this mechanism needs high temperature MgO -rich melts produced first. As we
503 discussed bellow, at normal mantle potential temperature garnet pyroxenite would easily partially melted
504 even when surrounding peridotite still subsolidus. Therefore, the low- MgO and high- MgO Cenozoic

505 OIB-type basalts from Eastern China are almost primary pyroxenite melts, indicating that caution should
506 be used when inferring the source lithology and petrogenesis of both low- and high-MgO basaltic rocks.
507 This also suggests that the commonly used criteria for primary mantle-derived magmas, such as high
508 MgO and $Mg^\#$ (Frey et al., 1978), are not suitable for these primary pyroxenite-derived mantle magmas.
509 A similar conclusion has been reached by Lambart et al.(2009). For Mesozoic basalts in Eastern China,
510 all of their compositional characteristics can be explained by partial melts from heterogeneous peridotite,
511 but pyroxenite sources cannot be fully excluded because high-MgO pyroxenite melts can also account
512 for the compositional characteristics of the basalts (Figs. 6 and 7).

513

514 **Fig. 7** $\text{FeO}^T/\text{CaO}-3*\text{MgO}/\text{SiO}_2$ versus MgO , $\text{Na}_2\text{O}+\text{K}_2\text{O}$, $\text{Mg\#}$, Fe/Mn and La/Yb of experimental peridotite and pyroxenite
 515 melts and representative basalts from Eastern China. Experimental peridotite and pyroxenite melts and Mesozoic and
 516 Cenozoic basalts in (a-d) are the same as those in Fig. 6. (e) is modified from Yang and Zhou (2013). Hannuoba basalts

517 (Liu et al., 2015; Zhi et al., 1990) that have the most evident compositional variations are indicated in (e). Hornblende
518 peridotite melts from (Pilet et al., 2008). Olivine fractionation and accumulation trends, and fractional (FC) and equilibrium
519 (EC) clinopyroxene fractionation trends are shown (Yang and Zhou, 2013). Two melting curves of average pyroxenite with
520 low-La/Yb (1.42) and high-La/Yb (2.78) sources are also shown. Metasomatized and PM-like peridotite have different
521 major element characteristics but the same assumed source La/Yb (1.58). The La/Yb values of peridotite and pyroxenite
522 melts are calculated using a non-modal batch melting model. $\text{FeO}^T/\text{CaO}-3*\text{MgO}/\text{SiO}_2$ of peridotite and pyroxenite melts
523 are calculated using their quantitative relationships with melting degree (F%). 1σ of $\text{FeO}^T/\text{CaO}-3*\text{MgO}/\text{SiO}_2$ in (b) and (e)
524 are mainly due to experimental melt errors and the small pressure effect. The broken curves in (a-e) represent the highest
525 FC3MS value of peridotite-derived melts if olivine-dominated fractionation occurs. For other details, see Yang and Zhou
526 (2013). The Mn content of experimental peridotite melts has large analytical error compared with Fe and other major
527 elements. Thus there is no significant pressure relations with Fe/Mn ratio of melts for a specific source rock. Importantly,
528 recent experimental work (Davis et al., 2013) and our calculation shown in Fig. 4 suggest that Fe/Mn are largely controlled
529 by source composition.

530

531 There are also some Cenozoic alkaline basalts with very low SiO_2 content (~40-45wt%) in Eastern
532 China, and they are commonly have higher FeO^T contents than those high SiO_2 (>45wt%) basalts (e.g.
533 Sakuyama et al., 2013; Xu, 2014; Xu et al., 2014; Zeng et al., 2010). These low silica and high iron
534 basalts have been experimentally produced by melting of pyroxenitic rocks of hornblendite (Pilet et al.,
535 2008) and carbonated eclogite (Dasgupta et al., 2006), and of peridotitic rocks of carbonated peridotite
536 (Dasgupta et al., 2007; Dasgupta et al., 2013). Although carbonated peridotite has been suggested as
537 the source lithology of basalts in Eastern China (Sakuyama et al., 2013; Zeng et al., 2010), their partial
538 melts are too low in FC3MS value (< -0.2) (Dasgupta et al., 2007; Dasgupta et al., 2013) to explain the
539 high FC3MS (0.2~0.8, when $\text{MgO}>7.5\text{wt}\%$) basalts in Eastern China (e.g. Sakuyama et al., 2013; Xu,
540 2014; Xu et al., 2014; Zeng et al., 2010) (See Figure 3 in Yang and Zhou, 2013). Therefore, either
541 pyroxenitic source (hornblendite, carbonated eclogite or low silica pyroxenite) or large amount of
542 pyroxenite-derived melts are mixed with carbonated peridotite-derived melts can explain the major
543 elements characteristics of these low silica basalts in Eastern China. Because typical low FC3MS (< -0.2)
544 carbonated peridotite-derived basalts have not been reported along with these high FC3MS basalts, the
545 pyroxenitic rocks are the most likely source materials of basalts in Eastern China.

