**Origin of low $\delta^{26}\text{Mg}$ basalts with EM-I component: Evidence
for interaction between enriched lithosphere and carbonated
asthenosphere**

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Abstract

This study presents stable Mg isotopic data for Cenozoic potassic basalts from Wudalianchi and Erkeshan in northeastern China to determine the interactions between upwelling carbonated asthenosphere and enriched lithospheric mantle. Although the Wudalianchi and Erkeshan basalts have variable MgO contents of 4.45 to 9.47 wt.%, they exhibit a homogeneous Mg isotopic composition with $\delta^{26}\text{Mg}$ values ranging from -0.57 to -0.46‰ and averaging $-0.51 \pm 0.06\text{‰}$ (2SD, $n = 18$). This Mg isotopic composition is lighter than that of the average mantle ($\delta^{26}\text{Mg} = -0.25 \pm 0.07\text{‰}$) but similar to late Cretaceous (< 110 Ma) and Cenozoic basalts from the North China Craton and the South China Block ($\delta^{26}\text{Mg} = -0.60$ to -0.35‰). The high $\text{CaO}/\text{Al}_2\text{O}_3$ and Ba/Rb, and low Hf/Hf* ratios of the Wudalianchi and Erkeshan basalts are typical characteristics of carbonatitic metasomatism, suggesting that the light Mg isotopic composition could derive from involvement of recycled sedimentary carbonates in the mantle source. The high Dy/Er ratios (2.55 to 2.75) and excess of ^{230}Th ($^{230}\text{Th}/^{238}\text{U} = 1.24$ to 1.33) suggest presence of garnet in a relatively deep mantle source. Additionally, the seismic tomographic observations show the existence of the stagnant Pacific slab in the mantle transition zone (410-660 km) under eastern China. This carbonated mantle source should be located in the asthenosphere. However, compared to other low $\delta^{26}\text{Mg}$ basalts from eastern China with MORB-like Sr-Nd-Pb isotopic compositions and OIB-like trace element features, the Wudalianchi and Erkeshan basalts exhibit EM-I Sr-Nd-Pb isotopic compositions combined with

high SiO₂, Ba, K, Pb and LREE contents, high Ba/Th and low Ce/Pb ratios. These geochemical features require a contribution from another mantle source, most likely an EM-I lithospheric mantle. Therefore, an asthenosphere-lithosphere interaction model is proposed **for** determine the origin of the Wudalianchi and Erkeshan basalts. The original melt was derived from partial melting of the carbonated asthenospheric mantle that was metasomatized by carbonate melt from the stagnant Pacific slab in the mantle transition zone. This ascending melt with a low $\delta^{26}\text{Mg}$ signature subsequently interacted with the EM-I lithospheric mantle. The interaction dissolved pyroxene and crystallized olivine, releasing LILE, REE, Sr and Pb from dissolved minerals into the melt. Simultaneously, Mg was inherited by newly formed olivine. Therefore, this interaction modified the trace elements and the Sr-Nd-Pb isotopic compositions of the melt toward EM-I geochemical features but preserved the light Mg **isotope signatures**.

Key words: magnesium isotopes; continental basalts; asthenosphere-lithosphere interaction; recycled carbonates; Pacific slab subduction.

1. Introduction

It is widely accepted that the heterogeneity of the mantle observed in the composition of mantle-derived magmas is largely attributable to subduction and recycling of continental or oceanic crust into the deep mantle (Cohen and O’Nions, 1982; White and Hofmann, 1982; Zindler and Hart, 1986). Many studies have focused on the interactions between recycled materials and depleted mantle to explain the origins of EM and HIMU components (e.g., Cohen and O’Nions, 1982; White and Hofmann, 1982; Menzies et al., 1983; Zindler and Hart, 1986; Kogiso et al., 1997; Jackson et al., 2007). However, little is known about the interactions between recycled materials and enriched mantle and the associated consequences, e.g., late recycled crustal material overprints mantle previously enriched by earlier recycling events of the crust.

Recently, the light Mg isotopic compositions ($\delta^{26}\text{Mg} = -0.60$ to -0.35‰) of the < 110 Ma basalts from the North China Craton (NCC) and South China Block (SCB) have been attributed to the interaction between mantle peridotites and isotopically light carbonate melts derived from the subducted Pacific slab (Yang et al., 2012; Huang et al., 2015). This relationship is supported by seismic tomographic observations that suggest the stagnant Pacific slab existed in the mantle transition zone under eastern China (Huang and Zhao, 2006; Zhao et al., 2011). If this explanation is correct, the Cenozoic basalts from Northeast (NE) China should also contain light Mg isotopic compositions. The basalts from NE China have EM-I

Sr-Nd-Pb isotopic features that are distinct from the NCC and SCB basalts, indicating the contribution of an enriched mantle source (Choi et al., 2006; Chu et al., 2013). Therefore, Mg isotopic analyses of the basalts from NE China will help to determine the **interaction** between recycled sedimentary carbonates and an enriched mantle.

Here, we investigate the Mg isotopic compositions of Cenozoic potassic basalts with EM-I geochemical features from Wudalianchi and Erkeshan in NE China. Our results show that these basalts have homogeneous and light Mg isotopic compositions, with $\delta^{26}\text{Mg}$ values varying from -0.57 to -0.46‰. We argue that such a light Mg isotopic composition can only be explained by the involvement of recycled sedimentary carbonates in the asthenosphere mantle source. The melt originated from the carbonated mantle **and** subsequently interacted with an EM-I lithospheric mantle. Thus, this study provides a specific example of carbonated asthenosphere interaction with enriched lithospheric mantle.

