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RESEARCH ARTICLE

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Key Points:

- The $\delta^{18}\text{O}$ of the 106–60 Ma basalts in NCC were measured by SIMS
- The $\delta^{18}\text{O}$ of the basalts are different from that of the lithospheric mantle
- OIB-type rocks in NCC were most likely derived from the convective asthenosphere, rather than the lithospheric mantle

Supporting Information:

- Supporting Information S1
- Data Set S1
- Data Set S2

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Dynamic contribution of recycled components from the subducted Pacific slab: Oxygen isotopic composition of the basalts from 106 Ma to 60 Ma in North China Craton

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Abstract How the materials derived from the stagnant Pacific slab contributed to the mantle sources of the mafic rocks in east China is still in hot debate. In this work, $\delta^{18}\text{O}$ (Vienna standard mean ocean water) values of clinopyroxene phenocrysts in the oceanic island basalts (OIB)-type mafic rocks from 106 Ma to 60 Ma in the east North China Craton (NCC) were measured by secondary ion mass spectrometry. Our data show that for all of the samples, the $\delta^{18}\text{O}_{\text{Cpx}}$ are dominantly higher than that of the clinopyroxene from normal mid-oceanic ridge basalt (5.4–5.8‰), which confirms the role of recycled oceanic crust (ROC) in their mantle sources. Combining the $\delta^{18}\text{O}$ data of basalts and the lithospheric mantle in the literature, we found that in the southeast NCC, upper and lower ROC components alternately appeared in the mantle sources of basalts, but these ROC components are consistently different from that in the lithospheric mantle, while in the northern NCC, the recycled components in the sources seem to be persistently from upper ROC. These observations suggest that (1) these mafic OIB-type rocks are most likely derived from the convective asthenosphere and (2) the contribution of components from the Pacific oceanic slab into the NCC upper mantle was dynamic, without a simple temporal trend. This new knowledge calls for the reconsideration of the existing models of the thinning process of the NCC lithospheric mantle, and it warns against simply using the chemical composition of basalts to infer the evolution of lithosphere.

1. Introduction

Many seismic tomography studies have shown that the subducted Pacific oceanic slab has been stagnant within the mantle transition zone and has extended westward subhorizontally beneath East Asia [Fukao *et al.*, 1992; Huang and Zhao, 2006]. The western end of this stagnant slab has reached the North-South Gravity Lineament (NSGL) [Huang and Zhao, 2006]. The subduction of the Pacific plate and the subsequent extension is now regarded as one of the principal triggers of the destruction of the North China Craton (NCC) [Xu, 2007; Zhu *et al.*, 2011]. In addition to the dynamic impact, such as inducing the rift of the lithospheric mantle and the subsequent upwelling of asthenosphere, many studies have suggested that the components (hydrated fluids and/or melts) derived from this subducted oceanic slab have directly contributed to the metasomatism and thinning of the lithospheric mantle and/or the production of the mafic magmatism from the Early Cretaceous to the late Cenozoic [Niu, 2005; Zhao, 2004; Pang *et al.*, 2015; Meng *et al.*, 2015; Xia *et al.*, 2013; Zhang *et al.*, 2009; Wang *et al.*, 2011; Z. Xu *et al.*, 2012; Y. G. Xu *et al.*, 2012; Yang *et al.*, 2012; Sakuyama *et al.*, 2013; Liu *et al.*, 2015a, 2015b; Chen *et al.*, 2015; Wang *et al.*, 2015; Xu, 2014; Kuang *et al.*, 2012].

Recently, the role of the dehydrated fluids from the subducted Pacific slab in the thinning of the SCLM has been shown by quantitative or qualitative determinations of the water concentration of the lithospheric mantle during the Early Cretaceous [Xia *et al.*, 2013; Liu *et al.*, 2016; Pang *et al.*, 2015]. However, whether and how the slab-derived components were associated with the mantle sources of the Late Cretaceous to Cenozoic basalts are still under debate. Niu [2005], Guo *et al.* [2014], and Meng *et al.* [2015] strengthened the role of hydrous melts released from the subducted Pacific slab as the metasomatic agents for the mantle sources of the mafic rocks with ages <110 Ma, while they denied the contribution of recycled subducted marine carbonates and partial melts of the basaltic oceanic crust. In contrast, Zhang *et al.* [2009], Wang *et al.* [2011], and Z. Xu *et al.* [2012] argue that the $\delta^{18}\text{O}$ anomaly (which is generally lower than the $\delta^{18}\text{O}$ for N-MORB olivine, $5.16 \pm 0.09\text{‰}$ [Eiler *et al.*, 1997]) of the olivine and pyroxene phenocrysts in the late Cenozoic basalts in the

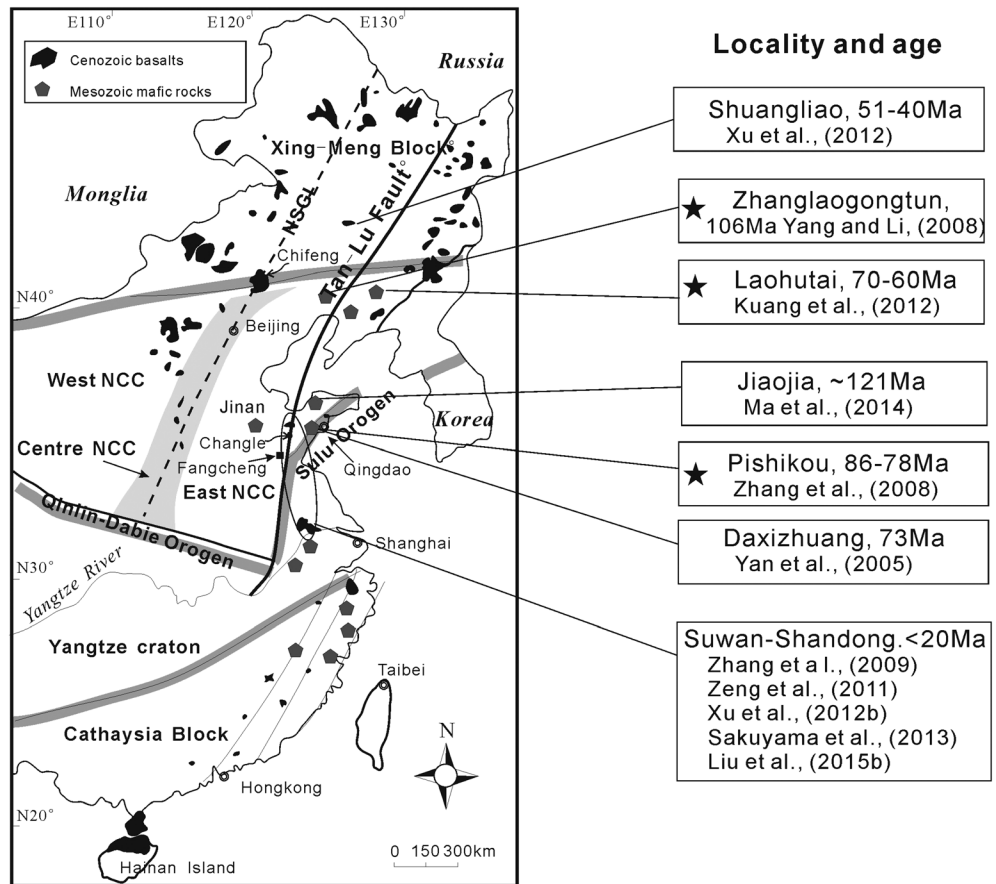


Figure 1. The simplified tectonic scheme for east China and the location of the 121 Ma to late Cenozoic basalts. NSGL represent the North-South Gravity Lineament. The dark stars mark the basalts in this study.

southeast NCC exclusively provides evidence for the involvement of components derived from the subducted oceanic crust. They further suggest that these recycled oceanic components had resided in a juvenile lithospheric mantle (formed by cooling of the upwelling asthenosphere) since the Early Cretaceous in the form of pyroxenites that were the product of the interaction between the peridotite and ROC-derived melts. However, very recently, Y. G. Xu et al. [2012], Kuang et al. [2012], and Xu [2014] investigated the temporal changes of geochemical compositions, such as Eu/Eu^* and $^{87}\text{Sr}/^{86}\text{Sr}$, for the basalts from 90 to 40 Ma in the eastern NCC, and they attributed the secular shifts of the geochemical characteristics to the differential melting of the recycled components from the upper to lower ROC in the upwelling asthenosphere.

The principal sensitivity to the interaction of rocks with the Earth's hydrosphere makes oxygen isotope geochemistry a sensitive tool for identifying recycled crustal material from mantle-derived rocks and minerals [e.g., Muehlenbachs, 1986; Eiler, 2001]. In this work, using secondary ion mass spectrometry (SIMS), we measured the oxygen isotopes of clinopyroxene (cpx) phenocrysts in 106 Ma to 60 Ma basalts from the west Liaoning and east Shandong region (Figure 1). We also compiled the oxygen isotopic data for the lithospheric mantle of the eastern NCC from the Early Cretaceous to late Cenozoic and compared them to the $\delta^{18}\text{O}$ data of basalts from this work and from the literature. The results will help us to explore (1) whether there were recycled oceanic crust components in the mantle sources of mafic rocks during the Late Cretaceous to early Cenozoic, (2) whether there was a temporal or regional variation for these ROC components, and (3) whether these ROC components resided in the juvenile lithospheric mantle since the Early Cretaceous or were incorporated into the upper mantle gradually on a long time scale. The answers to these questions would (1) improve our understanding of the recycling process of a subducted oceanic slab in the "back-arc" region and (2) provide new insight in the discussion of the thinning process of the NCC lithospheric mantle.

2. Geological Background and Samples in This Study

2.1. Mafic Magmatism From Mesozoic to Cenozoic in NCC

The NCC has been stable since its cratonization [Zhao and Zhai, 2013] in the Proterozoic. Extensive basaltic magmatism occurred from the Mesozoic on and can be divided into three stages, after Xu *et al.* [2009]. The first stage of magmatism is from the Late Jurassic and Early Cretaceous and is distributed both in the margin and interior of the NCC. This basaltic magmatism is characterized by a very negative $\epsilon\text{Nd}(t)$ value (down to -20) and an arc-type trace element pattern in the primitive mantle normalized diagram, which has been suggested to be derived from the enriched lithospheric mantle [Xu, 2001; Zhang *et al.*, 2002; Yang and Li, 2008; Huang *et al.*, 2012]. However, a recent study on an Early Cretaceous (~ 121 Ma) lamprophyre in the Jiaodong Peninsula shows $\epsilon\text{Nd}(t)$ up to 1.76 and OIB-type trace element patterns [Ma *et al.*, 2014]. The second stage of magmatism is from ~ 106 to 40 Ma [Zhang *et al.*, 2003; Yang and Li, 2008; Yan *et al.*, 2005; Zhang *et al.*, 2008; Kuang *et al.*, 2012; Z. Xu *et al.*, 2012; Cai *et al.*, 2013; Xu, 2014]. These basalts have positive $\epsilon\text{Nd}(t)$ values but more varied $^{87}\text{Sr}/^{86}\text{Sr}$ and OIB-type trace element patterns, and they show temporal variations in their Eu/Eu^* and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios [Z. Xu *et al.*, 2012; Xu, 2014; Kuang *et al.*, 2012]. The third stage of magmatism is in the late Cenozoic. The basalts in this stage include predominantly alkali basalts and subordinate basanites and tholeiites. The obvious features of these basalts are positive $\epsilon\text{Nd}(t)$ values and OIB-type trace element patterns [Song *et al.*, 1990; Zhi *et al.*, 1990; Zhang *et al.*, 2009; Zeng *et al.*, 2011]. Recently, it has been suggested that the component from ROC or recycled continental crust exists in their mantle source, despite the fact that there is still debate about whether the mantle source resides in the asthenosphere or in the lithospheric mantle [Liu *et al.*, 2008; Zhang *et al.*, 2009; Zeng *et al.*, 2011; Z. Xu *et al.*, 2012; Y. G. Xu *et al.*, 2012; Sakuyama *et al.*, 2013; Liu *et al.*, 2015a, 2015b]. Such a secular variation in magma composition was recently suggested to reflect the prolonged thinning of the NCC lithospheric mantle [Z. Xu *et al.*, 2012; Xu, 2001, 2014; Kuang *et al.*, 2012].

