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Magnesium isotopic composition of the deep continental crust

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Abstract:

To constrain the behavior of Mg isotopes during deep crustal processes and the Mg isotopic composition of the middle and lower continental crust, 30 composite samples from high-grade metamorphic terranes and 18 granulite xenoliths were investigated. The composites derive from 8 different high-grade metamorphic terranes in the two largest Archean cratons of China, including 13 TTG gneisses, 5 amphibolites, 4 felsic, 4 intermediate and 4 mafic granulites. They have variable bulk compositions with SiO_2 ranging from 45.7 to 72.5%, representative of the middle crust beneath eastern China. The $\delta^{26}Mg$ values of these samples vary from -0.40 to +0.12‰, reflecting heterogeneity of their protoliths, which could involve upper crustal sediments. The granulite xenoliths from the Cenozoic Hannuoba basalts also have a diversity of compositions with MgO ranging from 2.95 to 20.2%. These xenoliths equilibrated under high temperatures of 800–950 ^{o}C, corresponding to depths of the lower continental crust (> 30 km). They yield a large $\delta^{26}Mg$ variation of -0.76 ~ -0.24‰. The light Mg isotopic compositions likely result from interactions with isotopically light metamorphic fluids, probably carbonate fluids. Together with previously reported data, the average $\delta^{26}Mg$ of the middle and lower continental crusts is estimated to be -0.21 ± 0.07‰ and -0.26 ± 0.06‰, respectively. The bulk continental crust is estimated to have an average $\delta^{26}Mg$ of -0.24 ± 0.07‰, which is similar to the average of the mantle. The large Mg isotopic variation in the continental crust reflects the combination of several processes, such as continental weathering, involvement of supracrustal materials in the deep crust, and fluid metasomatism.

Keywords: magnesium isotope, deep continental crust, high-grade metamorphic terrane, granulite xenolith

Introduction

Magnesium is a fluid-mobile, major element, and has three isotopes of ^{24}Mg, ^{25}Mg and ^{26}Mg. 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 (Young and Galy, 2004; Tipper et al., 2006a; 2006b; 2010; Pogge von Strandmann et al., 2008a; 2008b; Li et al., 2010; Teng et al., 2010b; Huang et al., 2012; Liu et al., 2014). The mantle, upper continental crust and the hydrosphere have distinct Mg isotopic compositions. The mantle is nearly homogeneous with $\delta^{26}Mg$ values ranging from -0.48 to -0.06 ‰ (Teng et al., 2007; 2010a; Handler et al., 2009; Yang et al., 2009; Bourdon et al., 2010; Dauphas et al., 2010; Pogge von Strandmann et al., 2011; Xiao et al., 2013), whereas the upper continental crust is highly heterogeneous ($\delta^{26}Mg$ = -1.64 ~ +0.92‰) and on average heavier than the mantle (Shen et al., 2009; Li et al., 2010; Liu et al., 2010; Huang et al., 2013a; Teng et al., 2013). The hydrosphere has a very light Mg isotopic composition, as represented by seawater ($\delta^{26}Mg$ = -0.83 ± 0.09‰, 2SD) (Foster et al., 2010; Ling et al., 2011 and references therein) and the flux weighted average of major rivers ($\delta^{26}Mg$ = -1.09‰) (Tipper et al., 2006b). These Mg isotopic characteristics are considered to result from continental weathering, during which light Mg isotopes are preferentially partitioned into the hydrosphere, causing a shift in the weathered residues toward a heavier isotopic composition (Pogge von Strandmann et al., 2008b; Teng et al., 2010b; Tipper et al., 2010; Huang et al., 2012; Liu et al., 2014).

To better constrain the interaction between the crust and the hydrosphere, Mg isotopic composition of the middle and lower continental crustal materials should also be investigated since they contain large proportions of Mg in the crust. However, thus far, only one study on this issue has been reported. Teng et al. (2013) investigated two well-characterized suites of lower-crustal granulite xenoliths from the Chudleigh and McBride volcanic provinces, North Queensland, Australia. The McBride granulites display a very large variation in δ^{26}Mg values from -0.72 to +0.19‰, which was considered to reflect both source heterogeneity and metamorphic enrichment of garnet. Nonetheless, the Mg isotopic composition of the middle continental crust is still unknown since granulite xenoliths are generally considered to be representative of the lower crust (Rudnick and Gao, 2003). In addition, the δ^{26}Mg variation (-0.72 to +0.19‰) observed in the McBride lower crustal granulites is quite large. Whether it is a special case or a common phenomenon in the lower crust requires further research.