2. Geological Background and Sample Description

Cenozoic continental basalts are widespread in eastern China (Fig. 1). A considerable number of studies on these basalts have been conducted since the 1980s (e.g., Zhou and Armstrong, 1982; Peng et al., 1986; Zhou et al., 1988; Song et al., 1990; Zhi et al., 1990; Liu et al., 1994; Zou et al., 2000; Zhang et al., 2003, 2009; Xu et al., 2005; Tang et al., 2006; Yang and Li, 2008; Chen et al., 2009; Zeng et al., 2010, 2011; Wang et al., 2011; Yang et al., 2012; Huang et al., 2015). The Cenozoic basalts

in NE China, including Wudalianchi, Erkeshan, Keluo and Xiaogulihe volcanic fields, are located on the northern margin of the Songliao graben, encompassing an area of > 1,400 km² (Fig. 1). These basalts are highly potassic with EM-I geochemical features (Basu et al., 1991; Chen et al., 2007; Zhang et al., 1991, 1995; Zou et al., 2003; Kuritani et al., 2013; Sun et al., 2014, 2015), distinguishing them from other Cenozoic continental basalts in eastern China (Zhou and Armstrong, 1982; Song et al., 1990; Zhi et al., 1990; Basu et al., 1991; Fan and Hooper, 1991; Liu et al., 1994; Chen et al., 2007). They represent an enriched mantle endmember in eastern China (Basu et al., 1991; Zhang et al., 1991, 1995; Zou et al., 2003; Chen et al., 2007; Kuritani et al., 2013; Sun et al., 2014, 2015).

The samples studied here were collected from lava flows related to the eruptions (0.25 Ma to 1721 AD, Zhang et al., 1995) from the Wudalianchi volcanic area, and lava flows related to the Erkeshan eruption (0.56 Ma, Liu et al., 2001) from the Kedong volcanic area (Fig. 1). Their petrology, major and trace element compositions, Re-Os and Sr-Nd-Hf isotopic compositions have been reported in a previous study (Chu et al., 2013). Only a brief description is given below.

The Wudalianchi and Erkeshan basalts are characterized by high K₂O contents of 4.50% to 6.09%, with K₂O/Na₂O ratios of 1.09 to 1.67 (Chu et al., 2013). They have variable MgO (4.45 to 9.47 wt.%), SiO₂ (49.02 to 53.54 wt.%), Al₂O₃ (12.15 to 14.39 wt.%), CaO (5.09 to 7.44 wt.%), total FeO (8.19 to 9.64 wt.%) and TiO₂ (2.26 to 2.77 wt.%) contents. They have enriched LREEs and exhibit strong positive Rb, Ba, K and

Pb anomalies based on a spider diagram (Fig. 2). Accordingly, they display lower Ce/Pb (9.4 to 13.5) and higher Ba/Th (198.2 to 316.1) ratios than those of basalts from the NCC and SCB (Fig. 3a, b), reflecting input of sediment-derived fluid (Sakuyama et al., 2013). The high Nb/U ratios (38.9 to 54.2) of the Wudalianchi and Erkeshan basalts resemble those of basalts from the NCC and SCB (Fig. 3a), suggesting that their low Ce/Pb ratios are not caused by crustal contamination. The geochemistry of the Wudalianchi and Erkeshan basalts suggests contradictory sources. The high Dy/Er ratios (2.55 to 2.75) and significant excess of ^{230}Th ($^{230}\text{Th}/^{238}\text{U} = 1.24$ to 1.33) suggest the presence of garnet in a relatively deep mantle source (Zou et al., 2003; Chen et al., 2007). Based on thermodynamic calculations, the temperatures of the Wudalianchi magmas were estimated to have been $\sim 1250^\circ\text{C}$, higher than the projected maximum temperature of the lithospheric mantle beneath the Wudalianchi volcanic field (Kuritani et al., 2013). However, high SiO_2 contents (49.02 to 53.54 wt.%) of the Wudalianchi basalts require a shallow melting depth (Chen et al., 2007).

The EM-I Sr-Nd-Pb isotopic compositions of the Wudalianchi and Erkeshan basalts with high $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.7051 to 0.7057, low $^{143}\text{Nd}/^{144}\text{Nd}$ of 0.5123 to 0.5125 and unradiogenic Pb ($^{206}\text{Pb}/^{204}\text{Pb} = 16.7$ to 17.1) isotope compositions have been well documented (Basu et al., 1991; Zhang et al., 1991; Zou et al., 2003; Chu et al. 2013). However, their mantle source is still a subject of debate (Zhang et al., 1995, 1998, 2000; Zou et al., 2003; Choi et al., 2006; Chen et al., 2007; Kuritani et al., 2013; Chu et al., 2013). Several models exist that may explain the mantle source: (1)

subcontinental lithospheric mantle (SCLM) enriched by past metasomatism during the Proterozoic (Zhang et al., 1995, 1998, 2000), likely phlogopite-bearing garnet peridotites (Zou et al., 2003; Sun et al., 2015); (2) shallow asthenosphere containing fragments of delaminated SCLM due to the Sr–Nd–Pb–Hf isotopic characteristics resembling a DMM-EMI array (Choi et al., 2006); (3) mixed FOZO (Focal Zone; Hart et al., 1992) and LoMu (low U/Pb; Douglass et al., 1999) mantle sources (Chen et al., 2007); (4) mantle transition zone metasomatized by K-rich sediment-derived fluids approximately 1.5 Ga ago, due to the stagnation of an ancient slab (Kuritani et al., 2013) and (5) SCLM metasomatized by delaminated lower continental crust based on low Ce/Pb ratios and unradiogenic Pb isotope compositions (Chu et al., 2013).

3. Analytical Methods

Stable Mg isotopic compositions were determined at the State Key Laboratory of Geological Processes and Mineral Resources, China University of Geosciences (Beijing). Procedures for sample dissolution, column chemistry and instrumental analysis are similar to those reported in previous studies (Teng et al., 2007, 2010; Huang et al., 2015). A brief description is given below.