2.2. Samples in This Study: 106 Ma to 60 Ma Basalts in NCC

In this work, a subset of second-stage mafic rocks from Xu [2014], nominally the Pishikou basanites, Laohutai alkali-to-tholeiitic basalts [Kuang *et al.*, 2012], and Zhanglaogongtun alkali basalts (106 Ma, the first-appeared basalt with OIB-type trace element pattern and $\epsilon\text{Nd}(t) > 0$ in the northern margin of the NCC [Zhang *et al.*, 2003; Yang and Li, 2008]) (Figure 1 and supporting information Data Set S1), was chosen for analysis of their oxygen isotopic composition. A brief description of these rocks is given below in the order of decreasing age of eruption, and their geochemical characteristics are shown in Figure 2.

The Zhanglaogongtun Formation (ZLGT) alkali basalts erupted at 106 Ma (Figure 1), and they are dark gray and massive with well-developed columnar jointing [Zhang *et al.*, 2003; Yang and Li, 2008]. Most of the samples contain sparse olivine and plagioclase phenocrysts and possess small spinel-facies mantle xenoliths [Zhang *et al.*, 2003] and crustal-derived cpx xenocrysts as the core of some of the zoned cpx (this work; see Figure 3). In this work, a subset of the samples from Yang and Li [2008] (JG1-4) are selected. These sample suites have also been measured for Mg isotopic composition by Yang *et al.* [2012].

The Pishikou (PSK) dikes, with K-Ar ages from 86 to 78 Ma, intruded into the Laoshan Early Cretaceous alkali granites near Qingdao in Jiaodong Peninsula [Zhang *et al.*, 2008]. The dike is approximately 5–6 m wide and can be subdivided into the southern and northern parts. The samples in this study were sampled from the southern part, which contains abundant peridotite and granulite xenoliths. All samples are dark green and show porphyritic texture. Phenocrysts are small and dominantly clinopyroxene (cpx, $\sim 10\%$), and the matrix is composed of clinopyroxene, feldspar, aphanitic materials, and some accessory minerals of ilmenites and apatites. Cpx phenocrysts are usually zoned, with the core as mantle- or crustal-derived xenocrysts and rim growing from the magma (see section 4). The $\delta^{18}\text{O}$ of olivine as xenocrysts or in peridotite xenoliths hosted by these basanites have been measured by Guo *et al.* [2013] with secondary ion mass spectrometer (SIMS).

The Laohutai (LHT) Formation is located in the Fushun-Mishan graben near Fushun City, which is the northern extension of the Tan-Lu fault. It consists of volcanic successions and subordinate amounts of coal, gray to black shales, sandstones, and tuffs. The volcanic lava succession could be subdivided into three units, in which the lowermost is alkali basalts and the middle and upper units are tholeiitic basalts [Kuang *et al.*, 2012]. Ar-Ar dating shows that they were emplaced at 70 Ma (alkali basalts) and 60 Ma

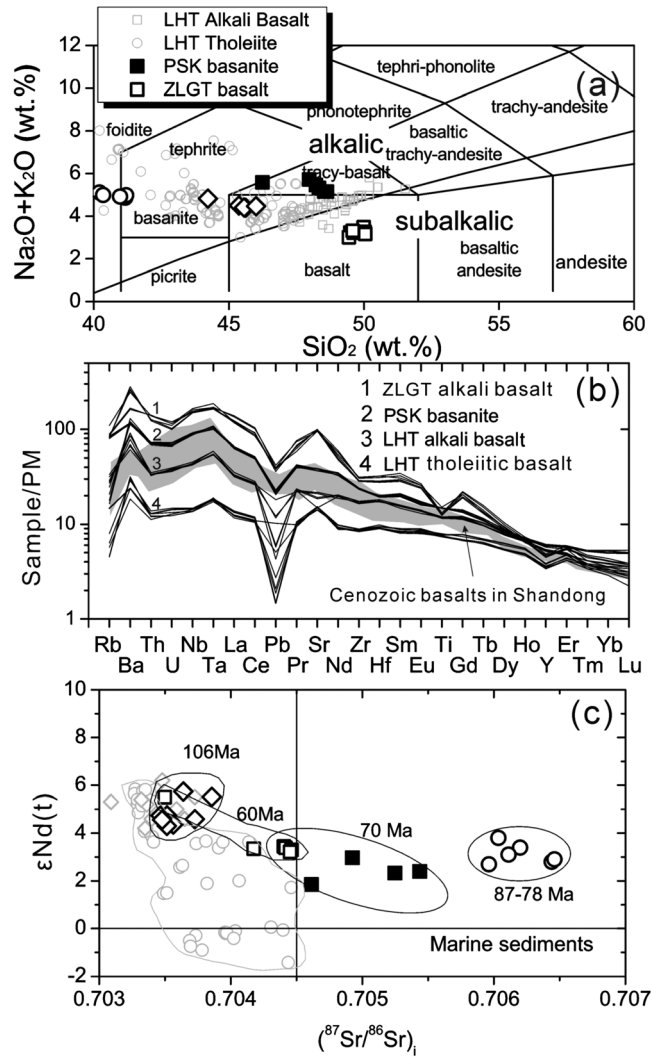


Figure 2. Major, trace and Sr-Nd isotopic characteristics of the 106–60 Ma basalts in this study. LHT represents Laihutai, PSK represents Pishikou, and ZLGT represents Zhanglaogongtun. The data source of these basalts can be found in the main text. (a) Total alkali versus SiO₂ plots. Mafic rocks erupted in Late Cretaceous (thin gray square, data from Zhang et al. [2002] and Gao et al. [2008]) and late Cenozoic basalts (thin gray circle) are also shown for comparison. (b) Primitive mantle normalized trace element patterns. The late Cenozoic basalts in Shandong [Zeng et al., 2011] are shown in the gray region. (c) Sr-Nd isotopes. Thin gray circles are late Cenozoic basalts in Shandong [Zeng et al., 2011]. The PM normalization value is from McDonough and Sun [1995].

analysis was performed to examine the alteration and small melt or fluid inclusions [Liu et al., 2015a]. Cpx separates without alteration-contaminants and free of inclusions were mounted again for chemical composition and oxygen isotope analysis.

3.1. Electron Microprobe

The chemical compositions of cpx phenocrysts were measured with a Shimadzu Electron Probe Microanalyzer (EPMA 1600) at CAS-CMME, USTC. Backscattered Electron (BSE) images were used to check the homogeneity of these phenocrysts. During the quantitative analysis, the operating conditions were set as follows: 15 kV accelerating voltage, 20 nA beam current, and <5 μm beam diameter. Natural minerals and synthetic oxides were used as standards, and the ZAF procedure was used for data correction. A portion of the Electron Microprobe (EMP) analyses was conducted with a JEOL at Hefei University of Technology.

(tholeiitic basalts) [Kuang et al., 2012]. The basalts are black and massive. The phenocrysts are olivine (<2%) and augite (5–10%), and the groundmass is composed of plagioclase (20–40%), pyroxene (30–40%), and minor olivine and magnetite. In this study, a subset of alkali basalts (LHT1, LHT9, and LHT19) and tholeiitic basalts (LHT20 and LHT25) from Kuang et al. [2012] was chosen. Note that Kuang et al. [2012] did not publish any geochemical data for sample LHT19, and its rock type was determined by comparison of the trace element patterns of cpx phenocrysts from this sample and other type-known samples (see supporting information Figure S1 for details).

3. Analysis Methods

Following the procedure of Liu et al., [2015a, 2015b], we used secondary ion mass spectrometry (SIMS) to measure the δ¹⁸O value of the cpx phenocrysts in these basalts. Compared with the laser fluorination method (LF), this approach allows us to avoid the bias caused by alteration as much as possible. In addition, the reason for our preference for cpx phenocrysts rather than olivine is that (1) olivine phenocrysts in some of our samples are rather sparse and (2) the matrix effect during the SIMS analysis for cpx could be more accurately constrained than for olivine. The rock samples were first cut and polished into double-polished thin sections (~150 μm thick), and then Fourier transform infrared spectrometry ana-

