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Magnesium isotopic systematics of continental basalts from the North China craton: Implications for tracing subducted carbonate in the mantle

Wei Yang [a,b,*], Fang-Zhen Teng [b], Hong-Fu Zhang [a], Shu-Guang Li [c,d]

[a] State Key Laboratory of Lithospheric Evolution, Institute of Geology and Geophysics, Chinese Academy of Sciences, P.O. Box 9825, Beijing 10029, China
[b] Isotope Laboratory, Department of Geosciences and Arkansas Center for Space and Planetary Sciences, University of Arkansas, Fayetteville, AR 72701, USA
[c] CAS Key Laboratory of Crust-Mantle Materials and Environments, School of Earth and Space Sciences, University of Science and Technology of China, Hefei 230026, China
[d] State Key Laboratory of Geological Processes and Mineral Resources, China University of Geosciences, Beijing 100083, China

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ABSTRACT

To explore the possibility of tracing recycled carbonate by using Mg isotopes and to evaluate the effects of the western Pacific oceanic subduction on the upper mantle evolution of the North China craton, Mg isotopic compositions of the Mesozoic–Cenozoic basalts and basaltic andesites from the craton have been investigated. The samples studied here come from a broad area in the craton with variable ages of 125–6 Ma, and can be divided into two groups based on geochemical features: the >120 Ma Yixian basalts and basaltic andesites, and the <110 Ma Fuxin and Taihang basalts. Our results indicate that these two groups have distinct Mg isotopic compositions. The >120 Ma Yixian basalts and basaltic andesites, with low Ce/Pb, Nb/U ratios and lower-crust like Sr-Nd-Pb isotopic compositions, have a mantle-like Mg isotopic composition, with δ^{26}Mg values ranging from −0.31‰ to −0.25‰ and an average of −0.27 ± 0.05‰ (2SD, $n = 5$). This suggests that continental crust contamination may strongly modify their Ce/Pb, Nb/U ratios and Sr-Nd-Pb isotopic compositions but do not influence their Mg isotopic compositions. By contrast, the <110 Ma basalts from Fuxin and Taihang exhibit lower δ^{26}Mg values of −0.60‰ to −0.42‰, with an average of −0.46 ± 0.10‰ (2SD). Since these basalts still preserve mantle-like Sr-Nd-Pb isotopic compositions and Ce/Pb, Nb/U ratios and have high U/Pb and Th/Pb ratios similar to those of HIMU basalts, the light Mg isotopic composition is most likely derived from interaction of their mantle source with isotopically light recycled carbonate melt. Since the Tethys and Mongolia oceanic subductions from south and north toward the North China craton were terminated in the Triassic and light Mg isotopic signature in basalts did not appear before 120 Ma, the subducted Pacific oceanic crust could be the major source of the recycled carbonate. Therefore, this study not only presents an example to trace recycled carbonate using Mg isotopes but also confirms the important role of the western Pacific oceanic subduction in generating the <110 Ma basalts in the North China craton.

1. Introduction

The “carbon cycle” constitutes one of the most important areas of the Earth science research in this century. The deep carbon cycle refers to carbon ingassing to the mantle through subduction and outgassing through magmatic and volcanic processes. Altered oceanic crust contains a significant fraction of carbonate (Alt and Teagle, 1999), and subduction zone dehydration do not significantly remove this carbonate (Yaxley and Green, 1994; Molina and Poli, 2000; Kerrick and Connolly, 2001). This carbonate delivered to the mantle then significantly affects chemical and physical properties of the mantle.

Carbon isotopes are generally used to trace the subducted carbonate, which are efficient to identify recycled organic carbon but not very sensitive to inorganic carbon. However, the subducted carbon is dominated by inorganic carbonate (Plank and Langmuir, 1998; Alt and Teagle, 1999). Therefore, additional tracers are required to better identify such recycled inorganic carbonate in the mantle.

Magnesium (Mg) isotopes could be one of such tracers. Magnesium isotope fractionation is limited during high temperature processes (Teng et al., 2007; 2010a; Handler et al., 2009; Yang et al., 2009; Bourdon et al., 2010; Liu et al., 2010), but is significant during low temperature processes (Galy et al., 2002; Young and Galy, 2004; Tipper et al., 2006a; 2006b; 2008; 2010; Pogge von Strandmann et al., 2008; Higgins and Schrag, 2010; Li et al., 2010; Teng et al., 2010b). The mantle is homogeneous in its Mg isotopic composition, with an average δ^{26}Mg value of −0.25 ± 0.07‰ (Teng et al., 2007; 2010a; Handler et al., 2009; Yang et al., 2009; Bourdon et al., 2010; Liu et al., 2011). The upper continental crust is heterogeneous and on average similar

* Corresponding author at: State Key Laboratory of Lithospheric Evolution, Institute of Geology and Geophysics, Chinese Academy of Sciences, P.O. Box 9825, Beijing 10029, China. Tel.: +86 10 82998514; fax: +86 10 62010846.
E-mail address: yangw@mail.iggcas.ac.cn (W. Yang).

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to the mantle (Shen et al., 2009; Li et al., 2010; Liu et al., 2010). Notably, carbonates have significant light Mg isotopic compositions with δ^{26}Mg value ranging from −5.31 to −1.09 (Young and Galy, 2004; Tipper et al., 2006a; Brenot et al., 2008; Pogge von Strandmann, 2008; Hippler et al., 2009; Higgins and Schrag, 2010), which makes Mg isotopes a potential tracer of recycled carbonate in the mantle.

The Mesozoic and Cenozoic basalts from the North China craton (NCC) are ideal samples to explore Mg isotopes as a potential tracer of recycled carbonate in the mantle. There is an abrupt change in the chemical compositions between basalts formed before 120 Ma and those formed after 110 Ma. The >120 Ma basalts with strong continental geochemical signatures could be derived from the mantle that has been contaminated by delaminated lower continental crust (Huang et al., 2007; Liu et al., 2008), whereas the <110 Ma basalts with mantle-like Sr-Nd-Pb isotope and HIMU-like trace element signatures could be originated from the mantle affected by subducted oceanic crust (Yang and Li, 2008; Zhang et al., 2009; Xu et al., 2010; Wang et al., 2011). Recent studies suggest that the source of the <110 Ma basalts could be metasomatized by carbonate (Chen et al., 2009; Zeng et al., 2010). Therefore, Mg isotopic composition of these basalts could help us understand how recycled carbonate affects the mantle.

Here, we investigated Mg isotopic compositions of the >120 Ma basalts and basaltic andesites, and <110 Ma basalts from the North China craton. Our results indicate that Mg isotopic compositions of basaltic lavas change abruptly from >120 Ma to <110 Ma. The >120 Ma basalts and basaltic andesites have mantle-like Mg isotopic composition whereas the <110 Ma basalts are isotopically light (δ^{26}Mg = −0.60 to −0.42‰). This light Mg isotopic composition most likely results from metasomatism by recycled carbonate melt, which could be derived from subducted Pacific oceanic crust. This study not only presents an example to trace recycled carbonate by using Mg isotopes but also confirms the important role of the western Pacific oceanic subduction in generating the <110 Ma basalts in the North China craton.