Sample dissolution and column chemistry were performed in an ultra-clean laboratory. Approximately 1–5 mg sample powders were dissolved in Savillex screw-top beakers with HF–HNO₃ (~3:1) mixed acids followed by HCl–HNO₃ (~3:1) mixed acids. Then, the samples were refluxed with concentrated HNO₃. Finally, all

dried residues were dissolved in 1 M HNO₃ prior to ion exchange column chemistry. Magnesium was separated by cation exchange chromatography with Bio-Rad 200-400 mesh AG50W-X8 resin following the established procedures (Teng et al., 2007, 2010; Yang et al., 2009). The prepared samples were eluted using 1 M HNO₃ from the column. The same column procedure was performed twice to obtain pure Mg solutions for mass spectrometry. The eluted solutions were evaporated to dryness and dissolved in 3% HNO₃ prior to instrumental analysis. The concentrations of cations (e.g., Na, Al, K, Ti and Fe) in the solutions were measured by ICP-MS, with cation/Mg (mass/mass) ratios of < 0.02, which is negligible for the Mg isotope analyses. The BHVO-2, BCR-2 and GSR-3 standard materials and a blank were prepared and analysed following the same procedure to evaluate the precision, accuracy and blank.

Magnesium isotopic compositions were measured by the sample-standard bracketing method to correct for instrumental mass bias and drift using a Thermo-Finnigan Neptune plus MC-ICP-MS. Solutions containing 400 ppb Mg were introduced into the plasma (~ 100 µL/min) via a standard H-skimmer cone and an ESI PFA MicroFlow nebulizer with a quartz Scott-type spray chamber. Stable Mg isotopes were analysed in low-resolution mode, with ²⁶Mg, ²⁵Mg and ²⁴Mg measured simultaneously in separate Faraday cups (H3, IC and L3). The duration of each sample run was approximately 6-7 minutes. Repeated analyses of synthetic solution, mineral and rock standards are indicative of a long-term reproducibility of $\leq \pm 0.10\%$

for $\delta^{26}\text{Mg}$ (2SD, Table 1). The Mg content of the blank was < 10 ng, which was negligible for the Mg isotope analyses. The results are reported in standard δ -notation relative to DSM3, where $\delta^{25,26}\text{Mg} = [({}^{25,26}\text{Mg}/{}^{24}\text{Mg})_{\text{sample}}/({}^{25,26}\text{Mg}/{}^{24}\text{Mg})_{\text{DSM3}} - 1] \times 1000$ (Galy et al., 2003). The standard materials were also analysed, with $\delta^{26}\text{Mg} = -0.20 \pm 0.09\text{‰}$ for BCR-2, $\delta^{26}\text{Mg} = -0.27 \pm 0.01\text{‰}$ for BHVO-2 and $\delta^{26}\text{Mg} = -0.48 \pm 0.03\text{‰}$ for GSR-3. These values are consistent with previously published data (-0.36 to -0.16‰ for BCR-2 and -0.32 to -0.16‰ for BHVO-2; Teng et al., 2007, 2015; Huang et al., 2009, 2011, 2015; An et al., 2014).

4. Results

Stable Mg isotopic compositions of the Wudalianchi and Erkeshan basalts and standard reference materials are listed in Table 1. The $\Delta^{25}\text{Mg}'$ ($\Delta^{25}\text{Mg}' = \delta^{25}\text{Mg}' - 0.521 \times \delta^{26}\text{Mg}'$, where $\delta^{25,26}\text{Mg}' = 1000 \times \ln[(\delta^{25,26}\text{Mg} + 1000)/1000]$; Young and Galy, 2004) values for the Wudalianchi and Erkeshan basalts are within $0 \pm 0.02\text{‰}$ (Table 1).

Overall, the Wudalianchi and Erkeshan basalts have a homogeneous Mg isotopic composition (Fig. 4a and Table 1), with $\delta^{25}\text{Mg}$ values ranging from -0.29 to -0.23‰ and $\delta^{26}\text{Mg}$ values ranging from -0.57 to -0.46‰ . These Mg isotopic compositions are similar to those of < 110 Ma basalts from NCC ($\delta^{26}\text{Mg} = -0.60$ to -0.42‰ ; Yang et al., 2012) and Cenozoic basalts from SCB ($\delta^{26}\text{Mg} = -0.60$ to -0.35‰ ; Huang et al., 2015), but lighter than that of average mantle ($\delta^{26}\text{Mg} = -0.25 \pm 0.07\text{‰}$; Teng et al., 2010)

and > 120 Ma basaltic rocks from NCC ($\delta^{26}\text{Mg} = -0.27 \pm 0.05\text{‰}$, $n = 5$, 2SD; Yang et al., 2012) (Fig. 4a).

The $\delta^{26}\text{Mg}$ values of the Wudalianchi and Erkeshan basalts are invariant with MgO and TiO_2 contents, in accordance with those of the low $\delta^{26}\text{Mg}$ basalts from the NCC and SCB (Fig. 4). They have low Hf/Hf^* (0.62 to 1.07; $\text{Hf}/\text{Hf}^* = \text{Hf}_\text{N}/(\text{Sm}_\text{N} \times \text{Nd}_\text{N})^{0.5}$), high Ba/Rb (14.4 to 19.9) and $\text{CaO}/\text{Al}_2\text{O}_3$ (0.36 to 0.57) ratios compared to MORB (Hofmann, 1988), but similar to the low $\delta^{26}\text{Mg}$ basalts from the NCC and SCB (Fig. 5).

5. Discussion

5.1 Characteristics of the primary melt

The chemical index of alteration (CIA) values of the Wudalianchi and Erkeshan basalts vary from 41.0 to 45.0 (Table 1), which fall within the fresh basalt range of 30 to 45 (Nesbitt and Young, 1982). These basalts exhibit a relatively large $\text{Mg\#}$ variation of 55.3 to 70.6, which may reflect compositional evolution from the primary melts (Langmuir et al., 1977). To discuss their source characteristics, the geochemical features of the primary melt should be first^{ly} evaluated.

The Wudalianchi and Erkeshan basalts show positive correlations between $\text{Mg\#}$ and both TFeO (Fig. 6a) and CaO (Fig. 6b), indicating olivine and pyroxene fractional crystallization. Accordingly, $\text{CaO}/\text{Al}_2\text{O}_3$ ratios of these basalts display a large variation and show a positive correlation with $\text{Mg\#}$ (Fig. 6c). This correlation could

be attributed to pyroxene fractional crystallization rather than plagioclase crystallization, because no obvious Eu anomaly is observed in these basalts (Fig. 2). In addition, the negative correlation between TiO_2 and Mg# (Fig. 6d) with no negative Ti anomaly (Fig. 2) indicates limited ilmenite fractional crystallization. The variations of major oxides (e.g., MgO, K_2O and CaO) can be modelled by fractional crystallization processes using MELTS software (Fig. 6a-d; Gualda and Ghiorso, 2015). The results indicate fractional crystallization of olivine, orthopyroxene and clinopyroxene (Fig. 6a-d). The major elements of the primary melt associated with the Wudalianchi and Erkeshan basalts are estimated from the samples with the highest MgO/ Al_2O_3 ratios (YQ4 and HSS6).