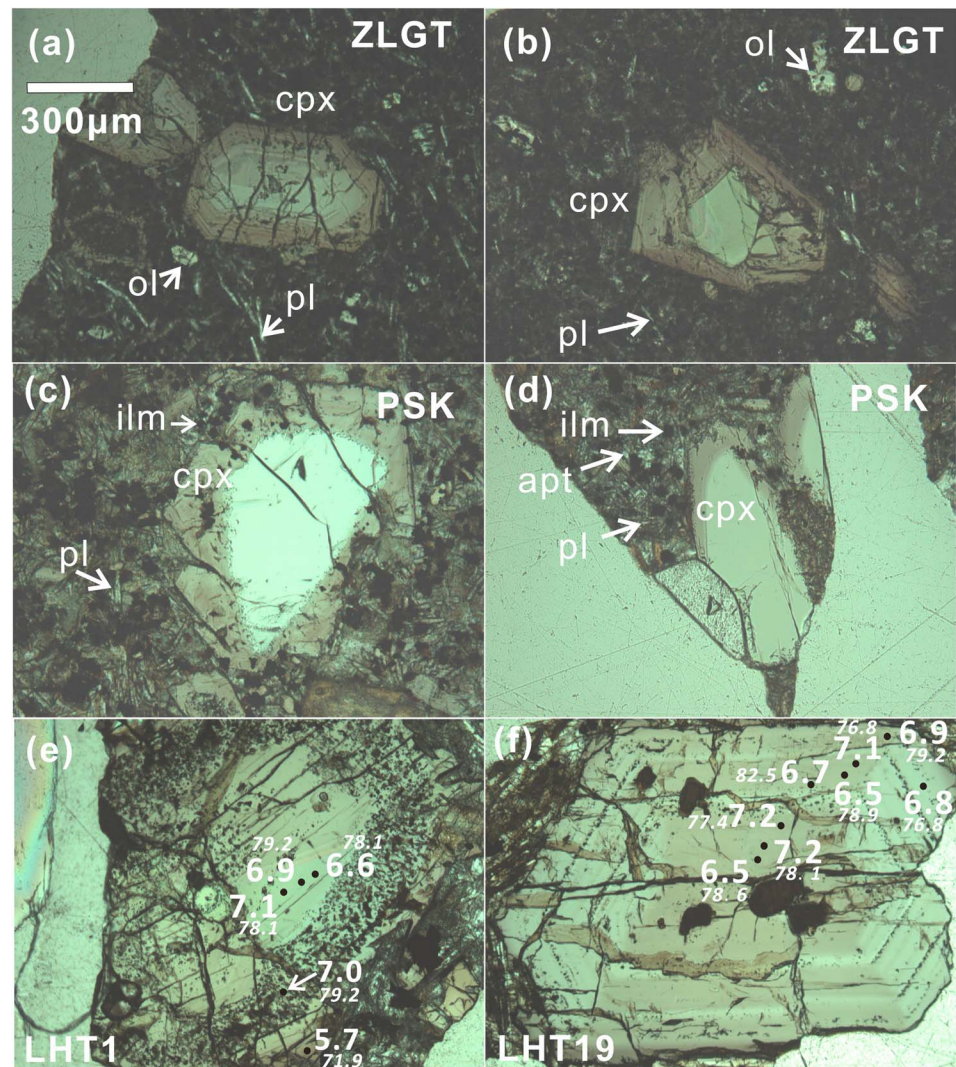


Figure 3. Petrologic photograph of clinopyroxene phenocrysts in the studied samples. All of the photographs are taken under plane-polarized light, and all slices have an approximate thickness of 100 μm . The dimensions for all photos are the same to that shown in Figure 3a. (a and b) Zoned cpx in Zhanglaogongtun (ZLGT) basalts. In Figure 3a, the core is magmatic and has higher Mg# than the rim, while in Figure 3b the core is crustal-derived xenocryst. (c and d) Zoned cpx in Pishikou (PSK) basanite. The cores are all crustal xenocrysts. The comparison of chemical composition of the cores and igneous rims is shown in Figure 4. (e and f) Cpx phenocrysts from Laohutai alkali basalts. The dark spots show the positions of SIMS analyses. The white bold numbers show the $\delta^{18}\text{O}$ values from secondary ion mass spectrometer (SIMS) analyses, and the italic small white numbers are the Mg# of the cpx analysis spots. Abbreviations: apt, apatite; ilm, ilmenite; pl, plagioclase.

Analytical conditions are similar to that at USTC. Cross comparison of the results from these two labs show that there are no systematic differences. The BSE images and Mg# calculated from the point-by-point data were used to identify the zonation and xenocrystic cores for all of the cpx families.

3.2. In Situ SIMS

The ion microprobe analyses were carried out using the CRPG-CNRS Cameca IMS-1280 instrument at Nancy (undated from IMS-1270). The analysis details are the same as in Liu *et al.* [2015a]. Four cpx megacrysts (NSH14, 2, 5, and 8) from the Nushan basanite, eastern China, with Mg# from 71.6 to 80.3 [Xia *et al.*, 2004], were used as standards to correct for the matrix effect. In each mount, a piece of cpx megacryst NSH6 (with an Mg# of 75.9) was measured in bracket to trace the possible instrumental drift. All of the

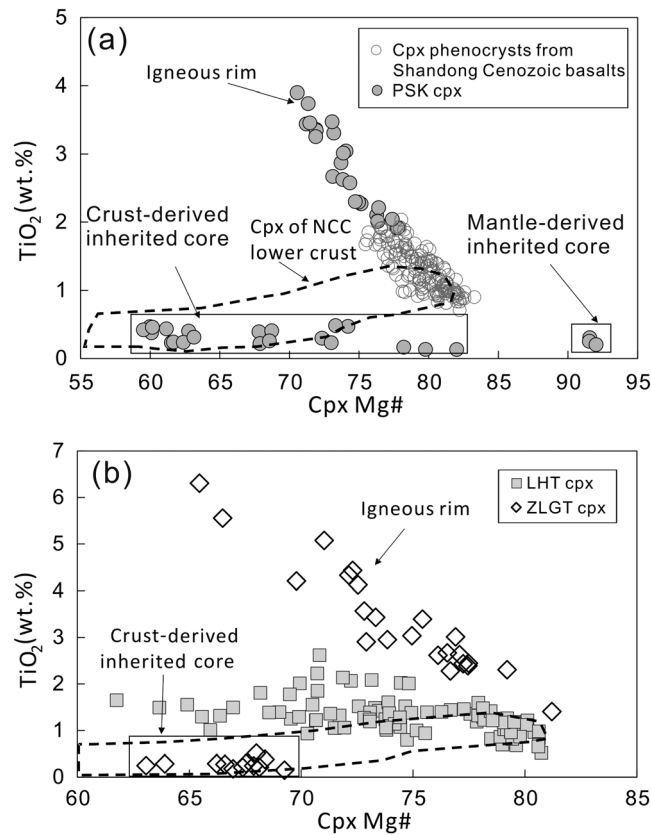


Figure 4. (a and b) TiO₂ versus Mg# plots for the cpx in the studied samples. The compositions of the cpx cores shown in Figure 3 are marked by real rectangles. In Figure 4a, the cpx phenocrysts from the late Cenozoic alkali basalts in Shandong are also plotted for comparison (data from Liu *et al.* [2015b]). The dashed lines mark the composition region of the cpx from the lower continental crust in NCC (data from Zhang *et al.* [2008, and references therein]).

(1 day) based on replicate analyses of standards is ~0.40‰ (2 SD). When the average of the analyses on one cpx grain are reported as the δ¹⁸O of that grain, the standard error of the mean are better than 0.3‰ (2 SE).

4. Results

4.1. Major Element Composition of Clinopyroxene Phenocrysts

The major element data of cpx phenocrysts are listed in supporting information Data Set S2. For the ZLGT alkali basalt, the cpx phenocrysts are usually chemically zoned. According to the differences in chemical composition and crystal form, the cpx cores could be classified in two groups: (1) In group 1, the cores are irregular and anhedral, their Mg# are lower than 70 (62–69), and their TiO₂ contents are lower than 0.5 wt%, which is consistent with the chemical composition of the cpx from the NCC lower continental crust (Figure 4a); (2) and in group 2, the cores are euhedral, and their TiO₂-Mg# plot shows the same trend defined by their igneous overgrowth rims (Figure 4a), which indicates that they are the early crystallized magmatic phenocrysts. Overall, the cpx crystallized from the magma is augitic to diopsidic, with Mg# from 82.4 to 65.4. For the cpx phenocrysts in the PSK basanites, the situation is similar to that of the ZLGT alkali basalts. Nearly all of the cpx phenocrysts have core-rim zoning, while the cores are xenocrysts from granulites or lithospheric mantle. As shown in Figure 4b, the igneous cpx have distinctively higher TiO₂ contents, and their Mg# ranges from 77 to 69. For the LHT alkali basalts and tholeiites, all of the cpx has TiO₂ higher than 0.50 wt%, and this value increases with decreasing Mg# (Figure 4a) along a wide trend, which indicates that all of these cpx are igneous phenocrysts. For all samples from these three locations, the highest Mg# of

mounts were coated with Au to prevent charging. An approximately 5–8 nA, 10 kV Cs⁺ primary ion beam current was used in focused mode, with a spot size of approximately 20 μm. The incidence of an electron flood gun was used to compensate for any sample charging. The secondary O⁻ ions were accelerated at 10 kV and analyzed at a mass resolution power of 3000, to resolve the ¹⁷OH isobaric interference on ¹⁸O. Oxygen isotopes were measured in multicollection mode, using two off-axis Faraday cups, L'2 for ¹⁶O, and H1 for ¹⁸O. Each analysis takes approximately 4 min, including 60 s of presputtering, ~40 s of automated centering and 150 s of integrating oxygen isotope signals. The internal precision for one spot analysis was typically better than 0.1‰ (2σ). For each cpx grain, three to eight spots were analyzed. The “matrix” effect, as described by Gurenko *et al.* [2001], was taken into account when using the correlation of instrumental mass fractionations (IMF) relative to the Mg# (Mg# = Mg/(Mg + Fe) mol) of the reference cpx (see supporting information Figure S2 for the calibration lines). The reproducibility of δ¹⁸O during a single analytical session

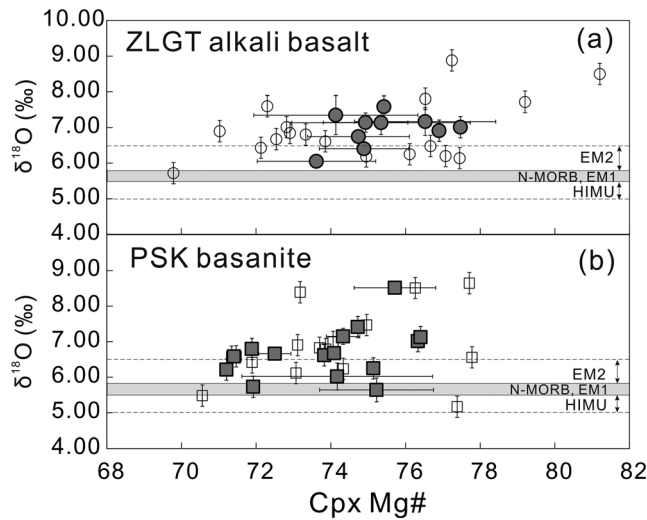


Figure 5. Oxygen isotope compositions of the cpx phenocrysts in the ZLGT and PSK basalts versus Mg#. The empty symbols are the $\delta^{18}\text{O}$ values for each single analysis spot, while the ones filled with dark gray color are the average for one specific cpx grain. The range of $\delta^{18}\text{O}$ values of cpx phenocrysts from N-MORB, EM1, EM2, and HIMU OIB, calculated from published $\delta^{18}\text{O}$ values of olivine phenocrysts [Eiler et al., 1997; Widom and Farquhar, 2003] and an equilibrium fractionation between cpx and olivine of 0.4‰, is indicated by the gray bars or double headed arrows. The error bar for a single point $\delta^{18}\text{O}$ is taken as 0.3‰ (1 SD), while the error bar for the average $\delta^{18}\text{O}$ for one cpx grain varies from 0.14 to 0.4‰ (1 SE).

For the ZLGT alkali basalts, the cpx $\delta^{18}\text{O}$ values (5.74–8.88‰) are all higher than the values for cpx from N-MORB (5.70‰) and are similarly scattered in the Mg# range of 70 to 80 (Figure 5a). For the PSK samples, the single point $\delta^{18}\text{O}$ values vary from 5.48‰ to 8.64‰, except for one point with $\delta^{18}\text{O}$ of 5.17‰ (Figure 5b). For the LHT alkali basalts (LHT1, LHT9, and LHT19), the majority of the cpx suite $\delta^{18}\text{O}$ values are in the range of 5.67‰ to 8.02‰, while three cpx grains in LHT19 and LHT1 exhibit $\delta^{18}\text{O}$ slightly lower than the N-MORB value (Figures 6a and 6b). For the LHT tholeiitic basalts, all of the cpx grains exhibit $\delta^{18}\text{O}$ close to or higher than the N-MORB value (Figure 6c).