546 Using the olivine-addition method of Hainan basalts, Wang and co-workers propose that the mantle
547 potential temperature varies from 1,500 to 1,580°C in the Hainan area (Wang et al., 2012). They argue
548 that this high mantle potential temperature and the high-Fo olivine phenocrysts and recycled oceanic

549 crust in their source region provide independent petrological evidence for the Hainan plume model.
550 However, their model primary Hainan basalts, with the FC3MS value varying from 0.11 to 0.74
551 ($Mg^\# = 72-73$) (Wang et al., 2012), are highly inconsistent with peridotite melts (Fig. 7c). In addition,
552 high-Fo (89-91) olivines in Hainan basalts show a large variation in CaO content (Fig. 3), regardless of
553 their textural characteristics (Wang et al., 2012). Although olivines with $CaO < 0.1\%$ have been
554 considered mantle xenocrysts, high-CaO olivines with $CaO = 0.1-0.25\%$ may also disaggregated from
555 high-temperature re-equilibrated peridotite (Fig. 3). Note that high-Fo olivine-bearing primitive samples
556 ($MgO > 9$ wt% and $CaO > 8$ wt%), which have been used to calculate the primary melt compositions of
557 Hainan basalts, have FC3MS values varying from 0.56 to 0.89 ($Na_2O + K_2O = 3.34-4.96$ wt%) (Wang et al.,
558 2012), which are the typical pyroxenite melt characteristics and cannot be accounted for by peridotite
559 melts (Fig. 7a, b). Thus, the model partial melting temperature and pressure of primary Hainan basalts
560 based on a peridotite source exhibit high uncertainties. Trace element ratios, such as La/Yb, Dy/Yb and
561 Zr/Hf, have been frequently used to trace the garnet signature in basalt sources. Using this method,
562 Huang et al. (2013) argued that Cenozoic OIB-like basalts from Guangdong in Southeast China may
563 represent the partial melting of the asthenospheric mantle beneath the thickened lithosphere. This
564 argument holds only for peridotite sources. However, these OIB-like basalts ($MgO = 8-12\%$,
565 $Na_2O + K_2O = 4.6-6.6\%$, $Mg^\# = 55-63$) exhibit FC3MS values (0.53-0.92) (Huang et al., 2013) that are not
566 sufficiently low to be explained by peridotite-derived melts (Fig. 7a-c). If the basalts are
567 pyroxenite-derived products, the typical garnet signature implied by the LREE/HREE ratios will not
568 suggest a high-pressure garnet peridotite source because garnet can be stable at low pressures for
569 pyroxenite sources. Therefore, the primary magma composition and its origin temperature and pressure
570 cannot be accurately constrained if pyroxenite-derived basalts are erroneously considered peridotite
571 sources.

572 For our pyroxenite model, at normal mantle potential temperatures of 1,350-1,400°C, adiabatic
573 upwelling of the mantle will result in 0-40% partial melting of the pyroxenite M5-40-like source rock when
574 melting occurs between 2 and 3 GPa, but the peridotite is still near-solidus at the same temperature and
575 pressure (Fig. 8). The high degree partial melting of pyroxenite seems cannot explain the enriched trace
576 elements of the Cenozoic basalts from Eastern China, however, low degree partial melts derived from
577 MORB and/or asthenosphere may react with peridotite to generate trace element-enriched pyroxenite,
578 which can produce basaltic melts with enriched trace elements under very high degree partial melting