Although the Wudalianchi and Erkeshan basalts have undergone significant olivine and pyroxene fractional crystallization, their Ce/Pb, Nb/U, Hf/Hf* and Ba/Rb ratios and Sr-Nd isotopes do not vary with Mg# values (Fig. 6e-j), indicating limited influence on these values during fractional crystallization. The trace element ratios (e.g., Ce/Pb, Nb/U, Hf/Hf* and Ba/Rb) and Sr-Nd-Pb isotopic compositions of the primary melt are denoted with the variations (see Table 2 for summarized data). For comparison, data for the low $\delta^{26}\text{Mg}$ basalts from the NCC and SCB are also listed in Table 2.

5.2 Origin of the light Mg isotopic composition

The $\delta^{26}\text{Mg}$ values of the Wudalianchi and Erkeshan basalts do not vary with

MgO contents (Fig. 4a), indicating limited Mg isotope fractionation during magma differentiation. This observation is consistent with previous studies of Mg isotope fractionation during magma differentiation (Teng et al., 2007, 2010; Liu et al., 2010) and of the low $\delta^{26}\text{Mg}$ basalts from the NCC and SCB (Fig. 4a; Yang et al., 2012; Huang et al., 2015). In addition, the high Nb/U (38.9 to 54.2) ratios of the Wudalianchi and Erkeshan basalts suggest limited input of crustal materials. Therefore, the light Mg isotopic composition of the Wudalianchi and Erkeshan basalts is inherited from the mantle source, which may result from two processes: accumulation of ilmenite and/or involvement of carbonates.

A recent study on lunar high-Ti basalts indicates that ilmenite could have low $\delta^{26}\text{Mg}$ values ($< -0.61 \pm 0.03\text{‰}$), and the accumulation of ilmenite may hence result in light Mg isotopic compositions (Sedaghatpour et al., 2013). However, this origin of low $\delta^{26}\text{Mg}$ basalts can be ruled out for the Wudalianchi and Erkeshan lavas. First, the Wudalianchi and Erkeshan basalts have lower TiO_2 (2.3 to 2.8 wt.%) contents than the lunar high-Ti ($\text{TiO}_2 > 6.0$ wt.%) basalts. Second, the trace elements Nb and Ta are compatible in ilmenite and the partition coefficients $D_{\text{Ta}}/D_{\text{Nb}}$ are approximately ~ 1.3 between ilmenite and mafic melts (Dygert et al., 2013). Accumulation of ilmenite in the mantle source should generate a negative correlation between Nb/Ta ratios and TiO_2 (wt.%). However, the Wudalianchi and Erkeshan basalts display a positive correlation between Nb/Ta and TiO_2 (wt.%) (Fig. 7). Third, unlike the lunar high-Ti basalts, no correlation between $\delta^{26}\text{Mg}$ and TiO_2 is observed for the Wudalianchi and

Erkeshan basalts (Fig. 4b). Thus, the accumulation of isotopically light ilmenite is unlikely to have affected the mantle source of the Wudalianchi and Erkeshan basalts.

The light Mg isotopic compositions ($\delta^{26}\text{Mg} = -0.60$ to -0.35‰) of the < 110 Ma basalts from the North China Craton (NCC) and South China Block (SCB) have been attributed to the interaction between mantle peridotites and isotopically light carbonate melts derived from the subducted Pacific slab (Yang et al., 2012; Huang et al., 2015). The high $\text{CaO}/\text{Al}_2\text{O}_3$ (0.57) and Ba/Rb (14.4 to 19.9), and low Hf/Hf^* ratios (0.63 to 1.07) of the Wudalianchi and Erkeshan basalts (Fig. 5) are similar to those of the low $\delta^{26}\text{Mg}$ basalts from the NCC and SCB (Table 2). These are typical characteristics of carbonatitic metasomatism because carbonatite is enriched in Ba, Th and La and depleted in Zr, Hf and Ti (Hoernle et al., 2002; Bizimis et al., 2003). A binary mixing calculation based on N-MORB and carbonate melts indicates that the involvement of $\sim 10\%$ carbonate melts can form the light Mg isotopic composition in the basalts from eastern China (Huang and Xiao, 2016). Therefore, the light Mg isotopic composition of the Wudalianchi and Erkeshan basalts could also derive from a mantle source involving sedimentary carbonates (Yang et al., 2012; Huang et al., 2015).

The distribution of low $\delta^{26}\text{Mg}$ basalts defines a large area with a light Mg isotope anomaly in the mantle beneath eastern China (Fig. 1), which coincides with the subducted Pacific slab in the mantle transition zone, as revealed by seismic tomography (Fukao et al., 1992; Huang and Zhao, 2006; Zhao et al., 2011). Based on

experimental studies, the subducted carbonated oceanic crust underwent slab melting to produce carbonatitic melts in the mantle transition zone (410-660 km) because the majority of slabs will intersected the melting curve of carbonated oceanic crust at depths of approximately 300-700 km (Dasgupta and Hirshmann, 2010; Thomson et al., 2016). Therefore, the low $\delta^{26}\text{Mg}$ mantle source was formed by interaction between the mantle and the carbonate melt from the subducted oceanic crust above the mantle transition zone (Fig. 8). The carbonated asthenospheric mantle could partially melt at a depth of $\sim 300\text{km}$ (Dasgupta and Hirschmann, 2010) to generate the Wudalianchi and Erkeshan basalts, which is supported by their high Dy/Er ratios (2.55 to 2.75) and significant excess of ^{230}Th ($^{230}\text{Th}/^{238}\text{U} = 1.24$ to 1.33) (Zou et al., 2003; Chen et al., 2007).