5. Discussion

5.1. Origin of the Oxygen Isotopic Anomaly

The $\delta^{18}\text{O}$ values of the cpx phenocrysts in all of these basalts (Figures 5 and 6) are dominantly higher than that of the cpx from typical MORBs and peridotite xenoliths (5.4–5.8‰ [Mattey et al., 1994; Eiler et al., 1997]), and many of them are even higher than cpx $\delta^{18}\text{O}$ from EM2-type OIB lavas (5.8–6.5‰ [Eiler et al., 1997; Widom and Farquhar, 2003]). The range of $\delta^{18}\text{O}$ for the cpx in one basalt sample is similar to that of the cpx phenocrysts in the Klyuchevskoy volcano, Kamchatka (5.5–8.0‰) (calculated from $\delta^{18}\text{O}$ of olivine phenocrysts and $\Delta^{18}\text{O}_{\text{cpx-ol}} = 0.4\text{‰}$ [Auer et al., 2009]). Due to the small fractionation between cpx phenocrysts and basaltic melt ($\sim 0.1\text{‰}$, calculated from the $\Delta^{18}\text{O}_{\text{cpx-ol}} = 0.4\text{‰}$ and $\Delta^{18}\text{O}_{\text{melt-ol}} = \sim 0.4$ to 0.5‰ [Eiler, 2001]), the measured cpx $\delta^{18}\text{O}$ could be approximately considered as that of the basaltic melt. Multiple factors, such as magmatic processes (partial melting, fractional crystallization, and devolatilization), secondary alteration, and crustal contamination (or AFC processes), could change the primary oxygen isotopic composition of magma. In the following, the roles of these factors are discussed one by one.

A large number of natural observations and theoretical considerations show that for basaltic melts with MgO contents between 8 and 3 wt %, the partial melting, fractional crystallization, and devolatilization would not change the $\delta^{18}\text{O}$ of the magma to a level higher than 0.1‰ [Eiler, 2001, and references therein; Zhao and Zheng, 2003]. Thus, for our samples, their high bulk MgO contents (higher than 8 wt % [Kuang et al., 2012; Yang and Li, 2008; Zhang et al., 2008; this work]; see supporting information Data Set S1) would indicate

cpx is in equilibrium with the bulk rock if the Fe-Mg exchange coefficient (Kd) is assumed to be 0.275 ± 0.13 [Putirka et al., 2003].

4.2. Oxygen Isotopic Composition of Cpx Phenocrysts

The instrumental mass fractionation (IMF)-corrected oxygen isotope analyses of cpx phenocrysts (note that data for cpx xenocrysts and cpx with Mg# less than 70 are not shown) are plotted with Mg# in Figures 5 and 6, in which both the $\delta^{18}\text{O}$ of single points and the average for those in single grains are shown (Table 1). Overall, the main obvious characteristics of these $\delta^{18}\text{O}$ results are (1) oxygen isotopic compositions are rather heterogeneous, both on the intergrain and intragrain scale, and (2) except for very few spots (8 of 201 analyses) with $\delta^{18}\text{O}$ lower than 5.4‰, all other analyzed spots exhibit $\delta^{18}\text{O}$ higher than the normal mantle value (5.40–5.70‰ [Mattey et al., 1994; Eiler et al., 1997]).

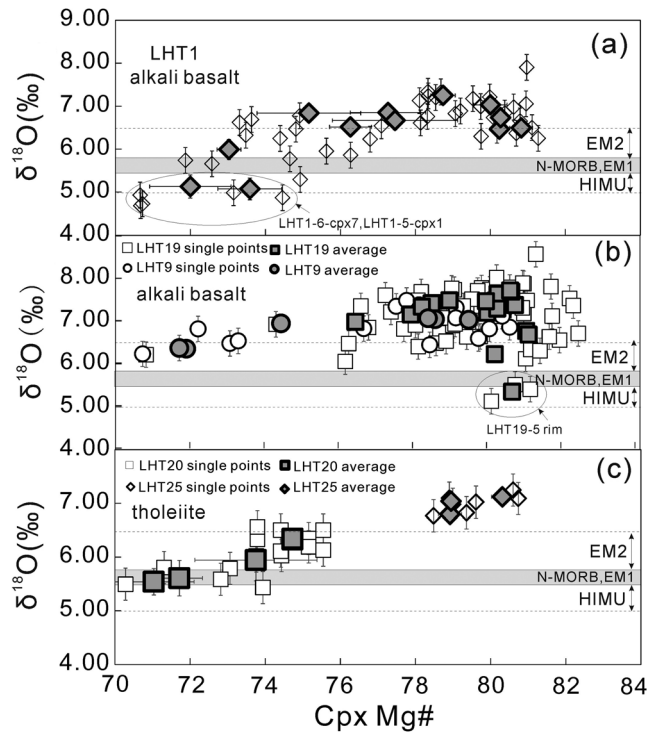


Figure 6. Oxygen isotope compositions of the cpx phenocrysts in the LHT alkali and tholeiitic basalts. The meaning of symbols and range of cpx from MORB and OIB are the same as in Figure 5. Note that most of the error bars (1 SE) of the grain average values are smaller than the scale of the symbols. The ellipses-marked analysis spots are that effected by mineral inclusions or the unperfect surface (see Figure S3).

rather similar MORB and OIB-like Nb/U ratios (36.2 for LHT25 and 35.8 for LHT20, respectively), which indicate that the relatively low $\delta^{18}\text{O}$ (close to N-MORB value) could not be explained by the crustal assimilation of LHT25 magma. Instead, this apparent trend between Mg# and $\delta^{18}\text{O}$ would be explained by the fact that the cpx in LHT20 are crystallized from a melt with relatively low $\delta^{18}\text{O}$ and more evolution (like fractional crystallization of mafic minerals in the plumbing system). At last, for all of the samples, the high $\delta^{18}\text{O}$ values are not correlated with the LOI (loss on ignition) values of the bulk rocks, the grain sizes, or the cracks in the cpx grains (Figure 3), which indicate that water-rock interaction after magma eruption does not shift the $\delta^{18}\text{O}$ value of cpx. However, the few spot of SIMS analyses with $\delta^{18}\text{O}$ lower than 5.40‰ in sample LHT19 and LHT1 (Figures 6a and 6b) could be due to the contaminant of tiny mineral inclusions (see supporting information Figures S3d and S3f) and the relief or kink in the mineral rim caused by unperfected polishing (see supporting information Figure S3a).

Overall, we consider that the dominantly high $\delta^{18}\text{O}$, especially for the cpx with higher Mg#, reflects the signature of the primary magmas, and thus the characteristics of the mantle sources. The spread of $\delta^{18}\text{O}$ among cpx grains in a single basalt sample is similar to the case in the Taihang and Shandong Cenozoic basalts ($\delta^{18}\text{O}$ interval up to 2‰ in one basalt sample [Liu et al., 2015a, 2015b]), the Canary Island basalts (La Gomera) [Gurenko et al., 2011], the Borgarhraun basalt flow in northern Iceland [Winpenny and Maclennan, 2014], and the Chifeng Cenozoic continental flood basalts [Wang et al., 2015], for which the $\delta^{18}\text{O}$ interval for minerals in one basalt sample could be up to 2‰. These types of heterogeneous $\delta^{18}\text{O}$ values were usually explained by a complex magma plumbing system for a very heterogeneous mantle-melting region [Gurenko et al., 2011; Winpenny and Maclennan, 2014], which is not the main scope of this paper.

5.2. Recycled Oceanic Components in the Mantle Sources

According to the discussion above, the higher-than-normal mantle $\delta^{18}\text{O}$ values of the cpx phenocrysts in all of these basalts (Figure 5) were inherited from their parental magmas and, consequently, the mantle sources,

that the oxygen anomaly were not caused by the simple magma evolution. In addition, the crustal contamination or assimilation also did not play a significant role. The bulk rock Ce/Pb and Nb/U ratios for the ZLGT, PSK, and LHT alkali basalts are ~25 and ~42, 19–24 and 40–46, and 27–70 and 32–40, respectively, which are consistent with the values for the global primary OIB and MORB basalts (Ce/Pb = 25 ± 5 , Nb/U = 40 ± 7 [Hofmann et al., 1986]). The bulk rock SiO₂ contents for the PSK basanites are 40–42 wt % (this study), and the bulk rock ⁸⁷Sr/⁸⁶Sr for LHT25 (tholeiite) is 0.70356 [Kuang et al., 2012]. Overall, these indexes argue against a significant role of the continental crust, which has rather low Ce/Pb and Nb/U ratios, high SiO₂ content, and enriched radiogenic isotopes [Rudnick and Gao, 2003].

Table 1. The In Situ $\delta^{18}\text{O}$ of Cpx Phenocryst in the ZLGT, PSK, and LHT Basalts