2. Geological settings and sample description

2.1. Geological settings

The North China craton is one of the world's oldest Archean cratons, which preserves >3.8 Ga crustal remnants (Liu et al., 1992). It can be divided into three regions: the Eastern Block, the Western Block and the Trans-North China Orogen. The Trans-North China Orogen was formed by the collision between the Eastern Block and Western Block at ~1.85 Ga, marking the final amalgamation of the NCC (Zhao et al., 2008). Two linear geological and geophysical zones, the NEE strike Tan-Lu fault zone and Daxing'anling–Taihangshan gravity lineament crosscut the craton (Fig. 1). The NCC experienced multiple circum-craton subductions and collisions, which are manifested by the Paleozoic to Triassic Qinling-Dabie-Sulu ultrahigh-pressure belt in south, the Central Asian Orogenic Belt in north and the Mesozoic–Cenozoic subduction of Pacific plate in the east (Fig. 1).

The Mesozoic and Cenozoic basalts occurred widely in the NCC. The basalts erupted before 120 Ma (e.g., Yixian basalts) have distinctive chemical compositions from those erupted after 110 Ma (e.g. Fuxin and Taihang basalts). The >120 Ma basalts are characterized by negative ε_{Nd} (down to −20), variable ^{87}Sr/^{86}Sr, low radiogenic Pb isotope ratios, and typical "continental" geochemical signatures, such as enrichment of large ion lithophile elements (LILE, e.g., Rb and Ba) and depletion of high field strength element (HFSE, e.g., Nb and Ta) (Guo et al., 2001; 2003; Qiu et al., 2002; Zhang et al., 2002; 2003; Zhang, 2007; Liu et al., 2008; Yang and Li, 2008). By contrast, the <110 Ma basalts have relatively depleted Nd and Sr isotopic compositions and HIMU-like trace element patterns, e.g. depletion of Rb and Pb and enrichment of Nb (Peng et al., 1986; Song, 1990; Zhi et al., 1990; 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).

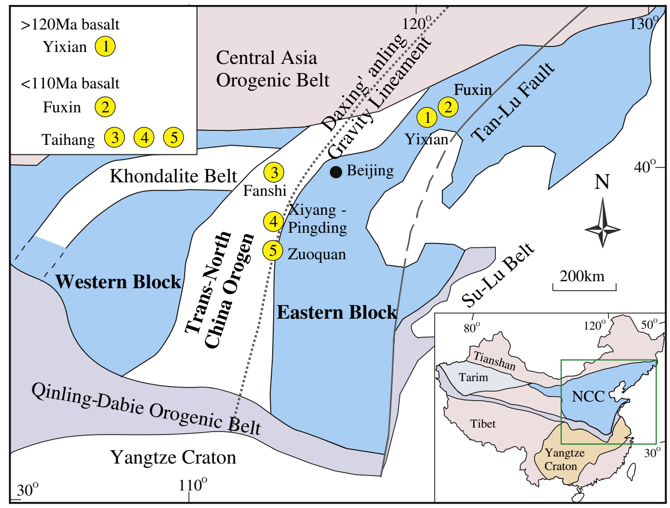

Fig. 1. Simplified geological map showing sample localities and subdivisions of the North China craton (modified from Tang et al., 2010). The North China craton is cut by two major linear zones, Tan-Lu fault zone to the east and Daxing'anling gravity lineament to the west. The basalt samples from Fanshi, Xiyang-Pingding and Zuoquan are all called Taihang basalts in this paper. Inset shows location of the NCC relative to other blocks and fold belts.

2.2. Sample description

The samples studied here are collected from Yixian and Fuxin in the Eastern Block, and Taihang in Trans-North China Orogen, respectively (Fig. 1). Their petrology, chemical and Sr-Nd-Pb isotopic compositions have been presented in previous studies (Zhang et al., 2003; Tang et al., 2006; Yang and Li, 2008). Only a brief description is given below.

Yixian is located to the west of Tan-Lu fault zone at the northern margin of the NCC (Fig. 1). The Yixian lavas mainly consist of basalts, andesites and rhyolites dated to 125–120 Ma by zircon U-Pb method (Yang and Li, 2008). Like many >120 Ma basalts in the craton, the Yixian basalts and basaltic andesites studied here have typical "lower crustal" signatures, such as depletion of HFSEs, low radiogenic Pb (^{206}Pb/^{207}Pb = 16.21–16.66, ^{207}Pb/^{204}Pb = 15.22–15.29) and low ε_{Nd}(T) (−13.4 to −9.7) (Yang and Li, 2008). Their low Ce/Pb ratios (4.6 to 8.2) are similar to the average Ce/Pb ratio (~5.0) of the lower continental crust (Rudnick and Gao, 2003) (Fig. 2a), suggesting the involvement of lower crustal materials in producing these basalts. The samples investigated here include three basalts and two basaltic andesites, and have relatively high SiO_2 contents (52.8–56.7%) and low CaO/Al_2O_3 ratios (0.36–0.46; Fig. 2c). These samples are fresh as indicated by petrographic studies (Yang and Li, 2008).

Fuxin is located near Yixian at the northern margin of the NCC (Fig. 1). Taihang is more than 700 km west from Yixian and Fuxin, and Taihang samples are from three locations: Fanshi, Xiyang-Pingding and Zuoquan (Fig. 1). Although these basalts (26–6 Ma, Ar–Ar method; Tang et al., 2006) erupted >70 Myrs later than the Fuxin basalts (106–100 Ma, K-Ar and Ar–Ar methods; Zhang et al., 2003; Yang and Li, 2008), they have similar geochemical characteristics to the Fuxin basalts. They both are characterized by OIB-type trace element distribution patterns, with enriched light rare earth elements (LREEs) and Nb and depleted Rb and Pb. They have lower ^{87}Sr/^{86}Sr (0.7036–0.7054), higher ε_{Nd}(T) (−4.0–5.1) and Pb isotopic ratios (^{206}Pb/^{207}Pb = 16.76–18.32, ^{207}Pb/^{204}Pb = 15.28–15.46) than those of Yixian basalts (Zhang et al., 2003; Tang et al., 2006). Their Ce/Pb and

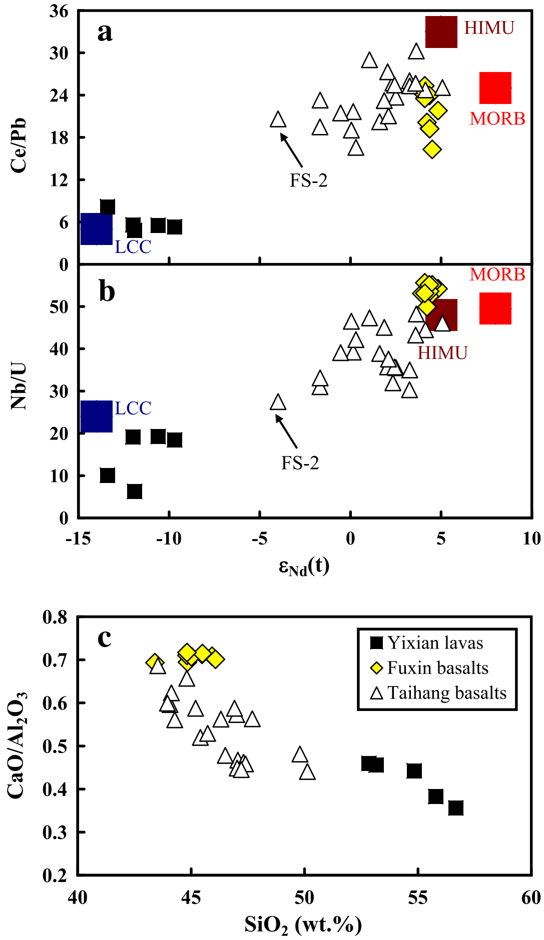

Fig. 2. (a) Ce/Pb vs. $\varepsilon_{Nd}(t)$, (b) Nb/U vs. $\varepsilon_{Nd}(t)$, and (c) CaO/Al_2O_3 vs. SiO_2 for the Yixian lavas, and the Fuxin and Taihang basalts studied here. Database: MORB and HIMU from Sun and McDonough (1989), lower continental crust (LCC) from Rudnick and Gao (2003), the Yixian lavas, and Fuxin and Taihang basalts from Zhang et al. (2003), Tang et al. (2006) and Yang and Li (2008).