To constrain the behavior of Mg isotopes during deep crustal processes and the Mg isotopic compositions of the middle and lower continental crusts, two suits of samples from China have been investigated. One is a set of high-grade metamorphic rocks from Archean terranes, and the other are granulite xenoliths from Damaping, Hannuoba. Both suites have been systematically studied and are considered as representative samples for the middle and lower continental crusts of eastern China (Gao et al., 1998a; Liu et al., 2001; 2004; Teng et al., 2008). Our results reveal a large Mg isotopic variation in the deep continental crust, which likely results from the combination of several processes, such as continental weathering, involvement of supracrustal materials in the deep crust, and fluid metasomatism. Nonetheless, the bulk continental crust on average still has a mantle-like Mg isotopic composition.

Samples and geological background

The deep continental crust can be divided into two layers based on seismological studies: the middle crust and the lower crust (Rudnick and Fountain, 1995). Two types of samples can be used to determine the composition of the deep continental crust: high-grade metamorphic terranes and lower crustal xenoliths. The former is often considered to be representative of the middle crust (Bohlen and Mezger, 1989) and the latter to be representative of the lower crust (Rudnick and Gao, 2003). Thirty samples from the high-grade metamorphic terrane and 18 granulite xenoliths from Eastern China are studied here. The geological background, sample description, major and trace-element abundances and Sr, Nd, Pb and Li isotopic compositions of the studied samples have been previously reported (Gao et al., 1998a; Liu et al., 2001; 2004; Teng et al., 2008). Only a brief summary is given below.

High-grade metamorphic rocks from Archean terranes

The samples were collected from 8 different high-grade metamorphic terranes in the two largest Archean cratons of China (Fig. 1). The Kongling amphibolite-granulite-facies terrane is from the Yangtze Craton, and the other 7 terranes are from the North China craton, including Wutai and Dengfeng amphibolite-facies terranes, Fuping, Hengshan and Taihua amphibolite-granulite-facies terranes, Jinning and Wulashan granulite-facies terranes (Gao et al., 1999; Qiu et al., 2000). The samples, including 13 TTG gneisses, 5 amphibolites, 4 felsic, 4 intermediate and 4 mafic granulites, are composites that were produced by mixing equal amounts of individual rock samples (n=1 to 15) having the same age and lithology from the same tectonic unit. Sm-Nd and zircon U-Pb dating of these samples yielded 3.3 – 2.5 Ga (Gao et al., 1998a). The individual rock samples were collected along road cuts, riverbanks, or mountain valleys and are very fresh, as indicated by petrographic studies (Gao et al., 1996; 1998a; 1999; Qiu et al., 2000). These composites are thus considered to be representative of most Archean units exposed in eastern China.

The bulk compositions of these composites vary from mafic to felsic, with SiO_2 ranging from 45.7 to 72.5% and MgO from 0.4 to 7.7% (Fig. 2a). They are considered to be representative of the middle crust beneath eastern China (Gao et al., 1998a; 1998b). They have relatively restricted Li isotopic composition with δ^7Li ranging from +1.7 to +7.5‰ (Teng et al., 2008) (Fig. 2b).

Granulite xenoliths from Damaping, Hannuoba, China

The lower crustal xenoliths were collected from the Cenozoic Hannuoba basalts (Zhang et al., 2013), which is situated in the central orogenic belt of the North China

Craton (Fig. 1). These xenoliths have a diversity of compositions, with SiO_2 ranging from 44.2 to 60.3% and MgO from 2.95 to 20.2% (Gao et al., 2000; Chen et al., 2001; Liu et al., 2001; 2004; Zhou et al., 2002). They are 4 to 20 cm in diameter and range in composition from pyroxenite, plagioclase-rich mafic granulite to intermediate granulite. All these xenoliths equilibrated under high temperatures (800 – 950 ^{o}C), corresponding to depths greater than 30 km (Chen et al., 2001; Liu et al., 2003) (Fig. 3). U-Pb zircon chronology on these granulite xenoliths indicates that basaltic magma intruded Precambrian lower crust at ~160–140 Ma and induced subsequent granulite-facies metamorphism (Liu et al., 2004).