Sample	Cpx Grain	Spot	Measured $\delta^{18}\text{O}$	1 SD	Mg#	IMF ^a	Drift ^b	$\delta^{18}\text{O}$ Corrected	Average	1 SE	
QDPS7	7-5-cpx2	1-1-3	5.54	0.10	75.1	-1.09	0.38	6.25	6.25	-	
		1-2-3	6.40	0.09	73.1	-0.78	0.28	6.91			
	7-4-cpx1	1-2-4	6.12	0.11	71.9	-0.60	0.30	6.42	6.66	#NAME?	
		1-3-3	6.58	0.10	75.0	-1.06	0.17	7.47			
QDPS5	7-3-cpx1	1-3-4	6.15	0.10	73.7	-0.87	0.20	6.83	7.15	#NAME?	
		1-8-3	5.73	0.09	73.8	-0.89	0.00	6.62			
	7-4-cpx2	1-4-3	6.26	0.10	74.7	-1.03	-0.13	7.41	7.41	-	
		1-5-2	6.85	0.09	77.7	-1.48	-1.48	8.65			
	5-2-cpx3	1-5-4	6.98	0.10	76.3	-1.26	-1.26	8.51	8.52	#NAME?	
		1-5-6	7.39	0.10	73.2	-0.79	-0.79	8.39			
		1-10-2	5.13	0.10	71.9	-0.60	0.00	5.73			
		1-11-2	5.27	0.13	74.3	-0.97	0.00	6.24			
		1-11-3	6.06	0.10	74.1	-0.93	0.00	6.99			
		1-11-4	5.89	0.12	73.9	-0.90	0.00	6.79			
1-17-3		5.84	0.10	76.4	-1.28	0.00	7.13				
1-18-1-2		5.74	0.12	71.9	-0.60	-0.43	6.80				
1-18-2-2	3.22	0.13	77.4	-1.43	-0.52	5.17					
1-18-2-3	4.79	0.10	73.1	-0.78	-0.55	6.12					
QDPS2	2-4-cpx2	1-6-3	5.72	0.10	71.2	-0.50	0.00	6.21	6.21	-	
		1-16-3	5.40	0.11	76.3	-1.27	-0.34	7.01			
QDPS3	3-5-cpx1	1-15-1	4.71	0.10	70.6	-0.40	-0.37	5.48	7.01	-	
		1-15-2	4.67	0.08	77.8	-1.49	-0.40	6.56			
QDPS9	9-6-cpx2	1-13-2	6.06	0.11	71.3	-0.52	0.00	6.58	6.02	0.38	
		1-13-3	6.05	0.08	71.5	-0.54	0.00	6.59			
		1-13-3	6.05	0.08	71.5	-0.54	0.00	6.59			
JG2	JG2-1-CPX1	5-1-1	6.06	0.10	76.5	-1.20	-0.54	7.80	7.14	0.27	
		5-1-4	5.56	0.10	73.3	-0.75	-0.50	6.80			
		JG2-6-1	5-2-4	6.11	0.10	72.8	-0.67	-0.23			7.01
		5-2-5	5.05	0.11	76.7	-1.22	-0.21	6.48			
		JG2-3-1B	5-12-4	5.98	0.11	73.9	-0.82	0.19			6.61
JG3	JG3-1-1	5-12-5	6.32	0.09	79.2	-1.58	0.19	7.72	7.17	0.39	
		5-3-1-1	5.47	0.08	76.9	-1.25	-0.19	6.91			
		JG3-7-1	5-4-1	6.64	0.11	81.2	-1.87	0.01			8.50
		5-4-2	6.07	0.10	72.5	-0.63	0.03	6.67			
		5-4-3	7.06	0.09	72.3	-0.60	0.06	7.60			
	JG3-4-2	5-5-2	6.26	0.13	72.9	-0.69	0.10	6.85	7.14	0.33	
		5-5-3	5.04	0.13	77.1	-1.28	0.12	6.20			
		5-5-4	4.95	0.09	77.5	-1.33	0.14	6.14			
		5-5-5	6.02	0.09	72.1	-0.57	0.17	6.43			
		JG3-2-1	5-7-1	5.40	0.11	74.9	-0.98	0.21			6.19
JG1	JG1-1-2	5-7-3	5.34	0.09	76.1	-1.14	0.23	6.25	6.40	0.14	
		5-8-2	6.88	0.10	75.4	-1.04	0.34	7.59			
		JG3-1-3	5-11-1	5.86	0.11	77.5	-1.34	0.19			7.01
		5-6-1	6.97	0.10	77.2	-1.30	-0.59	8.88			
		5-6-5	5.91	0.11	71.0	-0.42	-0.57	6.90			
		5-6-5	5.91	0.11	71.0	-0.42	-0.57	6.90			
		5-6-5	5.91	0.11	71.0	-0.42	-0.57	6.90			
LHT1	LHT1-3-cpx5	2-11-1	4.75	0.09	80.79	-0.96	-0.95	6.66	6.47	0.09	
		2-11-2	4.60	0.07	80.18	-0.89	-0.95	6.44			
		2-11-3	4.50	0.08	79.75	-0.85	-0.95	6.30			
		2-13-1	4.26	0.08	81.27	-1.01	-0.99	6.25			
		2-13-2	4.87	0.08	80.08	-0.88	-0.99	6.74			
	LHT1-2-cpx1	2-13-3	4.52	0.08	81.11	-0.99	-0.99	6.50	6.50	0.11	
		2-13-4	5.50	0.06	78.54	-0.72	-0.99	7.20			
		2-13-5	5.50	0.06	73.63	-0.20	-0.99	6.69			
	LHT1-2-cpx1'	2-13-6	5.47	0.08	73.31	-0.17	-0.99	6.62	6.84	0.15	
		2-12-1	4.99	0.09	77.09	-0.57	-0.99	6.54			
		2-12-2	4.71	0.09	76.79	-0.54	-0.99	6.23			
		2-12-3	5.45	0.08	74.91	-0.34	-0.99	6.78			
	LHT1-3-cpx4	2-10-1	5.09	0.09	80.95	-0.97	-1.00	7.06	6.52	0.13	
		2-10-2	5.06	0.08	79.06	-0.77	-1.00	6.83			
		2-10-3	5.35	0.09	79.98	-0.87	-1.00	7.22			
		2-10-4	5.65	0.08	78.34	-0.70	-1.00	7.34			
		2-10-5	5.55	0.07	78.34	-0.70	-1.00	7.24			

Table 1. (continued)

Sample	Cpx Grain	Spot	Measured $\delta^{18}\text{O}$	1 SD	Mg#	IMF ^a	Drift ^b	$\delta^{18}\text{O}$ Corrected	Average	1 SE
		2-10-6	5.36	0.09	79.53	-0.82	-1.00	7.18	7.25	0.04
	LHT1-6-cpx2	3-3-1	3.99	0.09	80.38	-1.71	-1.14	6.84		
		3-3-2	4.24	0.10	79.74	-1.61	-1.14	6.99		
		3-3-3	3.46	0.11	80.69	-1.76	-1.15	6.37	6.74	0.15
	LHT1-6-cpx2'	3-3-4	4.34	0.10	74.39	-0.74	-1.17	6.25		
		3-3-5	4.92	0.09	80.97	-1.80	-1.18	7.90	7.08	0.58
	LHT1-6-cpxX	3-2-2-1	4.02	0.10	72.57	-0.44	-1.19	5.66		
		3-2-2-2	4.54	0.09	73.48	-0.59	-1.20	6.33	5.99	0.24
	LHT1-6-cpxx	3-2-3-1	4.18	0.09	78.32	-1.38	-1.20	6.76		
		3-2-3-2	4.69	0.10	78.68	-1.43	-1.21	7.33		
		3-2-3-3	4.45	0.11	74.81	-0.81	-1.22	6.47	6.85	0.21
	LHT1-6-cpx7	3-1-1	3.73	0.09	74.65	-0.78	-1.26	5.78		
		3-1-2	3.58	0.09	70.66	-0.13	-1.22	4.93 ^c		
		3-1-3	3.38	0.11	70.67	-0.14	-1.17	4.68 ^c	5.13	0.27
	LHT1-5-cpx1	3-5-1	4.67	0.12	78.11	-1.34	-1.12	7.13		
		3-5-2	4.30	0.10	79.20	-1.52	-1.07	6.89		
		3-5-3	4.24	0.10	78.14	-1.35	-1.02	6.61		
		3-5-4	4.44	0.09	71.86	-0.33	-0.97	5.74		
		3-5-5	4.31	0.11	80.62	-1.75	-0.92	6.98	6.67	0.22
	LHT1-5-cpx4	3-4-1	4.24	0.12	75.62	-0.94	-0.77	5.95		
		3-4-2	4.03	0.10	76.27	-1.04	-0.78	5.86		
		3-4-3	3.68	0.13	74.93	-0.82	-0.79	5.29 ^c	5.91	#NAME?
		3-4-4	3.32	0.10	74.45	-0.75	-0.80	4.86 ^c		
		3-4-5	3.64	0.12	73.15	-0.54	-0.81	4.98 ^c		
		3-4-6	3.76	0.12	70.72	-0.14	-0.82	4.72 ^c		
LHT11	LHT11-cpxN1	3-6-1	4.02	0.12	79.43	-1.56	-1.11	6.69		
		3-6-2	4.40	0.09	79.49	-1.57	-1.11	7.08		
		3-6-3	4.67	0.10	79.49	-1.57	-1.12	7.36		
		3-6-4	4.15	0.09	79.07	-1.50	-1.13	6.77	6.97	0.13
LHT19	LHT19-5-cpx6	2-9-1	5.02	0.07	78.74	-0.74	-0.95	6.71		
		2-9-2	5.62	0.08	80.83	-0.96	-0.95	7.53		
		2-9-3	5.44	0.08	80.47	-0.92	-0.95	7.31	7.18	0.20
	LHT19-5-cpx6'	2-9-4	5.44	0.07	80.02	-0.87	-0.95	7.27		
		2-9-52-9-52-9-5	5.96	0.08	79.86	-0.86	-0.95	7.77		
		2-9-62-9-62-9-6	5.94	0.10	81.00	-0.98	-0.95	7.87	7.64	0.15
	LHT19-5 core	2-9-1-2	4.17	0.07	81.05	-0.98	-0.95	6.10		
		2-9-1-3	4.37	0.06	81.19	-1.00	-0.95	6.32	6.21	0.08
	LHT19-5 rim	2-9-1-4	3.27	0.07	80.13	-0.89	-0.95	5.11 ^c		
		2-9-1-5	3.45	0.09	81.17	-1.00	-0.95	5.39 ^c		
		2-9-1-6	3.60	0.08	80.77	-0.95	-0.95	5.50 ^c	5.33	0.10
	LHT19-5-cpx1	2-7-2-1	5.14	0.12	80.61	-1.78	-0.48	7.40		
		2-7-2-2	5.00	0.11	80.47	-1.76	-0.48	7.24		
		2-7-2-3	5.07	0.09	79.78	-1.66	-0.48	7.21	7.28	0.05
	LHT19-5-cpx1'	2-7-2-4	5.17	0.10	79.76	-1.66	-0.48	7.31		
		2-7-2-5	5.66	0.09	76.61	-1.21	-0.48	7.35		
		2-7-2-6	5.53	0.08	80.62	-1.79	-0.48	7.79	7.48	0.12
	LHT19-5	2-7-1-1	4.09	0.08	81.96	-1.98	-0.48	6.54		
		2-7-1-2	4.52	0.10	79.47	-1.62	-0.48	6.62		
		2-7-1-3	4.69	0.10	81.76	-1.95	-0.48	7.11	6.76	0.15
	LHT19-5-cpx2	2-8-1	5.61	0.08	79.13	-1.57	-0.56	7.73		
		2-8-2	5.68	0.09	78.24	-1.44	-0.56	7.68		
		2-8-3	5.09	0.09	80.02	-1.70	-0.56	7.35		
		2-8-4	4.96	0.07	82.23	-2.02	-0.56	7.53		
		2-8-5	4.77	0.10	82.33	-2.03	-0.56	7.36		
		2-8-6	5.35	0.09	78.92	-1.54	-0.56	7.44		
		2-8-7	5.10	0.11	79.16	-1.58	-0.56	7.23		
		2-8-8	5.11	0.11	79.85	-1.68	-0.56	7.34	7.46	0.06
	LHT19-5-cpxN1	2-3-1	5.17	0.09	80.79	-1.81	-0.62	7.60		
		2-3-2	4.98	0.10	80.50	-1.77	-0.59	7.34		
		2-3-3	4.76	0.09	81.00	-1.84	-0.57	7.17	7.37	0.10
	LHT19-5-cpx4	2-4-1-1	5.48	0.10	80.09	-1.71	-0.55	7.74		

Table 1. (continued)