Nb/U ratios vary from 16.3 to 30.3 and 27.5 to 55.6, respectively (Fig. 2a, b), close to those of MORB and HIMU basalts (Sun and McDonough, 1989). They have low SiO_2 contents (43.4–50.1%) and high CaO/Al_2O_3 ratios (0.44–0.72; Fig. 2c). Nine Fuxin basalts and 21 Taihang basalts were analyzed for Mg isotopes here.

3. Analytical methods

Magnesium isotopic analyses were performed at the Isotope Laboratory of the University of Arkansas, Fayetteville, following the established procedures (Teng et al., 2007; 2010a; Yang et al., 2009; Li et al., 2010). Only a brief description is given below.

All chemical procedures were carried out in a clean laboratory environment. Depending on Mg concentration, 1 to 25 mg of sample powder was weighted in Savillex screw-top beakers in order to have >50 μg Mg in the solution and was then dissolved in a mixture of concentrated $HF\text{-}HNO_3\text{-}HCl$ solution. Separation of Mg was achieved by cation exchange chromatography with Bio-Rad 200–400 mesh AG50W-X8 resin in 1N HNO_3 media following the established procedures (Teng et al., 2007; 2010a; Yang et al., 2009; Li et al., 2010). Magnesium isotopic compositions were analyzed by the standard bracketing method using a Nu Plasma MC-ICP-MS. Magnesium isotope data are reported in standard δ-notation relative to DSM3: $\delta^X Mg = [(\delta^X Mg/^{24}Mg)_{sample}/(\delta^X Mg/^{24}Mg)_{DSM3} - 1] \times 1000$, where X = 25 or 26. $\Delta^{25}Mg' = \delta^{25}Mg' - 0.512 \times \delta^{26}Mg'$, where $\delta^X Mg' = 1000 \times \ln[(\delta^X Mg + 1000)/1000]$ (Young and Galy, 2004).

The internal precision of the measured $^{26}Mg/^{24}Mg$ ratio based on ≥4 repeat runs of the same sample solution during a single analytical session is <±0.1‰ (Teng et al., 2010a). Multiple analyses of olivine KH-1 yielded $\delta^{26}Mg$ values of −0.33 to −0.27 (Table 1), which is in agreement with that reported by Teng et al. (2010a) ($\delta^{26}Mg$ = −0.27 ± 0.07‰; 2SD, n = 16). The synthetic granite standard (IL-granite) yielded a $\delta^{26}Mg$ value of −0.06‰ (Table 1), within uncertainties of the reported value of −0.01 ± 0.06‰ (2SD, n = 13; Teng et al., 2010a). San Carlos olivine standard studied by Chakrabarti and Jacobsen (2010) was also analyzed here for inter-laboratory comparison, which yielded $\delta^{26}Mg$ values of −0.33 to −0.30‰ (Table 1), similar to those reported by Young et al. (2009) and Liu et al (2010), but heavier than the value ($\delta^{26}Mg$ = −0.55‰) reported by Chakrabarti and Jacobsen (2010). The cause for these differences is unknown.

4. Results

Magnesium isotopic compositions of reference materials and the Mesozoic and Cenozoic basalts from the NCC are reported in Table 1 along with their chemical compositions (Zhang et al., 2003; Tang et al., 2006; Yang and Li, 2008). In a plot of $\delta^{25}Mg$ vs. $\delta^{26}Mg$ (Fig. 3), all samples fall along the terrestrial equilibrium mass fractionation curve with a slope of 0.521. The $\Delta^{25}Mg'$ values of all samples are within ±0.04‰ (Table 1). Overall, Mg isotopic compositions of these basalts from NCC vary significantly, with $\delta^{26}Mg$ values ranging from −0.60‰ to −0.25‰ and $\delta^{25}Mg$ values from −0.32‰ to −0.14‰ (Table 1).

The Yixian basalts and basaltic andesites have homogenous Mg isotopic compositions (Fig. 4 and Table 1), with $\delta^{26}Mg$ ranging from −0.31 to −0.25‰ and an average of −0.27 ± 0.05‰ (2SD, n = 5), which is within analytical uncertainties of the average $\delta^{26}Mg$ of the mantle (−0.25 ± 0.07‰; Teng et al., 2010a).

29 out of the 30 Fuxin and Taihang basalts exhibit a variation of 0.18‰ in $\delta^{26}Mg$ values from −0.60 to −0.42‰, which are obviously lighter than those of the average mantle (−0.25 ± 0.07‰; Teng et al., 2010a) but close to those of some wehrlites (−0.48 to −0.43‰) in the North China craton (Yang et al., 2009). Sample FS-2 deviates from the population of the others with a $\delta^{26}Mg$ value of −0.30 ± 0.06‰ (Fig. 4). Notably, this sample also has the lowest $\varepsilon_{Nd}(t)$ value of −4.0 and lowest Nb/U ratio of 27.5 among all Fuxin and Taihang basalts (Fig. 2b).

5. Discussion

Magnesium isotopic composition of basalts is generally expected to be mantle-like because basaltic differentiation does not fractionate Mg isotopes (Teng et al., 2007; 2010a) and Mg isotopic composition of the mantle is difficult to be modified by recycled materials, which generally have low-Mg content compared to the mantle (MgO = ~37.8%; McDonough and Sun, 1995). For example, the >120 Ma Yixian basalts are considered to be derived from the mantle source hybridized by continental crustal materials based on their low Ce/Pb and Nb/U ratios and enriched Sr-Nd isotopic compositions (Yang and Li, 2008). However, they still preserve mantle-like Mg isotope composition, with $\delta^{26}Mg$ ranging from −0.31 to −0.25‰ and an average of −0.27 ± 0.05‰ (2SD, n = 5).

By contrast, most of the <110 Ma basalts have lighter Mg isotopic composition than the average mantle (Teng et al., 2010a), with $\delta^{26}Mg$ values from −0.60 to −0.42‰. Similar light Mg isotopic composition has been previously observed in the wehrlites from the Beiyan area, NCC (Yang et al., 2009). In general, Mg isotopic compositions of basalts and peridotites from the NCC can be divided into two groups (Fig. 5). One group, including the basalts and two wehrlites, has relatively light Mg isotopic composition (Yang et al., 2009 and this study). The other group has mantle-like Mg isotopic composition, represented by the majority of peridotites (Yang et al., 2009; Teng et al., 2010a). This may suggest that the isotopically light wehrlites could be either the mantle

Table 1
Magnesium isotopic composition of standards and volcanic rocks from Yixian, Fuxin and Taihang Mountains, North China craton.