5.3 The contribution of an EM-I mantle source

Although the origin of the Wudalianchi and Erkeshan basalts is still a subject of debate, many studies suggested contributions of at least two components with different geochemical features (Zhang et al., 1995, 1998, 2000; Zou et al., 2003; Choi et al., 2006; Chen et al., 2007; Kuritani et al., 2013; Chu et al., 2013). The debate focuses on the characteristics of these two components and their origins.

As discussed above, one mantle source was the carbonated asthenospheric mantle, which is similar to that inferred from the NCC and SCB basalts. However, the Wudalianchi and Erkeshan basalts exhibit many distinct geochemical features from

those of the NCC and SCB basalts (Table 2): e.g., high SiO₂ (49.4 wt.%) and K₂O (4.5 wt.%), low MgO (8.8 wt.%) and TFeO (9.4 wt.%) contents, high Ba/Th (198.2 to 316.1) and low Ce/Pb (9.4 to 13.5) ratios, and EM-I Sr-Nd-Pb isotopic compositions.

Thus, the second mantle source should have EM-I signatures.

Several sources for the EM-I signatures were previously proposed: the mantle transition zone metasomatized by K-rich sediment-derived fluids ~1.5 Ga ago (Kuritani et al., 2013), or the subcontinental lithospheric mantle (SCLM) metasomatized by delaminated lower continental crust (Chu et al., 2013) or by sediment-derived fluid during the Proterozoic (Zhang et al., 1995, 1998, 2000; Zou et al., 2003; Sun et al., 2014, 2015).

Based on the high temperature of the Wudalianchi magmas (~ 1250 °C), Kuritani et al. (2013) proposed that the EM-I signatures of the Wudalianchi basalts may be derived from the mantle transition zone. However, the following evidence argues against this view. First, the continental basalts from NCC and SCB, which were also derived from the mantle transition zone (Yang et al., 2012; Huang et al., 2015), do not show EM-I signatures. This indicates that the mantle transition zone may not exhibit EM-I signatures. In addition, K-hollandite is required in the model of Kuritani et al. (2013) to explain the high K, Rb, Sr and Ba contents in the Wudalianchi basalts. However, this mineral is highly depleted in Nb, which will result in extremely low Nb/U ratios. This is not consistent with the high Nb/U (38.9 to 54.2) ratios observed in the Wudalianchi and Erkeshan basalts.

Chu et al. (2013) suggested the EM-I signatures originated from a subcontinental lithospheric mantle contaminated by delaminated lower continental crust because the low Ce/Pb ratios and unradiogenic Pb isotope compositions are typical features of the lower continental crust. However, this process decreases not only Ce/Pb, but also Nb/U ratios, which is not supported by the high Nb/U (38.9 to 54.2) ratios of the Wudalianchi and Erkeshan basalts.

Alternatively, the EM-I signatures are most likely derived from a SCLM metasomatized by sediment-derived fluid during the Proterozoic (Zhang et al., 1995, 1998, 2000; Zou et al., 2003). The enrichments of Rb, Ba and Pb and high Ba/Th ratios are typical characteristics of sediment-derived fluid inputs (Sakuyama et al., 2013), which must have occurred at least ~1.5 Ga ago to produce such low $^{206}\text{Pb}/^{204}\text{Pb}$ ratios (16.7 to 17.1) (Basu et al., 1991; Zhang et al., 1991; Zou et al., 2003; Kuritani et al., 2013).

5.4 Interaction between carbonated asthenosphere and enriched lithosphere

Both carbonated asthenosphere and enriched lithosphere contributed to the Wudalianchi and Erkeshan basalts. Two stages are, therefore, required: partial melting of the carbonated peridotite to produce low $\delta^{26}\text{Mg}$ basaltic melt (Stage 1), and interaction of the basaltic melt with the enriched lithospheric mantle to obtain EM-I Sr-Nd-Pb isotopic compositions (Stage 2) (Fig. 9).

However, why did interaction with the EM-I SCLM significantly modify the

trace **element** and Sr-Nd-Pb isotopic **composition** of the melt, but have little effect on the Mg isotopes? Two possible **mechanisms** for **interaction** between melt derived from **the** asthenosphere and the EM-I SCLM should be considered.

(1) Mixing between melts derived from the asthenosphere and the EM-I SCLM. Given the limited fractionation of Mg isotopes during partial melting of the mantle (Teng et al., 2007, 2010), the basaltic melt from the EM-I SCLM should exhibit a mantle-like Mg isotopic composition. Thus, when the low $\delta^{26}\text{Mg}$ melt derived from the carbonated asthenosphere mixed with the melt from the EM-I SCLM, **the** Sr-Nd-Pb and Mg isotopic compositions were synchronously modified based on **the relevant** mixing proportions. However, although the Wudalianchi and Erkeshan basalts have typical EM-I Sr-Nd-Pb isotopic features, they still preserve light Mg isotopic **compositions**, similar to those of basalts from the NCC and SCB (Yang et al., 2012; Huang et al., 2015). **This suggests** that the interaction was **more complex than** simple melt mixing.

(2) A reaction between the low SiO_2 basaltic melt derived from the asthenosphere and the EM-I SCLM. Partial melting of carbonated peridotite produces low SiO_2 alkaline melts (e.g., nephelite, basanite, alkaline basalt) (Dasgupta et al., 2007). The primary melt of the Wudalianchi and Erkeshan basalts exhibits higher SiO_2 (49.4 wt.%) and K_2O (4.5 wt.%), **and** lower MgO (8.8 wt.%) and TFeO (9.4 wt.%) contents than those of low $\delta^{26}\text{Mg}$ basalts from the NCC and SCB (Table 2). **This indicates** that the subsequent interaction elevated SiO_2 and K_2O , and decreased MgO and TFeO

concentrations of the melt. This inference is consistent with the interaction between low SiO₂ melt and peridotite, which converts pyroxene to olivine, as reported in previous studies (Kelemen et al., 1992; Edwards and Malpas, 1996; Zhou et al., 1996, 2014; Suhr et al., 1998):