Sample	Cpx Grain	Spot	Measured $\delta^{18}\text{O}$	1 SD	Mg#	IMF ^a	Drift ^b	$\delta^{18}\text{O}$ Corrected	Average	1 SE
		2-4-1-2	5.52	0.10	79.76	-1.66	-0.53	7.71		
		2-4-1-3	5.36	0.11	81.73	-1.94	-0.50	7.80		
		2-4-1-4	5.12	0.09	81.09	-1.85	-0.50	7.48	7.68	0.06
	LHT19-5-cpx5	2-6-1-1	5.66	0.10	78.14	-1.43	-0.40	7.48		
		2-6-1-2	5.05	0.10	77.77	-1.38	-0.38	6.81	7.14	0.24
	LHT19-5-cpx7	2-1-1	4.03	0.11	81.44	-1.90	-0.35	6.29		
		2-1-2	4.35	0.10	81.69	-1.94	-0.33	6.62		
		2-1-3	5.06	0.10	80.27	-1.73	-0.31	7.10	6.67	0.19
	LHT19-5-cpx4	2-2-1	5.18	0.11	80.96	-1.83	-0.29	7.30		
		2-2-2	6.02	0.11	80.27	-1.73	-0.26	8.02		
		2-2-3	5.78	0.11	74.32	-0.89	-0.24	6.91	7.41	0.26
	LHT19-5-cpx9	2-5-1-1	5.10	0.12	79.09	-1.57	-0.22	6.89		
		2-5-1-2	5.67	0.09	79.60	-1.64	-0.20	7.51		
		2-5-1-3	6.51	0.10	81.33	-1.89	-0.18	8.57		
		2-5-1-4	5.90	0.10	80.97	-1.83	-0.15	7.89	7.71	0.30
	LHT19-5-CPX8	4-14-1-1	6.36	0.08	77.27	-1.21	-0.03	7.60		
		4-14-1-2	5.68	0.12	78.13	-1.34	-0.02	7.04		
		4-14-1-3	5.80	0.09	79.37	-1.55	0.00	7.35	7.33	0.13
	LHT19-5-CPX8	4-14-2-1	6.28	0.10	79.06	-1.50	0.02	7.76		
		4-14-2-2	6.07	0.09	70.82	-0.16	0.03	6.20		
		4-14-2-3	5.44	0.08	79.52	-1.57	0.05	6.96	6.97	0.37
	LHT19	5-10-1	5.53	0.10	79.82	-1.67	0.57	6.63		
		5-10-2	5.49	0.11	78.15	-1.43	0.54	6.39		
		5-10-3	5.97	0.10	78.08	-1.42	0.50	6.90		
		5-10-4	5.75	0.10	76.28	-1.17	0.46	6.46		
		5-10-5	5.30	0.09	76.19	-1.15	0.42	6.04	6.48	0.13
	LHT19	5-9-1	5.40	0.11	78.64	-1.50	0.43	6.47		
		5-9-2	6.24	0.10	78.16	-1.43	0.45	7.23		
		5-9-3	6.35	0.10	77.42	-1.33	0.47	7.21		
		5-9-4	5.14	0.08	82.47	-2.05	0.49	6.70		
		5-9-5	5.49	0.10	78.91	-1.54	0.51	6.52		
		5-9-6	6.08	0.11	78.79	-1.52	0.54	7.07		
		5-9-7	5.90	0.11	79.15	-1.58	0.56	6.92		
		5-9-8	6.18	0.10	76.82	-1.24	0.58	6.84	6.87	0.10
LHT9	LHT9-CPX10	4-4-1	5.38	0.08	73.08	-0.53	-0.56	6.46		
		4-4-2	5.53	0.09	70.73	-0.14	-0.54	6.22	6.34	0.09
	LHT9-CPX11	4-3-1	5.27	0.08	79.18	-1.51	-0.52	7.31		
		4-3-2	5.51	0.11	78.18	-1.35	-0.51	7.37		
		4-3-3	4.54	0.09	78.47	-1.40	-0.49	6.43	7.04	0.25
	LHT9-CPX8	4-1-1-1	5.66	0.09	77.56	-1.25	-0.43	7.34		
		4-1-1-2	4.55	0.09	79.78	-1.61	-0.41	6.58		
		4-1-1-3	5.55	0.09	77.92	-1.31	-0.40	7.26	7.06	0.20
	LHT9-CPX8'	4-1-2-1	5.69	0.10	71.72	-0.30	-0.37	6.36	6.36	-
	LHT9-cpx7	3-11-1	5.02	0.12	73.31	-0.56	-0.94	6.52		
		3-11-2	5.23	0.10	77.84	-1.30	-0.95	7.48		
		3-11-3	5.46	0.10	72.22	-0.39	-0.96	6.81	6.94	0.23
	LHT9-cpx1	3-10-1	4.09	0.10	80.62	-1.75	-1.00	6.85		
		3-10-2	4.38	0.11	80.42	-1.72	-1.01	7.11		
		3-10-3	4.81	0.11	80.11	-1.67	-1.02	7.50		
		3-10-4	4.52	0.07	79.17	-1.51	-1.03	7.07		
		3-10-5	4.10	0.12	80.08	-1.66	-1.04	6.81		
		3-10-6	4.65	0.10	76.69	-1.11	-1.05	6.81	7.02	0.10
LHT20	LHT20-2-cpx3	4-6-1	4.63	0.09	74.43	-0.75	-0.65	6.03		
		4-6-2	4.63	0.10	73.06	-0.52	-0.64	5.79		
		4-6-3	4.76	0.09	73.76	-0.64	-0.62	6.02	5.95	0.06
	LHT20-1-cpx1	4-5-3	5.17	0.11	70.28	-0.07	-0.26	5.49		
		4-5-4	4.81	0.08	72.82	-0.48	-0.29	5.59	5.54	0.13
	LHT20-3-CPX3	4-7-1	4.96	0.10	71.31	-0.24	-0.61	5.81		
		4-7-2	4.99	0.08	69.90	-0.01	-0.59	5.59		
		4-7-3	4.19	0.07	73.94	-0.66	-0.57	5.43	5.61	0.09
	LHT20-4-CPXN1	4-11-1	5.41	0.09	74.41	-0.74	-0.35	6.50		

Table 1. (continued)

Sample	Cpx Grain	Spot	Measured $\delta^{18}\text{O}$	1 SD	Mg#	IMF ^a	Drift ^b	$\delta^{18}\text{O}$ Corrected	Average	1 SE	
LHT25	LHT20-4-N2	4-11-2	5.15	0.08	75.15	-0.86	-0.33	6.35	6.33	0.07	
		4-11-3	5.38	0.10	73.79	-0.64	-0.32	6.33			
		4-11-4	4.90	0.08	75.54	-0.93	-0.30	6.13			
		4-8-1	5.19	0.08	74.41	-0.74	-0.18	6.11			
		4-8-2	5.17	0.07	75.15	-0.86	-0.16	6.19			
	LHT25-1-cpx1	4-8-3	5.77	0.10	73.79	-0.64	-0.15	6.56	6.34	0.10	
		4-8-4	5.45	0.09	75.54	-0.93	-0.13	6.50			
		3-9-1	4.41	0.09	79.36	-1.54	-0.87	6.82			
		3-9-2	4.48	0.09	78.48	-1.40	-0.88	6.77			
		3-8-2-1	4.60	0.09	78.99	-1.48	-0.89	6.98			
	LHT25-3-cpx1'	3-8-2-2	4.72	0.09	78.90	-1.47	-0.90	7.10	7.04	0.04	
		LHT25-3-cpx1	3-8-1-1	4.41	0.10	80.73	-1.77	-0.91			7.09
		3-8-1-2	4.58	0.11	80.60	-1.74	-0.92	7.25			
		3-8-1-3	4.50	0.10	79.61	-1.58	-0.93	7.02			
											7.12

^aIMF is instrumental mass fractionation.

^bThe instrumental drift, which is revealed by the bracketing measurements of standard mounted in the sample block and is calculated according to a linear relationship with time.

^cThese measured values were affected by the mineral inclusion and/or relief of mineral rim (see Figure S3).

which indicates that recycled crust or crustal derivatives with high $\delta^{18}\text{O}$ values must have been involved in their mantle sources [Eiler *et al.*, 1997; Eiler, 2001]. The possible candidates specifically point to continental crust [Eiler, 2001], fluids/melts expelled from the subducted slab [Auer *et al.*, 2009, and references therein; Martin *et al.*, 2011] or oceanic crust that has experienced low- T ($<350^\circ\text{C}$) water-rock interaction [Muehlenbachs, 1986; Taylor and Sheppard, 1986; Eiler, 2001] with or without marine sediments. Similar to the discussion of Liu *et al.* [2015a] for the origin of high cpx $\delta^{18}\text{O}$ values in Taihang basalts, the high $(\text{Nb}/\text{La})_n$ (n represent primitive mantle normalization), Nb/U, and Ce/Pb ratios in our samples (Figure 3) argue against the significant role of continental crust ($(\text{Nb}/\text{La})_n$, Ce/Pb, and Nb/U ratios of the average continental crust are 0.4, 3.9, and 6.1, respectively [Rudnick and Gao, 2003]) or fluids expelled from the subducted slab because of their low corresponding values [Tatsumi *et al.*, 1986; Grove *et al.*, 2002] ($(\text{Nb}/\text{La})_n$ ratios of fluids in the mantle wedge are always as low as 0.19–0.57, although the Ce/Pb ratio could be up to ~ 23). Thus, the high $\delta^{18}\text{O}$ values in our samples exclusively indicate the involvement of components from oceanic crust in the mantle sources. This is consistent with the previous suggestions from studies on Mg stable isotopes for the ZLGT basalts ($\delta^{26}\text{Mg}$ low to -0.6‰ , usually attributed to marine carbonates [Yang *et al.*, 2012]) and a more enriched $^{87}\text{Sr}/^{86}\text{Sr}$ at a given $\epsilon\text{Nd}(t)$ value relative to the global MORB Sr-Nd isotopic trend for the LHT alkali basalts and PSK basanites (Figure 2c) [Kuang *et al.*, 2012; Zhang *et al.*, 2008].

5.3. The Temporal Variation of the Recycled ROC Components

In the following sections, we first compare the $\delta^{18}\text{O}$ values of the Late Cretaceous (PSK basanites) to late Cenozoic basalts in the southeast NCC with that of their hosted mantle-derived xenoliths, which helps us to identify how the ROC were incorporated into the upper mantle during this time period. We then focus on the temporal variation of $\delta^{18}\text{O}$ for cpx phenocrysts in the 106 to ~ 0 Ma basalts (e.g., LHT basalts, 70–60 Ma) from the northern margin of the NCC, which may provide a more convincing identification of the nature of recycled oceanic crust. Finally, some of the implications of these $\delta^{18}\text{O}$ data are discussed briefly.

5.3.1. Contrast $\delta^{18}\text{O}$ Between the Lithospheric Mantle and Basalts in SE NCC

As shown in Figure 7, the oxygen isotopic composition ($\delta^{18}\text{O}$) of the olivine phenocrysts in the Late Cretaceous PSK basanites (calculated from $\delta^{18}\text{O}_{\text{Cpx}}$ from this study with $\Delta^{18}\text{O}_{\text{Cpx-ol}} = 0.4\text{‰}$ [Mattey *et al.*, 1994]) and the late Cenozoic alkali-tholeiitic basalts in the Suwan-Shandong region [Zhang *et al.*, 2009; Z. Xu *et al.*, 2012; Wang *et al.*, 2011; Liu *et al.*, 2015b] are mainly higher than and lower than the normal mantle value, respectively.