Sample	SiO_2 [a]	MgO [a]	$\varepsilon_{Nd}(t)$ [a]	$\delta^{26}Mg$ [b]	2SD [c]	$\delta^{25}Mg$ [b]	2SD [c]	$\Delta^{25}Mg'$ [d]
Standards								
KH-1 Olivine[e]				−0.27	0.06	−0.17	0.10	−0.03
Repeat[f]				−0.28	0.06	−0.15	0.05	0.00
Repeat				−0.28	0.07	−0.13	0.05	+0.02
Repeat				−0.30	0.06	−0.15	0.04	+0.01
Repeat				−0.28	0.05	−0.14	0.07	+0.01
Repeat				−0.33	0.09	−0.13	0.05	+0.04
Repeat				−0.31	0.06	−0.15	0.05	+0.01
Repeat				−0.32	0.06	−0.17	0.05	0.00
Average				−0.30	0.04	−0.15	0.04	
San Calos Ol				−0.33	0.07	−0.17	0.05	0.00
Repeat				−0.30	0.07	−0.16	0.05	0.00
Average				−0.32	0.04	−0.17	0.01	
Il-granite[g]				−0.06	0.06	−0.02	0.05	+0.01
Repeat				−0.06	0.05	−0.05	0.07	−0.02
Average				−0.06	0.01	−0.04	0.04	
Yixian basalts and basaltic andesites (125–120 Ma)								
HBJ4-1	54.83	6.45	−10.6	−0.25	0.06	−0.14	0.05	−0.01
HBJ4-2	53.16	6.23	−12.0	−0.28	0.06	−0.14	0.06	+0.01
HBJ4-3	52.84	7.03	−9.7	−0.27	0.06	−0.15	0.06	−0.01
SHT-14	55.78	5.21	−13.4	−0.26	0.06	−0.15	0.06	−0.01
SHT-3	56.67	5.92	−11.9	−0.31	0.07	−0.19	0.05	−0.03
Average				−0.27	0.05	−0.15	0.04	
Fuxin basalts (106–100 Ma)								
JG-01	44.84	8.31	4.8	−0.46	0.06	−0.27	0.05	−0.03
JG-02	45.48	8.25	3.9	−0.54	0.06	−0.26	0.05	+0.02
Repeat				−0.52	0.05	−0.29	0.07	−0.02
JG-03	44.82	8.38	4.1	−0.48	0.06	−0.25	0.05	0.00
JG-04	45.92	8.09	4.4	−0.53	0.06	−0.28	0.05	0.00
JG-05	45.01	8.30	4.5	−0.46	0.06	−0.24	0.05	0.00
JG-06	46.07	8.06	4.2	−0.47	0.06	−0.25	0.05	−0.01
JG-07	43.40	8.39	4.4	−0.60	0.06	−0.32	0.05	−0.01
Repeat				−0.55	0.06	−0.27	0.04	+0.02
JG-08	45.50	8.16	4.4	−0.49	0.06	−0.25	0.05	+0.01
JG-09	44.82	8.30	4.1	−0.49	0.07	−0.24	0.05	+0.02
Average				−0.51	0.09	−0.27	0.05	
Taihang basalts (26–6 Ma)								
FS-1	44.07	8.81	3.3	−0.44	0.04	−0.22	0.06	+0.01
FS-8	44.28	8.30	2.3	−0.46	0.04	−0.24	0.06	0.00
FS-10	44.13	9.09	2.5	−0.46	0.04	−0.22	0.06	+0.02
FS-30	43.53	9.93	3.3	−0.44	0.04	−0.25	0.06	−0.02
FS-32	45.20	8.74	2.1	−0.49	0.04	−0.27	0.06	−0.01
FS-33	43.96	8.81	2.4	−0.48	0.04	−0.22	0.06	+0.03
HHL-1	45.41	8.10	1.6	−0.43	0.04	−0.22	0.06	0.00
HHL-2	45.73	8.26	2.1	−0.52	0.06	−0.26	0.05	+0.01
FS-2	47.01	8.36	−4.0	−0.30	0.06	−0.16	0.05	0.00
FS-3	47.06	7.43	−1.7	−0.53	0.06	−0.26	0.05	+0.02
FS-9	44.81	9.85	−1.7	−0.46	0.06	−0.24	0.05	0.00
FS-36	46.31	8.47	0.2	−0.51	0.06	−0.25	0.05	+0.02
FS-38	46.91	8.91	−0.5	−0.45	0.06	−0.23	0.05	0.00
FHS-1	47.70	7.83	1.1	−0.44	0.07	−0.25	0.04	−0.02
GB-2	47.31	6.72	3.6	−0.50	0.07	−0.27	0.04	−0.01
JD-1	47.03	6.48	4.1	−0.46	0.07	−0.26	0.04	−0.02
JX-1	47.40	6.72	1.9	−0.49	0.07	−0.25	0.04	+0.01
JX-3	46.51	6.94	5.1	−0.46	0.07	−0.25	0.04	−0.01
MAS-1	47.21	6.47	3.6	−0.54	0.07	−0.29	0.04	−0.01
ZQ-1	49.80	7.17	0.3	−0.42	0.07	−0.22	0.04	0.00
ZQ-4	50.13	6.67	0.1	−0.42	0.07	−0.22	0.04	0.00
Average				−0.46	0.10	−0.24	0.05	

[a] Data from Zhang et al. (2003), Tang et al. (2006), Yang and Li (2008).
[b] $\delta^{X}Mg = [(^{X}Mg/^{24}Mg)_{sample}/(^{X}Mg/^{24}Mg)_{DSM3}-1] \times 1000$, where X = 25 or 26 and DSM3 is Mg solution made from pure Mg metal (Galy et al., 2003).
[c] 2SD = 2 times the standard deviation of the population of > 20 repeat measurements of the standard during a session.
[d] $\Delta^{25}Mg' = \delta^{25}Mg' - 0.512 \times \delta^{26}Mg'$, where $\delta^{X}Mg' = 1000 \times \ln[(\delta^{X}Mg + 1000)/1000]$ (Young and Galy, 2004).
[e] KH-1 olivine is an in-house standard that has been analyzed through whole-procedural column chemistry and instrumental analysis with $\delta^{26}Mg = -0.27 \pm 0.07$ and $\delta^{25}Mg = -0.14 \pm 0.04$ ($n = 16$; 2SD) relative to DSM-3 (Teng et al., 2010a).
[f] Repeat of column chemistry and measurement of different aliquots of a stock solution.
[g] IL-granite is a synthetic solution with Mg:Al:Fe:Ca:K:Na:Ti:Ni = 1:30:5:5:20:10:0.1:0.1.

source of or cumulates from the < 110 Ma basalts. Three potential processes may account for such a light Mg isotopic composition in the < 110 Ma basalts and the two wehrlites: (1) Kinetic Mg isotope fractionation in the mantle source; (2) Interaction with silicate melts (either crustal contamination or mantle metasomatism by recycled crustal material); (3) Interaction with carbonate rocks.

Table 2
Parameters for model calculations.

	MgO (wt.%)	$\delta^{26}Mg$	FeO (wt.%)	Nd (ppm)	ε_{Nd}	Pb (ppm)	$^{206}Pb/^{204}Pb$
DMM [a]	37.8	−0.25		1.1	8	0.2	18
Primitive magma from SCLM [b]	12.0	−0.25		7.8	−9.7	1.2	18.5
Melt from LCC [c]	3.5	−0.60		28.9	−12.5	15.3	16.24
Melt from OC [d]	4.6	−0.83	7.3				
Mantle harzburgites [e]	45.2	−0.24	8.5				
Kd $^{Fe\text{-}Mg}$ between peridotite and melt: 0.3 [f]							

[a] DMM = Depleted MORB mantle. Database: MgO (McDonough and Sun, 1995); $\delta^{26}Mg$ (Teng et al., 2010a); ε_{Nd} (Jahn et al., 1999); Nd, Pb and $^{206}Pb/^{204}Pb$ (Sun and McDonough, 1989).
[b] SCLM = sub-continental lithospheric mantle. Database: $\delta^{26}Mg$ (Teng et al., 2010a); ε_{Nd} and $^{206}Pb/^{204}Pb$ (Zheng and Lu, 1999); MgO, Nd and Pb based on data of oceanic basalt (Sun and McDonough, 1989).
[c] LCC = lower continental crust of the northern margin of the NCC. $\delta^{26}Mg$ is assumed as light as −0.6 to demonstrate the light isotopic composition of the basalts could not derive from crustal contamination. Other data are from (Yang and Li, 2008).
[d] OC = oceanic crust. Database: MgO and FeO from partial melt of an oceanic gabbro G108 (Sobolev et al., 2005); $\delta^{26}Mg$ is assumed as light as −0.83 to demonstrate the light isotopic composition of the basalts could not derive from silicate melt metasomatism.
[e] Data are from two harzburgites of the NCC (Wu et al., 2006; Teng et al., 2010a).
[f] Data are estimated based on Kd $^{Fe\text{-}Mg}$ of olivine and pyroxene with melt (Roeder and Emslie, 1970; Brey and Kohler, 1990).