Such an interaction can explain all petrological and geochemical features of the Wudalianchi and Erkeshan basalts. First, the melt was derived from a deep source (carbonated asthenosphere) with garnet present, which is consistent with the high Dy/Er ratios (2.55 to 2.75), significant excess ²³⁰Th (²³⁰Th/²³⁸U = 1.24 to 1.33) (Zou et al., 2003; Chen et al., 2007), and the high temperature of the melt (~1250 °C; Kuritani et al., 2013). In addition, the high SiO₂ contents, previously considered to require a shallow melting depth (Chen et al., 2007), resulted from interaction with the SCLM. Moreover, the trace elements of the EM-I SCLM largely entered the melt during the interaction because the solid product was olivine. Consequently, the Ce/Pb and Ba/Th ratios were significantly modified, and the Sr–Nd–Pb–Hf isotopic characteristics resemble a DMM-EMI array (Choi et al., 2006). Finally, all Mg was transferred from Opx and Cpx into the newly formed olivine during the interaction. Thus, the Mg isotopic composition of the melt was not significantly affected.

Therefore, the following model is proposed (Fig. 8): The subduction of the western Pacific plate subduction started in the Jurassic (Maruyama et al., 1997; Wu et al., 2007), whereby the slab may have reached the bottom of the transition zone and

become stagnant slab in the Cretaceous (Niu, 2005). The convective upper mantle was hybridized by a partial melt from the recycled carbonates of the stagnant oceanic slab, forming carbonated peridotites above the mantle transition zone. Due to convection in the big mantle wedge that were proposed for the active tectonic and volcanic processes in northeast Asia based on multiscale tomographic images (Zhao et al., 2009), diapirs of carbonated peridotite ascended. When they reached the solidus of carbonated peridotite at a depth of approximately ~ 300 km (Dasgupta and Hirschmann, 2010), partial melting generated a low $\delta^{26}\text{Mg}$ melt with high Ce/Pb (~ 25) and Nb/U (~ 50), low Ba/Th (~ 53) and MORB-like Sr-Nd-Pb isotopic compositions. Because there is no EM-I lithospheric mantle beneath NCC and SCB, the geochemical features of this melt did not change in these regions during interaction between the asthenosphere-derived melt and lithosphere, producing the low $\delta^{26}\text{Mg}$ basalts with MORB-like Sr-Nd-Pb isotopic compositions of the NCC and SCB. In NE China, however, the melt interacted with the EM-I SCLM, forming the Wudalianchi and Erkeshan basalts with low $\delta^{26}\text{Mg}$ and EM-I Sr-Nd-Pb isotopic compositions (Fig. 8).

6. Conclusions

The stable Mg isotopic compositions of the Wudalianchi and Erkeshan basalts from Northeast China were determined at high precision. A number of conclusions for the origin of the basalts follow from the new data.

(1) The Wudalianchi and Erkeshan basalts have homogeneous and light Mg isotopic compositions, with $\delta^{26}\text{Mg}$ ranging from -0.57 to -0.46‰ and averaging -0.51 $\pm$ 0.06‰ (2SD, n = 18). These values are similar to those of low $\delta^{26}\text{Mg}$ basalts from the NCC and SCB. Such a light Mg isotopic composition reflects the involvement of recycled sedimentary carbonates in the mantle source, which is also supported by high CaO/Al₂O₃ and Ba/Rb, and low Hf/Hf* ratios. The recycled sedimentary carbonates likely originate from the stagnant Pacific slab beneath eastern China, which is consistent with the seismic tomography observations.

(2) Compared to other low $\delta^{26}\text{Mg}$ basalts from eastern China with MORB-like Sr-Nd-Pb isotopic compositions and OIB-like trace element features, the Wudalianchi and Erkeshan basalts exhibit EM-I Sr-Nd-Pb isotope signatures, high SiO₂, Ba, K, Pb, LREE contents, as well as high Ba/Th and low Ce/Pb ratios. These geochemical features require contributions from another mantle source, likely an EM-I lithospheric mantle.

(3) An asthenosphere-lithosphere interaction model is proposed for the origin of the Wudalianchi and Erkeshan basalts, which most likely originated from partial melting of carbonated asthenospheric mantle. This material subsequently interacted with an EM-I lithospheric mantle during melt ascent. This interaction dissolved pyroxenes and crystallized olivine, significantly modifying the trace element and Sr-Nd-Pb isotopic compositions of the melt, but with little effect on Mg isotopes.

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Figure Captions

Fig. 1 Simplified geological map of eastern China showing samples locations (modified from Yang et al., 2009). Wudalianchi and Erkeshan are denoted by red circles. The yellow triangles represent the locations of the previously reported low $\delta^{26}\text{Mg}$ basalts from the North China Craton and the South China Block (Yang et al., 2012; Huang et al., 2015). The dark grey area is the distribution of the low $\delta^{26}\text{Mg}$ basalts, indicating a large-scale light Mg isotope anomaly in the mantle beneath eastern China.

Fig. 2 Primitive mantle-normalized trace element patterns for the Wudalianchi and Erkeshan basalts and the low $\delta^{26}\text{Mg}$ basalts from NCC and SCB. The Wudalianchi and Erkeshan basalts show strong positive Rb, Ba, K and Pb anomalies compared to the trace element patterns of the NCC (North China Craton) and SCB (South China Block) basalts. Primitive mantle normalized values, MORB (mid-ocean ridge basalts) and OIB (ocean island basalts) data are from Sun and McDonough (1989). The NCC and SCB basalt data are from Tang et al. (2006) and Huang et al. (2015), respectively.

Fig. 3 (a) Nb/U and (b) Ba/Th vs. Ce/Pb for the Wudalianchi and Erkeshan basalts and the low $\delta^{26}\text{Mg}$ basalts from NCC and SCB. The arrow represents metasomatism by fluids from sediments, which elevates Ba/Th and decreases Ce/Pb. Database: MORB and HIMU from Sun and McDonough (1989) and Weaver et al. (1991), lower continental crust (LCC) from Rudnick and Gao (2003), EM-I from

Leroex et al. (1990) and Weaver et al. (1991). The Wudalianchi and Erkeshan basalts are from Chu et al. (2013). The NCC and SCB basalts are from Zhang et al. (2003), Tang et al. (2006) and Huang et al. (2015).