However, the lithospheric mantle at these two time periods shows the reverse $\delta^{18}\text{O}$ anomalies to their hosting basalts (Figure 7). The Late Cretaceous lithospheric mantle in the southeastern NCC was suggested to consist of a newly accreted fertile mantle and an overlying residual, old and refractory

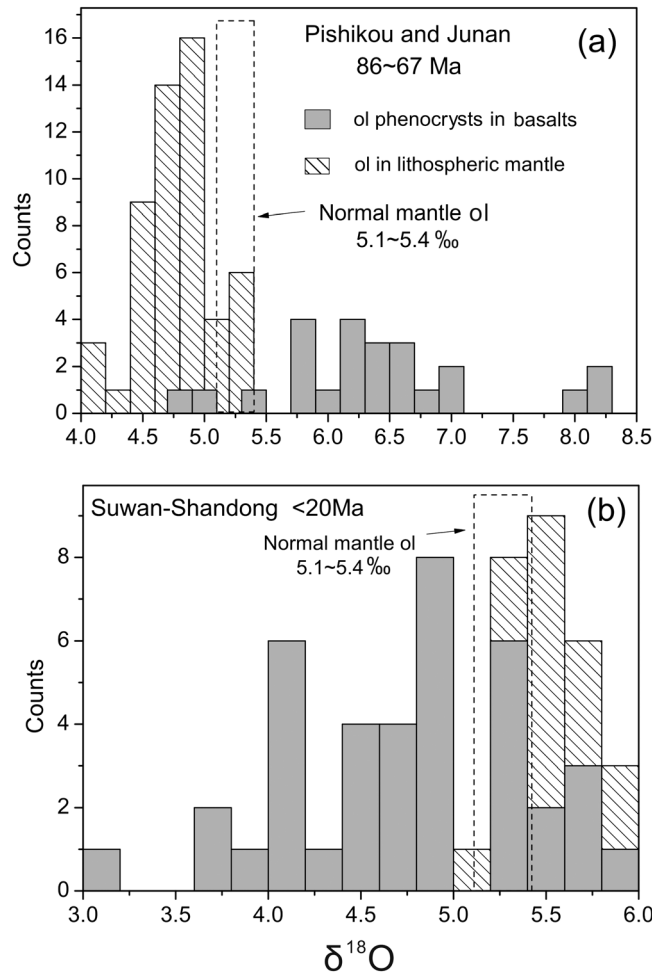


Figure 7. Comparison of the $\delta^{18}\text{O}$ of the olivine phenocrysts with that of olivine from the contemporaneous lithospheric mantle. The dashed vertical boxes mark the $\delta^{18}\text{O}$ range of olivine phenocrysts from N-MORB [Eiler et al., 1997]. The $\delta^{18}\text{O}$ of olivine phenocrysts from PSK basanite are calculated from the $\delta^{18}\text{O}_{\text{Cpx}}$ in this study and $\Delta^{18}\text{O}_{\text{ol-cpx}} = -0.4\text{‰}$ [Matthey et al., 1994]. The $\delta^{18}\text{O}_{\text{ol}}$ for the Suwan-Shandong late Cenozoic basalts includes those calculated from $\delta^{18}\text{O}_{\text{Cpx}}$ [Liu et al., 2015b] and those from Zhang et al. [2009], Wang et al. [2011], and Z. Xu et al. [2012] (the data set with $\Delta^{18}\text{O}_{\text{Cpx-ol}} < 0$ are filtered). The data of $\delta^{18}\text{O}_{\text{ol}}$ for 86–67 Ma and late Cenozoic NCC lithospheric mantle are from Guo et al., 2013 and Hao et al., 2015.

Late Cretaceous and late Cenozoic time is not clear now, the possible reasons, however, would be (1) the oxygen isotopic anomalies were formed during the cooling and accretion process of the upwelling asthenosphere, which had contain some components derived from the subducted oceanic slab; and/or (2) the long-time metasomatism by the slab-derived fluids/melt, which has been suggested to explain the oxygen isotopic disequilibrium between opx, cpx, and olivines [Hao et al., 2015]. Alternatively, these secular changes in $\delta^{18}\text{O}$ of lithospheric mantle may also reflect the spatial geochemical heterogeneity, considering that the Suwan-Shandong region is ~500 km away from the PSK region.

Overall, whatever the specific mechanism, the clearest information by this study is that in southeastern NCC, at Late Cretaceous and late Cenozoic, the oxygen isotopes of the basalts are distinct from that of the lithospheric mantle, which strongly indicate that these basalts were not derived from the lithospheric mantle. On the other hand, the secular changes in $\delta^{18}\text{O}$ anomaly for the lithospheric mantle and the contemporary basalts (Figure 7) could be best explained by that the components from the subducted Pacific slab (e.g., upper ROC versus lower ROC) were dynamically supplied to the upper mantle beneath

mantle [Ying et al., 2006; Zhang et al., 2011]. Guo et al. [2013] reported the low $\delta^{18}\text{O}$ ($<5.4\text{‰}$) of olivine as xenocrysts or in the peridotite hosted in Jūnan and PSK basalts. These olivines with Mg# ranging from 87 to 89 indicate these samples would come from a fertile part of the lithospheric mantle [Ying et al., 2006]. The authors explained these low $\delta^{18}\text{O}$ olivines as the separates from the mantle metasomatized or hybridized by the partial melts of a high- T hydrothermally altered ROC. This kind of metasomatized/hybridized production in the lithospheric mantle were widely suggested to be the source of Cenozoic basalts in NCC [Zhang et al., 2009; Z. Xu et al., 2012; Wang et al., 2011; Dai et al., 2016]. As the contrast, the late Cenozoic lithospheric mantle in Suwan-Shandong area has $\delta^{18}\text{O}$ higher than the normal mantle value (5.5‰ on average [Hao et al., 2015]). The studied peridotite xenoliths show olivine Mg# from 89 to 91, and the calculated equilibrium temperature are from 900 to 1200°C [Hao et al., 2015]. According to the typical Cenozoic geothermal gradient in eastern China [Huang and Xu, 2010], the depths of these xenoliths are estimated to be from 40 km to the spinel-garnet peridotite transition zone of ~65 km. This means that the high $\delta^{18}\text{O}$ should be the characteristics of large portion of the lithospheric mantle at this time period. The mechanism for the contrast $\delta^{18}\text{O}$ of lithospheric mantle in the

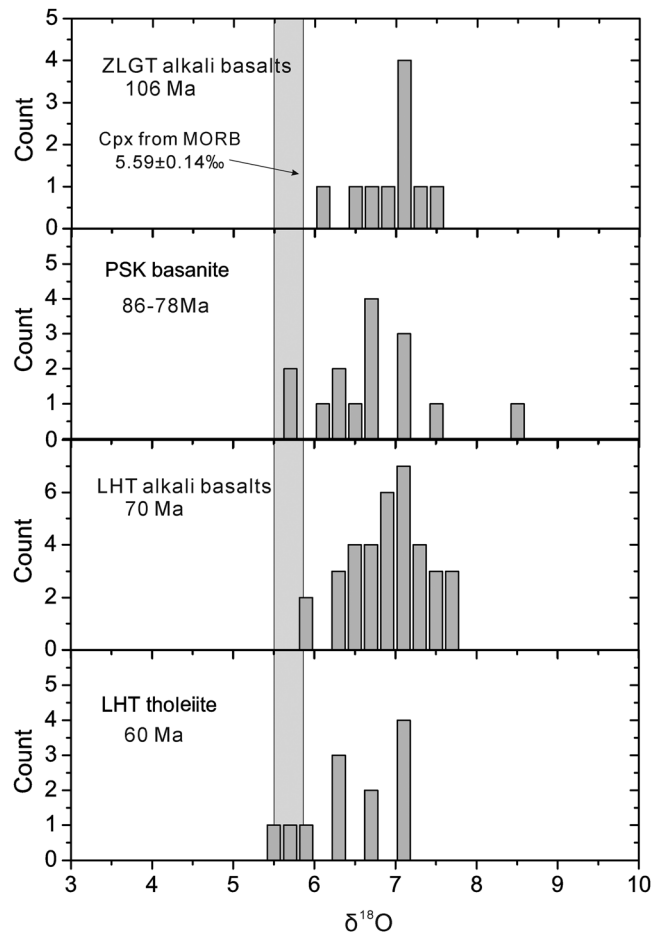


Figure 8. Histograms of $\delta^{18}\text{O}$ of cpx phenocrysts in the mafic rocks from 106 Ma to 60 Ma. All of the $\delta^{18}\text{O}$ values are for the single-spot analysis results. The light gray vertical box marks the $\delta^{18}\text{O}$ value for the cpx phenocrysts crystallized from N-MORB [Eiler *et al.*, 1997].

the southeast NCC over a long time period. This means that the recycled oceanic components responsible for the OIB-type basalts in eastern China were not always having resided in the lithospheric mantle since the Early Cretaceous [Zhang *et al.*, 2009; Z. Xu *et al.*, 2012].

5.3.2. Evolution of the ROC Components in the Mantle Sources of 106 Ma to Late Cenozoic Basalts in the Northern Margin of NCC

Figure 8 illustrates the variation of $\delta^{18}\text{O}_{\text{cpx}}$ for the 106–60 Ma basalts. In Figure 9, accompanying with the published data of the Cenozoic basalts in the Central NCC and Southeastern NCC, the average $\delta^{18}\text{O}_{\text{cpx}}$ within each sample are compared with the bulk rock Ce/Pb, $^{87}\text{Sr}/^{86}\text{Sr}$, Ba/Th, and Nb/La ratios.

As shown in Figure 8, in the northern margin of NCC, the $\delta^{18}\text{O}_{\text{cpx}}$ of basalts from 106 Ma to 60 Ma are persistently significantly higher than that of the normal mantle. Due to the considerably low $\delta^{18}\text{O}$ of the altered lower oceanic crust (3.4‰ to 5.8‰ [Gregory and Taylor, 1981]), the generally high $\delta^{18}\text{O}$ of these samples indicate that the recycled components in the mantle sources were derived from the upper altered ROC.

As shown in Figure 9, although the average $\delta^{18}\text{O}_{\text{cpx}}$ of basalt sample does not show any clear correlation with the bulk (Nb/La)*n*, Ba/Th, and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, the modeling shows that the high $\delta^{18}\text{O}_{\text{cpx}}$ of these basalts could be explained by the mixing of three end-member components, DMM, upper ROC and oceanic sediments. It could also be identified that the ZLGT basalts (106 Ma) have more contribution from the upper ROC than other younger counterparts, and both the alkali and tholeiitic LHT basalts seem have similar portion of contribution from those two hybridized sources, which means that the sediments components relative to the ROC components are variable with time.