Below, we will first evaluate which process could produce the observed light Mg isotopic composition in these basalts, and then discuss their origin together with their other geochemical characteristics.

5.1. Kinetic Mg isotope fractionation

Kinetic Mg isotope fractionation driven by chemical and thermal diffusion can occur at high temperatures as shown in experimental studies (Richter et al., 2008; Huang et al., 2009; 2010) but unlikely be the mechanisms that generate such light Mg isotopic compositions in the mantle source of the <110 Ma basalts. One recent study shows co-variation of Mg and Li isotopes in mantle xenoliths (Pogge von Strandmann et al., 2011), which is explained by chemical diffusion driven kinetic isotope fractionations during the transport of mantle xenoliths. However, such kinetic isotope fractionation by chemical diffusion could be ruled out to produce the isotopically light mantle source of the <110 Ma basalts from the NCC. First, lighter isotopes diffuse faster than heavier ones during chemical diffusion. When Mg diffuses from peridotites to host basalts, Mg isotopic composition of peridotites should be shifted towards heavy values, as shown in Pogge von Strandmann et al. (2011). This is the opposite to the wehrlites from the NCC. In addition, peridotites affected by diffusion also display large inter-mineral Mg isotope fractionation, with $\Delta^{26}Mg_{cpx\text{-}ol}$ up to 0.31‰ (Pogge von Strandmann et al., 2011). By contrast, the $\Delta^{26}Mg_{cpx\text{-}ol}$ values of all peridotite xenoliths from the NCC are <0.10‰, indicating that Mg isotopes reach equilibrium in these peridotite xenoliths (Yang et al., 2009). This further rules out chemical diffusion driven kinetic fractionation as causes of those isotopically light wehrlites.

Kinetic isotope fractionation by thermal diffusion could also be ruled out since the <110 Ma basalts studied here come from a very broad area and have variable ages (106–6 Ma). It is unlikely that thermal diffusion could operate on such a large scale and continue over such a long period of time (>70 Myrs from the Fuxian basalts to the Taihang basalts). Therefore, both chemical and thermal diffusion appear unable to create the light Mg isotopic composition in the <110 Ma basalts from the NCC.

5.2. Interactions with silicate melts

Crustal contamination by silicate wall rocks and mantle metasomatism by recycled silicate crustal material are both unlikely to produce such a light Mg isotopic composition in the <110 Ma basalts because crustal materials generally have low-Mg content compared to the mantle (MgO = ~37.8%; McDonough and Sun, 1995). The silicate rocks in continental crust have a relatively heavier Mg isotopic composition with $\delta^{26}Mg$ ranging from −0.44 to 0.90‰ (Shen et al., 2009; Li et al., 2010; Liu et al., 2010). Even assuming that they could

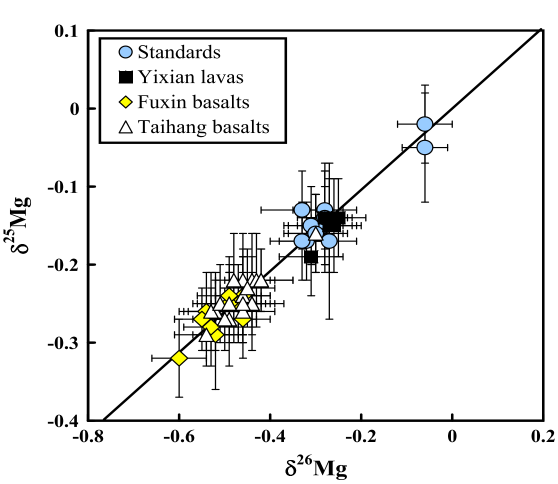

Fig. 3. Magnesium isotope plot of the samples in this study. The solid line represents the terrestrial fractionation line with a slope equal to 0.521. Error bars represent 2SD uncertainties. Data are from Table 1.

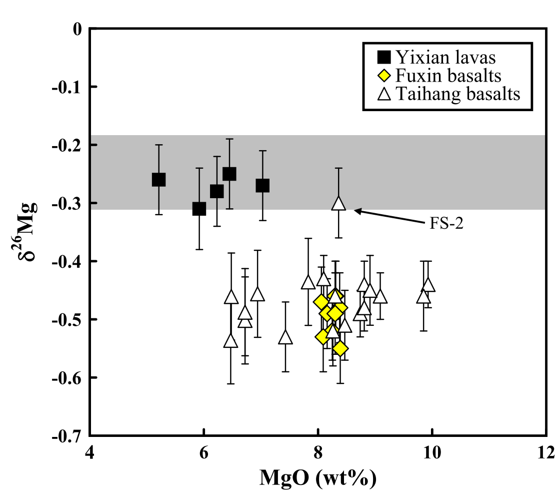

Fig. 4. $\delta^{26}Mg$ vs. MgO (wt.%) for the Yixian lavas, and the Fuxin and Taihang basalts. The grey bar represents global oceanic basalts ($\delta^{26}Mg$ = −0.25 ± 0.07‰; Teng et al., 2010a). Error bars represent 2SD uncertainties. Data are from Table 1.

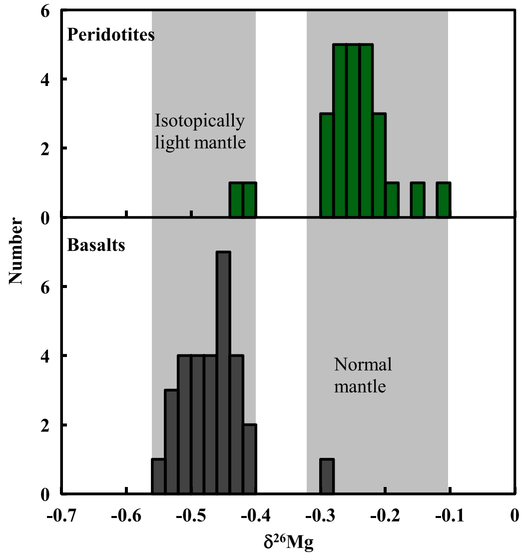

Fig. 5. Histogram for δ^{26}Mg values of the <110 Ma basalts and peridotites from the North China craton. The grey bars represent two kind of mantle sources with distinct Mg isotopic composition. Data are from Yang et al. (2009), Teng et al. (2010a) and Table 1.

have an average δ^{26}Mg value as low as −0.60‰, binary mixing model indicates that more than 60% crustal contamination is required to produce such a light Mg isotopic composition in the <110 Ma basalts (Fig. 6).