Fig. 4 $\delta^{26}\text{Mg}$ vs. (a) MgO (wt.%) and (b) TiO_2 (wt.%) for the Wudalianchi and Erkeshan basalts and the low $\delta^{26}\text{Mg}$ basalts from NCC and SCB. The arrow represents the trend for accumulation of ilmenite. Database: the NCC basalts from Yang et al. (2012), the SCB basalts from Huang et al. (2015), the Wudalianchi and Erkeshan basalts from Table 1 and Chu et al. (2013). The grey bar represents the average mantle ($\delta^{26}\text{Mg} = -0.25 \pm 0.07\text{‰}$; Teng et al., 2010). Error bars represent 2SD uncertainties.

Fig. 5 (a) Hf/Hf^* , (b) Ba/Rb and (c) $\text{CaO}/\text{Al}_2\text{O}_3$ vs. $\delta^{26}\text{Mg}$ for the Wudalianchi and Erkeshan basalts with the low $\delta^{26}\text{Mg}$ basalts from NCC and SCB. The pink star represents the average ratios of $\text{CaO}/\text{Al}_2\text{O}_3$ (152.4), Hf/Hf^* (0.01) and Ba/Rb (140.5) for magnesio- and calico-carbonatites (Hoernle et al., 2002; Bizimis et al., 2003). The arrows represent the involvement of recycled sedimentary carbonate in the mantle source. The NCC and SCB basalt data are from Yang et al. (2012) and Huang et al. (2015), respectively. MORB data are from Hofmann (1988). The $\delta^{26}\text{Mg}$ values of MORB and carbonatite are from Teng et al. (2010) and Wang et al. (2014), respectively. Error bar represents 2SD uncertainties. $\text{Hf}/\text{Hf}^* = \text{Hf}_\text{N}/(\text{Sm}_\text{N} \times \text{Nd}_\text{N})^{0.5}$, where subscript N means normalized to chondrites (Sun and McDonough, 1989).

Fig. 6 (a) TFeO (wt.%), (b) CaO (wt.%), (c) $\text{CaO}/\text{Al}_2\text{O}_3$, (d) TiO_2 (wt.%), (e)

$\epsilon_{\text{Nd(t)}}$, (f) $^{87}\text{Sr}/^{86}\text{Sr}$, (g) Ce/Pb, (h) Nb/U, (i) Hf/Hf* and (j) Ba/Rb vs. Mg# for the Wudalianchi and Erkeshan basalts. The red curves represent the fractional crystallization (olivine and/or pyroxene) processes modelled using MELTS software. The parental magma compositions of SiO_2 (45.90 wt.%), TiO_2 (1.65 wt.%), Al_2O_3 (10.10 wt.%), Fe_2O_3 (3.30 wt.%), FeO (7.05 wt.%), MgO (18.50 wt.%), CaO (5.60 wt.%), Na_2O (3.00 wt.%), K_2O (3.70 wt.%) and P_2O_5 (1.08 wt.%) is based on a large number of simulations. The given conditions are ~15 kbar and $\log f_{\text{O}_2} = \text{NNO} - 2.0$ (NNO corresponding to Ni-NiO buffer). Red circles on the fractional crystallization curves indicate 10 °C temperature decrements. Major oxides, trace elements and Sr-Nd isotopic data of the Wudalianchi and Erkeshan basalts are from Chu et al. (2013).

Fig. 7 Nb/Ta vs. TiO_2 (wt.%) for the Wudalianchi and Erkeshan basalts. The accumulation of isotopically light ilmenite should generate a negative correlation between Nb/Ta and TiO_2 (wt.%), which is not observed for the Wudalianchi and Erkeshan basalts.

Fig. 8 Cartoon showing the origin of the Wudalianchi and Erkeshan basalts. The thickness of the stagnant slab is based on Zou et al. (2008), Kameyama and Nishioka, (2012) and Li et al. (2013). The original melt derived from partial melting of the carbonated asthenospheric mantle, interacted with the EM-I lithospheric mantle during melt ascent. This interaction dissolved pyroxenes and crystallized olivine, significantly modifying the trace element and Sr-Nd-Pb isotopic composition of the

melt towards EM-I geochemical signatures. However, this interaction had little effect on Mg isotopes.

Fig. 9 $\delta^{26}\text{Mg}$ vs. $\epsilon_{\text{Nd}}(\text{t})$ for the Wudalianchi and Erkeshan basalts and the low $\delta^{26}\text{Mg}$ basalts from NCC. Stage 1: involvement of recycled sedimentary carbonate in the mantle source to produce low $\delta^{26}\text{Mg}$ basaltic melts. Stage 2: interaction of the melt with enriched lithospheric mantle to obtain melts with EM-I Sr-Nd-Pb isotopic compositions. The red curve represents binary mixing between depleted MORB (mid-ocean ridge basalts) and the carbonates, and circles on the binary modelling curves indicate 2% increments. Mg and Nd isotopic data for the NCC basalts are from Zhang et al. (2003), Tang et al. (2006) and Yang et al. (2012). Parameters for MORB ($\delta^{26}\text{Mg}$ of -0.25‰, MgO of 11.2 wt.%, $\epsilon_{\text{Nd}}(\text{t})$ of +8 and Nd of 7.3 ppm) are from Langmuir et al. (1977), Zindler and Hart (1986), Sun and McDonough (1989) and Teng et al. (2010), respectively; Carbonatite values ($\delta^{26}\text{Mg}$ of -2.508‰, MgO of 12.6 wt.%, $\epsilon_{\text{Nd}}(\text{t})$ of +4 and Nd of 500 ppm) are from Wang et al. (2014), Hoernle et al. (2002) and Bizimis et al. (2003), respectively. Error bar represents 2SD uncertainties.