(Fig. 7e). Different degrees of partial melts may erupt at the surface depending on the final extracting pressure, and all of these melts will present typical garnet signatures even when partial melting occurs at ~2 GPa. In the source region, the peridotite wall rock may re-equilibrate at the subsolidus temperature because of the adiabatic upwelling of the mantle. These near-solidus peridotite and other low-temperature mantle xenoliths may be accidentally picked up by pyroxenite-derived basaltic magmas during their ascent through the upper mantle. Below the solidus of peridotite, partial melting of low-MgO pyroxenite would yield primary melts not in equilibrium with the hosting mantle peridotite, this would lead pyroxenite-derived melt interact with surrounding mantle, the major elements characteristic of pyroxenite primary magma would not be preserved any more if they reach equilibrium with mantle peridotite. However, the pyroxenite source signature can be preserved if the magmas migrate to surface rapidly when the surrounding mantle is partial melted (Lambart et al., 2012). Therefore, for Hainan basalts and the other Cenozoic basalts from Eastern China shown in Fig. 7, pyroxenite-derived melts contaminated by small amount of disaggregated xenocrysts from peridotite can explain why many low-MgO basalts carry high-Fo olivines that show different minor element characteristics compared with olivines crystallized from typical peridotite melts.

Fig. 8 Experimentally determined solidus for typical peridotite (Hirschmann, 2000) and pyroxenites M5-40 (Lambart et al., 2012; Lambart et al., 2009; Lambart et al., 2013) and MIX1G (Kogiso et al., 2003), and stability fields for garnet pyroxenite M5-40 and typical garnet peridotite. Adiabats for mantle potential temperatures of 1,350, 1,400 and 1,570°C are shown, with a subsolidus adiabatic temperature gradient of 10°C/GPa and a liquid adiabat of 30°C/GPa (Mckenzie and Bickle,

1988). Dashed lines (0.2, 0.4 and 0.6) indicate the partial melting degree of pyroxenite M5-40. The temperature and pressure ranges of Hainan basalts, calculated through the olivine addition method for peridotite primary melts, are from (Wang et al., 2012).

604

5. Concluding remarks

The commonly used markers for the pyroxenite contribution in basalts are briefly summarized, and some new alternative interpretations using a peridotite model are explored to explain the pyroxenite signatures displayed by literature markers of the Cenozoic and Mesozoic basalts from Eastern China. The uncertainties of the commonly used markers for pyroxenite contribution in basalts from Eastern China may also occur for many other oceanic and continental basalts. Although the Cenozoic basalts with $\text{SiO}_2=45\text{-}53$ wt% collectively show compositional characteristics that are indistinguishable from those of peridotite-derived melts, their whole-rock FC3MS value, combined with $\text{Na}_2\text{O}+\text{K}_2\text{O}$, $\text{Mg}^\#$, MgO and La/Yb , suggests that they are mainly primary pyroxenite-derived products, and many low-MgO basalts are also pyroxenite-derived products. Caution should be paid when interpreting the petrogenesis of low-MgO basalts. For high-Fo olivine-bearing basalts, the origin of the "phenocrysts" should be carefully examined before using their chemical signature to infer source lithology. At the normal mantle potential temperature, a pyroxenite source can produce melts that explain the compositional characteristics of the Cenozoic basalts, suggesting that the origin temperature and pressure of the Cenozoic basalts determined by the traditional peridotite model are overestimated. For the Mesozoic basalts, compositionally diverse peridotite sources can account for their petrogenesis, but pyroxenite sources still cannot be excluded.

We acknowledge that the pyroxenite source may represent crustal materials (continental or oceanic crust) recycling (e.g. Liu et al., 2008; Gao et al., 2009; Zhang et al., 2009) and/or mantle metasomatism (Niu et al., 2005; Guo et al., 2014; Guo et al., 2016), which have important geodynamic implications, but to distinguish them is a much more complex issue, and which might not be easily resolved before enough understanding on the identification of source lithology of basalts. If the FC3MS value and/or other effective chemical markers can be quantitatively connected with source bulk composition including major element, trace element and isotopic characteristics, the pyroxenite origin may be resolved, but this needs much more numeric simulation on experimental petrological data and comprehensive high quality geochemical and mineralogical data of OIB-type basalts from Eastern China, and this will be discussed

631 in a subsequent study.

632

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