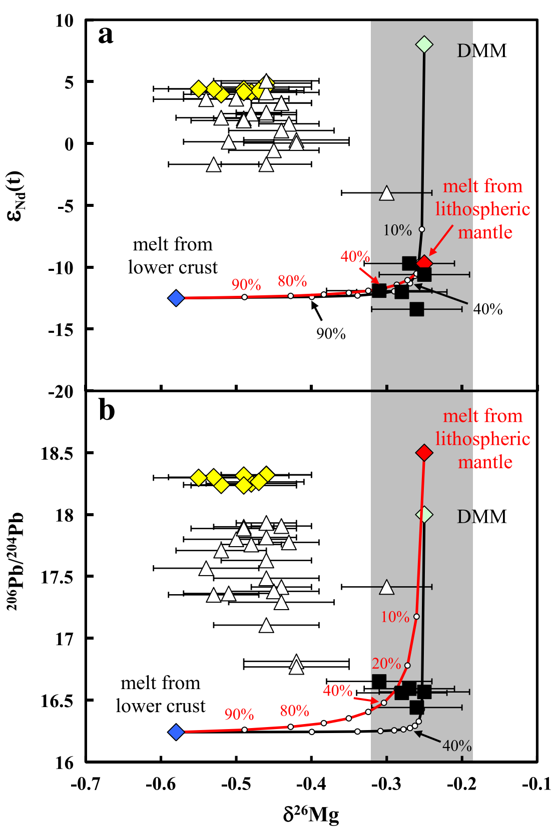

Fig. 6. Plots of $\varepsilon_{Nd}(t)$ and $^{206}Pb/^{204}Pb$ vs. δ^{26}Mg for the Yixian lavas, and the Fuxin and Taihang basalts. The symbols are the same as Fig. 4. The grey bar represents global oceanic basalts (δ^{26}Mg = −0.25 ± 0.07‰; Teng et al., 2010a). Error bars represent 2SD uncertainties. The curves represent mixing of two end members. Numbers at small circles represent fraction of the melt derived from the lower continental crust. Data are from Table 1. See Table 2 for modeling parameters.

This large amount of crustal contamination should have shifted Nd-Pb isotopic compositions and Ce/Pb, Nb/U ratios of these basalts to crustal-like compositions (Figs. 2 and 6). The mantle-like Nd-Pb isotopic compositions and Ce/Pb, Nb/U ratios in the <110 Ma basalts hence suggest that the contamination from silicate rocks in continental crust is limited and can be ruled out as causes of such light Mg isotopic compositions of these basalts.

For mantle metasomatism, the potential metasomatic agent could be originated from the delaminated continental crust (Liu et al., 2008; Zeng et al., 2011), the asthenospheric mantle (Xiao et al., 2010) or subducted oceanic crust (Yang and Li, 2008; Zhang et al., 2009; Xu et al., 2010; Wang et al., 2011). As mentioned above, the continental crust has a relatively heavier Mg isotopic composition with δ^{26}Mg ranging from −0.44 to 0.90 (Shen et al., 2009; Li et al., 2010; Liu et al., 2010), and the asthenosphere has a homogenous Mg isotopic composition as sampled by global MORBs (δ^{26}Mg = −0.25 ± 0.06, 2SD; Teng et al., 2010a). Their Mg isotopic compositions are both heavier than those of the <110 Ma basalts (−0.60 to −0.42).

Alternatively, the oceanic crust may have a relatively lighter Mg isotopic composition. First, seawater has a homogeneous and light Mg isotopic composition with δ^{26}Mg = −0. 832 ± 0.068 (Ling et al., 2011 and references therein). Water–rock interactions should hence lead to an isotopically light oceanic crust. In addition, the flux-weighted average δ^{26}Mg value of global runoff is −1.09 (Tipper et al., 2006b), which is even lighter than that of the seawater. Assuming a steady-state Mg isotopic composition for the oceans, the light Mg in the global runoff must balance the flux from the oceans to the oceanic crust, which suggests the oceanic crust has to be isotopically light (Teng et al., 2010b). Moreover, δ^{26}Mg value of a marine shale standard (SCo-1) is −0.94 ± 0.08 (Li et al., 2010).

However, even assuming that the silicate melt from the subducted oceanic crust has δ^{26}Mg value as low as seawater (−0.8‰), model calculation suggests that a total melt/rock ratio of 3.2–4.4 is required to form the isotopically light mantle source in the NCC (Fig. 7). Such a high melt/rock ratio may be possible when large volume of melt migrates through the mantle peridotite in a local area. However, the

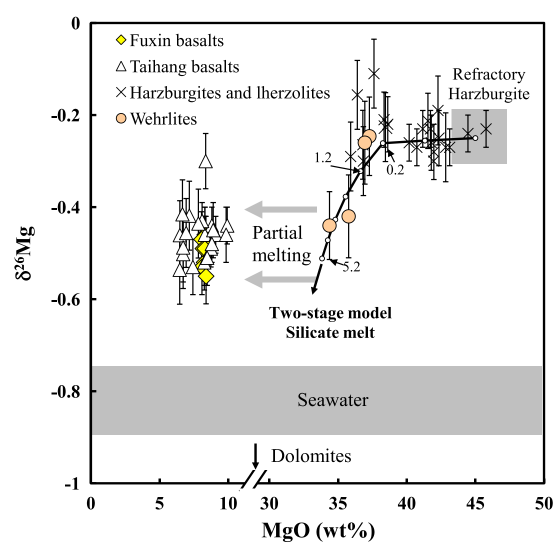

Fig. 7. δ^{26}Mg vs. MgO (wt.%) for the Fuxin and the Fanshi basalts with the peridotites in the North China craton. The grey bar represents seawater (δ^{26}Mg = −0.83 ± 0.07‰, 2SD) (Ling et al., 2011 and references therein). The grey box represents refractory harzburgite from the NCC (Teng et al., 2010a). Average δ^{26}Mg of dolomite is −2.2‰ (Higgins and Schrag, 2010). Numbers at small circles represent total melt/rock ratios. Data are from Yang et al. (2009), Teng et al. (2010a) and Tables 1 and 2. Error bars represent 2SD uncertainties. For modeling reaction of peridotite with silicate melt, two ideal processes are assumed. At the beginning the peridotite simply mixes with the melt, and then just exchange Fe-Mg with the melt. The latter process assumes that peridotite exchanges Fe-Mg with melt to achieve elemental and isotopic equilibrium but never mix with melts.

<110 Ma basalts studied here come from four localities in the NCC. In addition, migration of large volume of melt derived from subducted oceanic crust through the mantle should result in adakitic magma eruption (Rapp et al., 1999), which is inconsistent with current observation that nearly no <110 Ma adakitic rock has been found in the NCC.

Although mantle metasomatism by silicate melt cannot generate the light Mg isotopic composition in the <110 Ma basalts, this does not mean the mantle source of the <110 Ma basalts would have never metasomatized by silicate melt. The mantle could preserve its original Mg isotopic composition even thought it had been significantly metasomatized by silicate melts.

5.3. Interactions with high-Mg carbonate melts

High-Mg carbonate (e.g., dolomite) melt metasomatism however could easily modify Mg isotopic composition of the mantle or the basalts. Dolomite can have high MgO contents and light Mg isotopic composition, with δ^{26}Mg values ranging from −5.31 to −1.09‰ (Young and Galy, 2004; Tipper et al., 2006a; Brenot et al., 2008; Pogge von Strandmann, 2008; Hippler et al., 2009; Higgins and Schrag, 2010). For example, by assuming that dolomite has an average MgO of 20% and δ^{26}Mg of −2.2‰ (Higgins and Schrag, 2010). Both mixing with ~20% dolomitic melt in the mantle and contamination by ~5% dolomite wallrock can shift the basalts to such a light Mg isotopic composition.