Tables

Table 1 Mg isotopic compositions of volcanic rocks from Wudalianchi and Erkeshan, Northeast China

Sample ^a	Location ^a	MgO ^a	TiO ₂ ^a	CIA ^a	$\delta^{26}\text{Mg}^b$	2SD ^c	$\delta^{25}\text{Mg}^b$	2SD ^c	$\Delta^{25}\text{Mg}^d$
YQ1	Wudalianchi	6.17	2.45	44.4	-0.52	0.08	-0.27	0.04	0.00
YQ3	Wudalianchi	6.18	2.45	44.5	-0.49	0.05	-0.24	0.02	0.02
YQ4	Wudalianchi	9.47	2.39	41.0	-0.48	0.04	-0.24	0.06	0.01
YQ6	Wudalianchi	4.45	2.72	43.8	-0.51	0.03	-0.27	0.04	0.00
YQ7	Wudalianchi	7.40	2.35	41.9	-0.54	0.08	-0.28	0.07	0.01
YQ8	Wudalianchi	7.69	2.34	41.4	-0.48	0.10	-0.25	0.05	0.00
LHS1	Wudalianchi	7.45	2.30	43.7	-0.49	0.06	-0.26	0.04	-0.01
LHS3	Wudalianchi	7.46	2.29	43.8	-0.52	0.05	-0.25	0.03	0.02
LHS4	Wudalianchi	6.21	2.77	44.1	-0.52	0.05	-0.26	0.03	0.01
LHS6	Wudalianchi	5.96	2.34	44.9	-0.47	0.10	-0.24	0.06	0.00
HSS2	Wudalianchi	5.93	2.42	44.8	-0.49	0.03	-0.26	0.05	0.00
HSS5	Wudalianchi	5.99	2.42	45.0	-0.46	0.07	-0.26	0.08	-0.02
HSS6	Wudalianchi	8.10	2.26	41.0	-0.48	0.07	-0.25	0.04	0.00
HSS8	Wudalianchi	6.01	2.32	44.9	-0.48	0.07	-0.23	0.05	0.02
KD1	Erkeshan	6.87	2.57	44.5	-0.55	0.08	-0.28	0.05	0.01
KD3	Erkeshan	6.88	2.60	44.4	-0.57	0.10	-0.28	0.05	0.02
KD4	Erkeshan	6.83	2.61	44.5	-0.55	0.10	-0.29	0.05	0.00
KD5	Erkeshan	6.90	2.59	44.6	-0.54	0.10	-0.27	0.04	0.01
Standards									
BCR-2					-0.20	0.09	-0.12	0.06	-0.02
BHVO-2					-0.27	0.01	-0.12	0.03	0.03
GSR-3					-0.48	0.03	-0.24	0.02	0.01

^a The samples are from Wudalianchi and Erkeshan (Chu et al., 2013). CIA = Chemical Indexes of Alteration, defined as = molecular ratios of $\text{Al}_2\text{O}_3/(\text{Al}_2\text{O}_3 + \text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O}) \times 100$ (Nesbitt and Young, 1982).

^b $\delta^{25,26}\text{Mg} = \{(^{25,26}\text{Mg}/^{24}\text{Mg})_{\text{sample}}/(^{25,26}\text{Mg}/^{24}\text{Mg})_{\text{DSM3}} - 1\} \times 1000$, DSM3 is a Mg solution made from pure Mg metal (Galy et al., 2003).

^c 2SD = two times the standard deviation of the population of 4 repeat measurements of a sample solution.

^d $\Delta^{25}\text{Mg}' = \delta^{25}\text{Mg}' - 0.521 \times \delta^{26}\text{Mg}'$, where $\delta^{25,26}\text{Mg}' = 1000 \times \ln[(\delta^{25,26}\text{Mg} + 1000)/1000]$ (Young and Galy, 2004).

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Table 2 Composition of primary melt of the Wudalianchi and Erkeshan basalts, and the NCC and SCB basalts

	Wudalianchi & Erkeshan ^a		NCC ^a		SCB ^a	
	average ^b	1SD ^c	average ^b	1SD ^c	average ^b	1SD ^c
SiO ₂	49.4	0.5	44.2	0.9	41.9	1.2
MgO	8.8	1.0	9.9	0.1	11.1	1.3
CaO	7.2	0.4	9.0	1.3	9.7	0.3
Al ₂ O ₃	12.6	0.6	14.7	0.4	11.5	0.2
TiO ₂	2.3	0.1	2.5	0.2	2.3	0.2
TFeO	9.4	0.3	12.3	0.6	12.8	0.8
Na ₂ O	4.0	0.3	3.1	0.1	4.3	0.4
K ₂ O	4.5	0.1	1.2	0.5	1.9	0.6
CaO/Al ₂ O ₃	0.57		0.61		0.84	
Ba/Rb	14.4 - 19.9		9.5 - 67.8		9.6 - 23.9	
Hf/Hf* ^d	0.63 - 1.07		0.77 - 1.10		0.49 - 0.86	
Ba/Th	198.2 - 316.1		75.2 - 203.5		48.0 - 152.4	
Ce/Pb	9.4 - 13.5		16.3 - 30.3		12.6 - 33.6	
Nb/U	38.9 - 54.2		30.3 - 55.6		36.5 - 51.6	
Dy/Er	2.55 - 2.75		1.91 - 2.55		2.36 - 3.11	
⁸⁷ Sr/ ⁸⁶ Sr	0.7051 - 0.7057		0.7033 - 0.7054			
ε _{Nd}	-5.8 - -3.2		-1.7 - 5.1			
²⁰⁶ Pb/ ²⁰⁴ Pb	16.7 - 17.1		16.8 - 18.1			

786 ^a Wudalianchi and Erkeshan data from Basu et al., (1991), Zhang et al., (1991), Zou et al., (2003), Chu
787 et al. (2013); NCC data from Zhang et al. (2003), Tang et al. (2006); SCB data from Huang et al.
788 (2015).

789 ^b The major element composition of primary melt is estimated from the samples with the highest
790 MgO/Al₂O₃ ratios; the trace element ratios **denote the variability of the complete dataset.**

791 ^c 1SD = one standard deviation.

792 ^d Hf/Hf* = Hf_N/(Sm_N × Nd_N)^{0.5}

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9