At the Late Cretaceous, the situation in Southeastern NCC seems a bit different from the case in the northern margin of NCC. As we have shown in Figure 7, although the basalts (PSK) show obvious contribution from upper ROC, the $\delta^{18}\text{O}$ of the lithospheric mantle (lower than the normal mantle value) indicates that some derivatives from lower ROC have been involved in the upper mantle before 87 Ma. From early Cenozoic on, the lower ROC components increasingly appeared in the mantle sources of basalts eastern China widely. The $\delta^{18}\text{O}$ of cpx in basalts in Shuangliao (51 Ma to 40 Ma [Y. G. Xu *et al.*, 2012]), Northeast China, varies from 5.0 to 7.0‰ (basalts at 40 Ma have comparatively lower $\delta^{18}\text{O}_{\text{cpx}}$, from 4.0 to 6.0‰, Chen [2017]), which indicates that the upper and lower ROC have simultaneously played their roles in the mantle source. Such a situation was more extensive during the late Cenozoic. As shown in Figure 9, the nearly synchronical basalts in the Central NCC (Taihang and Chifeng 25 Ma–5.6 Ma and 25 Ma–2.5 Ma, respectively) and the southeastern NCC (Suwan-Shandong, <25 Ma) show obviously upper and lower ROC signatures, respectively. For Changle basalts, both upper and lower ROC have been involved in the source (Figures 9c and 9d).

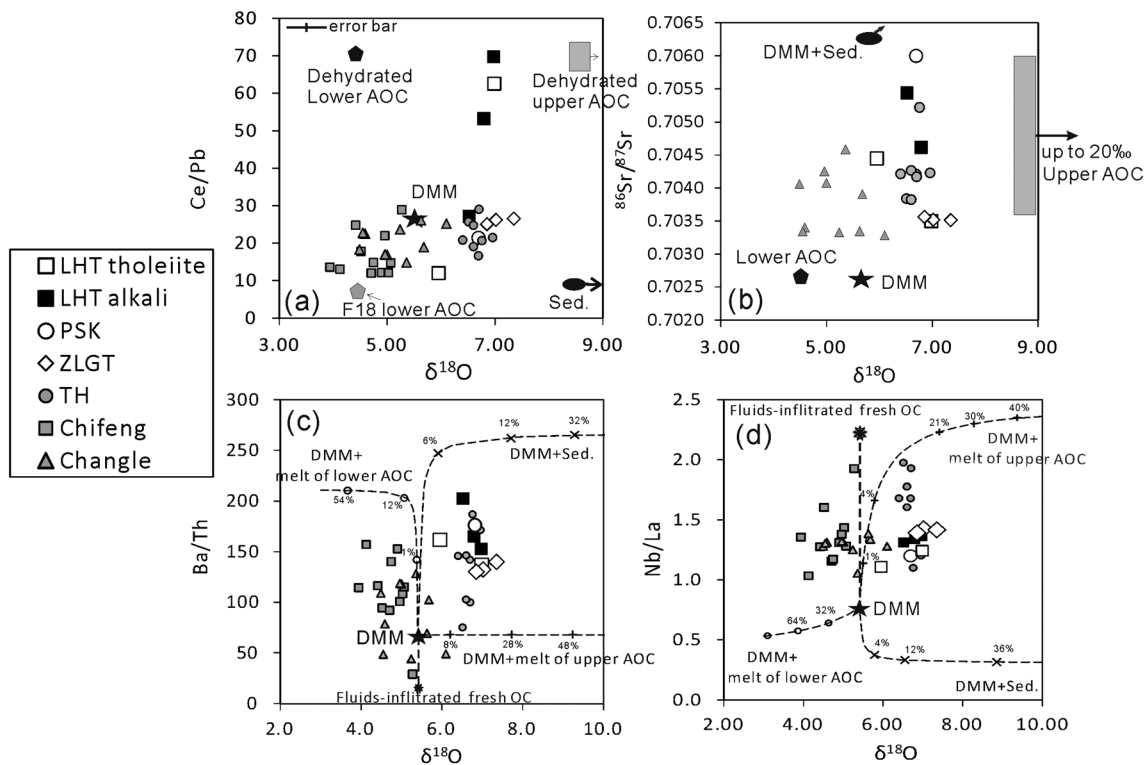


Figure 9. The comparison of oxygen isotopes of cpx with the (a, c, and d) bulk rock elemental ratios and (b) $^{87}\text{Sr}/^{86}\text{Sr}$. In Figure 9a, the Ce/Pb ratios of the dehydrated lower and upper ROC were calculated based on the mobility difference of these two elements [Kessel *et al.*, 2005], and F18 is the sample number in Bach *et al.*, 2001. In Figures 9c and 9d, the bold dashed lines show the modeled mixing among the depleted-MORB mantle (DMM) [Workman and Hart, 2005] and the marine sediments (Sed.) (GLOSS in Plank and Langmuir [1998]), partial melts of altered upper oceanic crust [Staudigel *et al.*, 1995], and altered lower oceanic crust (AOC) [Bach *et al.*, 2001]. The “fluids infiltrated fresh OC” means the oceanic crust effected by fluids but without of obvious change in oxygen isotopes [Bach *et al.*, 2001]. Note that the elemental loss in the subduction process have been considered based on the elemental mobility during the hydration process in the subduction zone are from Johnson and Plank [1999] and Kessel *et al.* [2005]. The composition of the partial melting of the subducted oceanic crust (eclogite) is calculated with the Excel sheet of Stracke *et al.* [2003]. The data for the Changle (Suwan-Shandong region) and Chifeng (northern margin of NCC) basalts are calculated from the $\delta^{18}\text{O}_{\text{ol}}$ by Y. G. Xu *et al.* [2012] and Wang *et al.* [2015], respectively, based on the equilibrium fractionation $\Delta^{18}\text{O}_{\text{cpx-ol}} = 0.4\text{‰}$ [Mattey *et al.*, 1994]. Taihang basalt data are from Liu *et al.* [2015a].

Overall, the oxygen isotope data for the basalts from 106 Ma to ~0 Ma do not support a monotonous change of the recycled components from upper ROC to lower ROC in the mantle sources [Kuang *et al.*, 2012; Y. G. Xu *et al.*, 2012]. Instead, they also indicate a dynamical contribution from different part of Pacific oceanic crust.

6. Implications

The changes in composition for mantle-derived mafic rocks have usually been used to infer the thickness evolution of the NCC’s lithospheric mantle [Xu, 2001; Yan *et al.*, 2003, 2005; Kuang *et al.*, 2012; Xu, 2014; Zhang *et al.*, 2003]. One critical step in this performance is the estimation of the depth where partial melting occurred, which usually relies on the peridotite-based experimental results showing that alkali basalts (Si deficient) would derive from a deeper source than tholeiitic basalts (>3 GPa versus 1.5–2.5 GPa) [Falloon *et al.*, 1988], or on the dependence of bulk FeO and Na₂O content on melting depth [Langmuir *et al.*, 1992]. However, the validity of these discriminate criteria is counteracted by (1) the widespread appearance of ROC components in the mantle sources of the mafic rocks from 106 Ma to the late Cenozoic, most likely in the form of pyroxenite and eclogite (metamorphosed from MORB, or the secondary pyroxenite) [this work; Zhang *et al.*, 2009; Kuang *et al.*, 2012; Z. Xu *et al.*, 2012; Y. G. Xu *et al.*, 2012; Zeng *et al.*, 2011; Xu, 2014; Liu *et al.*, 2015a, 2015b]; (2) the fact that the partial melting of these fusible components could produce silicon-excessive/deficient melts [e.g., Hirschmann *et al.*, 2003; Sobolev *et al.*, 2007]; and (3) the interaction between the pyroxenite-/eclogite-derived melt and the surrounding ambient mantle, which could produce residual melts with silicon-excessive or silicon-deficient components [Stracke and Bourdon, 2009; Mallik and Dasgupta, 2012, 2014; Lambart *et al.*, 2012].

In the “melting column” and the lithospheric lid effect model [Klein and Langmuir, 1987], the average melting depth of the upwelling asthenosphere (with ROC components) would be controlled by two factors at a given mantle temperature: the components of the mantle source and the thickness of the lithosphere. The mantle source with lower ROC is less fusible than the upper ROC and mainly fuses in a shallower mantle, because the lower ROC usually has higher MgO than the upper ROC and thus higher solidus temperature, and the intercept with mantle geotherm occurs at shallower depth [Pertermann and Hirschmann, 2003]. Following with model, Kuang *et al.* [2012] and Y. G. Xu *et al.* [2012] suggested that the secular increase in Eu/Eu* and a decrease in $^{87}\text{Sr}/^{86}\text{Sr}$ with time for the 90–40 Ma mafic rocks in the northern NCC and southern NEC (Northeast China) mirrored a shift in magma source from the upper to lower part of the ROC, which may finally reflect a continuous thinning of the lithospheric mantle from the Late Cretaceous to the early Cenozoic. However, as we have discussed in the last section, the $\delta^{18}\text{O}$ data in this study indicate that the recycled components in the mantle sources of basalts from 106 Ma to 60 Ma were dominantly upper ROC. In fact, the linkage between the Eu/Eu* variation of a basalt and the type of ROC in the source is not so direct. This can be best demonstrated by the observation that the recycled oceanic components in the mantle sources of the Cenozoic alkali basalts from Taihang mountain and West Shandong are upper and lower oceanic crust (their cpx $\delta^{18}\text{O}$ are higher and lower than that of the normal mantle, respectively [Liu *et al.*, 2015a, 2015b]), respectively, while their bulk rocks do not exhibit corresponding negative and positive Eu anomalies [Liu *et al.*, 2015a, 2015b]. Thus, the variation of Eu/Eu* and $^{87}\text{Sr}/^{86}\text{Sr}$ (and other geochemical indexes) could not be simply attributed to the shift from upper ROC to lower ROC components in the mantle sources and then used to suggest the thickness evolution of lithospheric mantle.

Zhang *et al.* [2009], Z. Xu *et al.* [2012], and Dai *et al.* [2016] have suggested that the ROC-derived components had been “frozen” in the “juvenile” lithospheric mantle since the Early Cretaceous and were responsible for the OIB-type basalts in the NCC from that time on. However, as discussed above, from 106 Ma to the late Cenozoic, the contribution of the ROC components into the upper mantle beneath the NCC was dynamic and alternating, which most likely came from the upwelling convective asthenosphere.

7. Conclusions

The $\delta^{18}\text{O}$ values of the cpx phenocrysts in the ZLGT basalts (106 Ma) and the LHT alkali-to-tholeiitic basalts (70–60 Ma) from the northern margin of the NCC and the PSK basanites (86–78 Ma) from the eastern NCC are all obviously higher than the normal mantle value. The data confirm that the recycled oceanic crust from the subducted Pacific oceanic slab were involved in the mantle sources of mafic rocks since 106 Ma. However, our data also clearly show the following:

1. The OIB-type mafic rocks in the NCC were most likely derived from the convective asthenosphere rather than from the lithospheric mantle.
2. From 106 Ma to 60 Ma, the recycled components in the northern NCC were dominantly upper ROC, and there was no visible temporal shift from upper ROC to lower ROC in the northern NCC.
3. In the southeast NCC, from the Late Cretaceous to the late Cenozoic, both upper and lower ROC were involved in the mantle sources, and these ROC components were incorporated into the source gradually and dynamically, rather than having resided in the lithospheric mantle since the Early Cretaceous.
4. The widespread involvement of ROC components in the mantle sources calls for more caution in the use of geochemical characteristics of basalts to infer the evolution of the lithosphere.

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