In addition, the isotopically light wehrlites in the NCC have higher CaO than those of other peridotites (see Fig. 5b in Yang et al., 2009). This CaO/Al_2O_3 variation in the wehrlites does not result from the mode abundances of clinopyroxene, because the isotopically light wehrlites have similar mode abundance of clinopyroxene (25%) to the "normal" wehrlites (27–28%) (Yang et al., 2009). Thus, the isotopically light wehrlites may be affected by carbonate. And if these wehrlites are cumulates from or source of the <110 Ma basalts, the basalts may also be modified by carbonates. Coincidentally, the isotopically light <110 Ma basalts have relatively higher CaO/Al_2O_3 than those of >110 Ma basalts (Fig. 8c), although this high CaO/Al_2O_3 may partially result from crystal fractionation of plagioclase.

5.3.1. Contamination by carbonate wallrock

Dolomites in the upper continental crust generally have light Mg isotopic composition (Higgins and Schrag, 2010). However, it is unlikely that the light Mg isotopic composition in the <110 Ma basalts could result from interactions with dolomite wallrock. First, the <110 Ma basalts studied here come from a very broad area and have variable ages (106–6 Ma). However, they have nearly identical Mg isotopic composition with δ^{26}Mg values ranging from −0.60 to −0.42‰. It is difficult to explain why they were all contaminated by ~5% dolomite wallrock. Second, dolomite only occupies a very small part of the upper crust. If all lavas have been contaminated by dolomite, they could also be largely contaminated by other crustal materials, which would significantly shift Nd isotopic compositions, Ce/Pb and Nb/U ratios. However, this is not observed in the <110 Ma basalts. Furthermore, the Yixian (~125 Ma) and the Fuxin (~106 Ma) basalts both come from western Liaoning area, thus have similar wallrocks. Although the former have enriched Nd isotopic composition (Yang and Li, 2008), their mantle-like Mg isotopic composition indicates that they maybe escaped from dolomite contamination. On the contrary, the latter with light Mg isotopic composition appears to be significantly contaminated by dolomite, still preserves depleted Nd isotopic composition (Yang and Li, 2008).

In addition, the two isotopically light wehrlites from Beiyan area could not be cumulates from the basalts based on their petrological and geochemical features (Xiao et al., 2010). Their olivines show undulatory extinction and kink bands which cannot be found in cumulates (Xiao et al., 2010). Furthermore, these wehrlites have lower $Cr^{\#}$ in spinel (<40) than those of cumulates (>50) (Xiao et al., 2010). Finally, their cpxs have sieve textures (Xiao et al., 2010), which could be a feature for melt-rock reactions as indicated by the experimental study (Shaw and Dingwell, 2008). This also indicates that there could be an isotopically light mantle source beneath the NCC after 110 Ma, and that a contamination model by dolomite wallrock is not necessary.

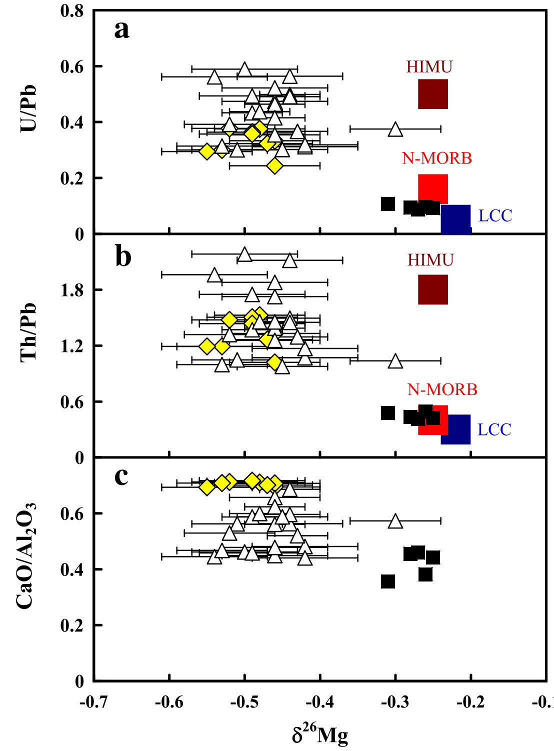

Fig. 8. (a) U/Pb, (b) Th/Pb and (c) CaO/Al_2O_3 vs. δ^{26}Mg for the Yixian lavas, and the Fuxin and Taihang basalts. The symbols are the same as Fig. 4. Error bars represent 2SD uncertainties. Database: U/Pb and Th/Pb ratios of HIMU and MORB from Sun and McDonough (1989), U/Pb and Th/Pb ratios of LCC from Rudnick and Gao (2003), Mg isotopic composition of HIMU, MORB and LCC from Teng et al. (2010a) and Li et al. (2010), other data from Table 1, Zhang et al. (2003), Tang et al. (2006) and Yang and Li (2008).

5.3.2. Mantle metasomatism by carbonate melt

To date, Mg isotopic data for mantle carbonatites are still not available. Thus, it is difficult to distinguish where the metasomatic agent (carbonatitic melt) was derived from. However, the observations below suggest that the metasomatic agent (carbonatitic melt) is more likely derived from oceanic crust than from the asthenosphere. First, if the carbonatitic melt from asthenosphere could modify the Mg isotopic composition of the mantle to light values, some of MORBs and OIBs should have light Mg isotopic composition. However, none of MORBs and OIBs has such a light Mg isotopic composition. Second, the geochemical data of the <110 Ma basalts suggest involvement of subducted oceanic crust materials in their mantle source. They are characterized by HIMU features, e.g., relatively higher U/Pb, Th/Pb, Ce/Pb and Nb/U ratios than those of >120 Ma basalts (Figs. 2 and 8). The dehydration of subducted oceanic crust may result in the increases of U/Pb, Th/Pb, Ce/Pb and Nb/U ratios (Hofmann, 1997; Kogiso et al., 1997). Some <110 Ma basalts have U/Pb ratios up to 0.5 and Th/Pb ratios up to 1.8, which are typical characteristics of the HIMU mantle end member (Fig. 8), and may reflect the signature of recycled subducted oceanic crust (Hofmann, 1997). Thus, the source of the <110 Ma basalts may also involve the carbonate in the recycled subducted oceanic crust.

The proportion of dolomite in subducted oceanic crust is not well constrained. Since modern deep water carbonates are dominated by calcite (Plank and Langmuir, 1998; Alt and Teagle, 1999) and dolomite is generally formed at continental shelves, it seems that no dolomite could be brought into the mantle by oceanic crust subduction. However, the subducted oceanic crust may indeed contain a lot of high-Mg carbonates. First, substantial quantities of the terrigenous sediments are known to enter the mantle at subduction zones as suggested by some oceanic island basalts (e.g., Samoa) with low Nd, very high Sr isotopic compositions and other trace element features (Hofmann, 2007; Jackson et al., 2007). Thus, it is possible these recycle terrigenous sediments contain continental shelf dolomite. In addition, as the oceanic crust subducting, low Mg carbonates can be transformed to high-Mg carbonates. The Ca-rich carbonate (e.g., limestone) exchange Ca-Mg with silicates to form dolomite or magnesite (Dasgupta and Hirschmann, 2010 and references therein). During this process, Mg diffuses from silicate minerals into carbonate which may also generate a light Mg isotopic composition in the carbonate. Future experimental studies are required for revealing the Mg isotope fractionation between carbonate and silicate during subduction.

5.4. Implications

5.4.1. Tracing recycled carbonate in the mantle by using magnesium isotopes

Cycling of carbon into and out of the mantle plays a key role in the global carbon cycle and influences the CO_2 budget of the Earth's atmosphere. Subducted oceanic crust contains a significant fraction of carbonate (Alt and Teagle, 1999), and subduction zone dehydration do not significantly remove this carbonate (Yaxley and Green, 1994; Molina and Poli, 2000; Kerrick and Connolly, 2001). These carbonated eclogites advected into the upper mantle would first produce carbonate melts at depths of 280–400 km (Dasgupta et al., 2004), which may then significantly modify chemical and physical properties of the mantle.

Carbon isotopes are efficient to identify recycled organic carbon but not very sensitive to inorganic carbonate. However, the subducted carbon is dominated by inorganic carbonate (Plank and Langmuir, 1998; Alt and Teagle, 1999). Huang et al. (2011) observed light Ca isotopic compositions of Hawaiian basalts with $\delta^{44}Ca_{SRM915a}$ ranging from 0.75‰ to 1.02‰, indicating recycled ancient marine carbonate involved in their mantle source. Thus, Ca isotopes could be a new tracer for Ca and C cycling (Huang et al., 2011).

Similarly, we observed light Mg isotopic compositions of the <110 Ma basalts from the NCC, indicating recycled carbonate in their mantle source. Therefore, Mg isotopes could be another useful tracer for interactions between mantle rocks and recycled carbonate. Since Mg isotopes cannot distinguish organic carbonate ($\delta^{26}Mg = -5.31$ to -1.88‰; Pogge von Strandmann, 2008; Hippler et al., 2009) from inorganic carbonate ($\delta^{26}Mg = -5.09$ to -1.09‰; Young and Galy, 2004; Tipper et al., 2006a; Brenot et al., 2008; Higgins and Schrag, 2010), combined Mg-Ca-C isotopic studies could shed light on tracing recycled carbonate in the mantle.

5.4.2. Constraints on the role of the Pacific subduction in generating continental basalts in the NCC

The influence of western Pacific subduction on the geological evolution of the south China block has been recognized (Li and Li, 2007; Sun et al., 2007; Ling et al., 2009). However, it remains uncertain whether Pacific subduction-related component was contributed to the late-Cretaceous and Cenozoic basalts in the NCC or not. The NCC has experienced three circum-craton subductions, which are manifested by the Paleozoic to Triassic Qinling-Dabie-Sulu ultrahigh-pressure belt in south, the Central Asian Orogenic Belt in the north and the Mesozoic–Cenozoic subduction of Pacific plate in the east (Fig. 1). Although several authors propose that the upper mantle beneath the NCC might have been affected by subduction (Yang and Li, 2008; Zhang et al., 2009; 2010; Xu et al., 2010), it is difficult to tell whether these signatures come from Pacific subduction or not. Magnesium isotopic data of the Mesozoic and Cenozoic basalts could provide spatial and temporal constraints on how the Pacific subducted crust affected the upper mantle beneath the NCC.

Magnesium isotopic data of the Mesozoic and Cenozoic basalts display an abrupt Mg isotopic variation between the >120 Ma basalt and <110 Ma basalt. This is consistent with the abrupt Nd isotopic variation in these basalts (Fig. 9), indicating that the upper mantle beneath the NCC has been significantly modified by melt from the recycled carbonate. Because carbonation lowers the solidus of peridotite (Dasgupta et al., 2007), if carbonated peridotites with light Mg isotopic composition existed in the upper mantle, they would melt first and generate isotopically light basalts. Light Mg isotopic signature in basalts did not appear before 120 Ma, suggesting that the isotopically light mantle source didn't form till 120 Ma. Only the Pacific plate was subducting beneath the NCC during the Mesozoic–Cenozoic, thus, is expected to play an important role in generating the <110 Ma basalts of the NCC.

The <110 Ma basalts investigated here are from both the Eastern Block and Trans-North China Orogen of the NCC (Fig. 1), indicating that the subduction of Pacific plate has affected the Taihang Mountain to the west, which is about 2000 km far from the Japan trench. Based on the seismic study, the Pacific slab is subducting beneath the Japan Islands and Japan Sea and becomes stagnant in the mantle transition zone under East China (Zhao and Ohtani, 2009). The upper mantle above the stagnant Pacific slab under East Asia may have formed a big mantle wedge which exhibits significantly low seismic velocity (Zhao and Ohtani, 2009). Slab melting may take place in the big mantle wedge. As a result, the upper mantle beneath the NCC has been metasomatized by the slab melt and shifts to HIMU geochemical features and light Mg isotopic composition.

6. Conclusions

Based on Mg isotopic compositions of the Mesozoic and Cenozoic basalts and basaltic andesites from the North China craton, we conclude:

(1) The >120 Ma Yixian basalts and basaltic andesites have mantle-like Mg isotopic composition, with $\delta^{26}Mg$ values varying from -0.31 to -0.25‰ and an average of -0.27 ± 0.05‰ (2SD). This indicates that the lithospheric mantle beneath the

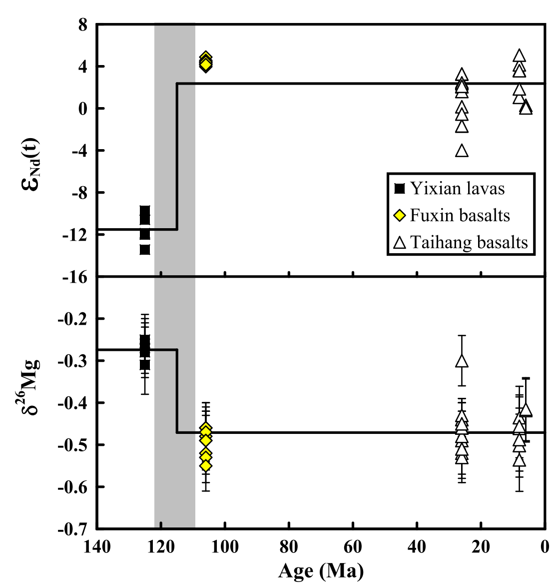

Fig. 9. Plots of $\varepsilon_{Nd}(t)$ and $\delta^{26}Mg$ vs. age for the Yixian lavas, the Fuxin and Taihang basalts. Error bars represent 2SD uncertainties. The grey box indicates the time period when the abrupt change occurred. Data are from Table 1.

NCC have a "normal" mantle-like Mg isotopic composition before 120 Ma.

(2) 29 out of 30 <110 Ma basalts from the NCC have light Mg isotopic composition, with δ^{26}Mg ranging from −0.60 to −0.42‰. The light Mg isotopic composition of the <110 Ma basalts reflects that of their mantle source rather than contamination of crustal materials because they still preserve mantle-like Sr-Nd-Pb isotopic compositions and Ce/Pb, Nb/U ratios.

(3) The light Mg isotopic composition in the upper mantle beneath the North China craton is most likely derived from metasomatism of isotopically light recycled carbonate melt, which is also supported by other geochemical features of the <110 Ma basalts, e.g. high U/Pb and Th/Pb ratios similar to those of HIMU basalts. Our study presents an example to trace recycled carbonate by using Mg isotopes.

(4) The temporal change of the Mg isotopic compositions between >120 Ma and <110 Ma basalts indicates that the isotopically light mantle component could not form till 120 Ma, thus the subducted Pacific Plate might play an important role during the lithospheric thinning in the NCC.

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