The samples can be divided into two groups based on MgO contents: high Mg xenoliths and low Mg xenoliths. The high Mg xenoliths include pyroxenites, two-pyroxene mafic granulites and garnet-bearing mafic granulites, with MgO ranging from 12.4 to 20.2%, while the low Mg xenoliths include plagioclase-rich mafic granulites and intermediate granulites, with MgO ranging from 2.95 to 6.97% (Fig. 2a). Both types have variable Li isotopic compositions, with δ^7Li of -9.6 ~ +4.3‰ and -5.1 ~ +13.8‰, respectively (Teng et al., 2008) (Fig. 2b). Such large variations of Li isotopic compositions were considered to mainly result from source heterogeneity (Teng et al., 2008). These xenoliths also show very large variations in Sr (^{87}Sr/^{86}Sr = 0.707 to 0.723), Nd (ε_{Nd} = -28.0 to -11.3) and Pb isotopic compositions (^{206}Pb/^{204}Pb = 16.16 to 17.91), probably reflecting mixing between preexisting Precambrian deep crust with the underplated basaltic magmas (Liu et al., 2001; 2004).

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; Li et al., 2010; Yang et al., 2009; Teng and Yang, 2014). Only a brief description is given below.

All chemical procedures were carried out in a clean laboratory environment.

Depending on Mg concentration, one to 25 mg of sample powder was weighted in Savillex screw-top beakers in order to have > 50 μg Mg in the solution. The sample powder was dissolved in a mixture of concentrated HF-HNO_3-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 (Multi-Collector Inductively Coupled Plasma Mass Spectrometry) at the University of Arkansas (Teng and Yang, 2014). Magnesium isotope data are reported in standard δ-notation relative to DSM3: $\delta^{26}Mg = [(\delta^{26}Mg/^{24}Mg)_{sample} / (\delta^{26}Mg/^{24}Mg)_{DSM3} - 1] \times 1000$. $\Delta^{25}Mg' = \delta^{25}Mg' - 0.521 \times \delta^{26}Mg'$, where $\delta^{25,\,26}Mg' = 1000 \times \ln[(\delta^{25,\,26}Mg + 1000)/1000]$ (Young and Galy, 2004).

The internal precision of the measured $^{26}Mg/^{24}Mg$ ratio based on ≥ 4 repeat runs of same sample solution during a single analytical session is < ± 0.07‰ (Teng et al., 2010a). Six replicate analyses of olivine KH-1 (Kilbourne Hole) yielded $\delta^{26}Mg$ values of -0.27 ± 0.05%, which is in agreement with that reported by Teng et al. (2015) ($\delta^{26}Mg$ = -0.27 ± 0.07%; 2SD, n=16). The external precision, as shown by replicate analyses of synthetic solution, mineral and rock standards, was ±0.06% for $\delta^{25}Mg$ and ±0.07% for $\delta^{26}Mg$ (2SD) (Teng et al., 2010a; 2015).

Results

Magnesium isotopic compositions of composite samples from high-grade metamorphic terranes are listed in Table 1 and granulite xenoliths in Table 2, along with their chemical compositions. In a plot of $\delta^{25}Mg'$ vs. $\delta^{26}Mg'$ (Fig. 4), all samples fall along the terrestrial equilibrium mass fractionation curve, with a slope of 0.521 (Young and Galy, 2004), with $\Delta^{25}Mg'$ values < ± 0.04‰ (Tables 1 and 2).

Overall, Mg isotopic compositions vary significantly, with $\delta^{26}Mg$ ranging from -0.76 to +0.12‰ and $\delta^{25}Mg$ from -0.39 to +0.06‰ (Tables 1 and 2). This variation is quite similar to that of the lower crustal granulite xenoliths from Chudleigh and McBride, Australia, with δ^{26}Mg from -0.72 to +0.19‰ (Teng et al., 2013), and falls within the range of the upper continental crust (δ^{26}Mg = -1.64 ~ +0.92‰) (Li et al., 2010; Liu et al., 2010; Huang et al., 2013a).

The samples from Archean high-grade metamorphic terranes have a large variation in Mg isotopic composition (Fig. 5), with δ^{26}Mg ranging from -0.40 to +0.12‰ and an average of -0.22 ± 0.19‰ (2SD, n=30). Two samples (14R110, +0.12 ± 0.06‰ and D148, -0.05 ± 0.09‰) have δ^{26}Mg values deviating the population of the others (-0.40 ~ -0.15‰, Fig. 6a). Samples from different localities do not display systematical variations (Fig. 6b).

The high and low Mg granulite xenoliths from Hannuoba exhibit quite distinct Mg isotopic compositions (Fig. 5). The high Mg granulite xenoliths show a limited variation of 0.11‰ in δ^{26}Mg value from -0.37 to -0.26‰. However, the low Mg granulite xenoliths display an obviously lighter Mg isotopic composition, with large variations in δ^{26}Mg ranging from -0.76 to -0.24‰. Such a light Mg isotopic composition of low Mg granulite xenoliths is comparable with the McBride granulites, which have δ^{26}Mg values of -0.72 to +0.19‰ (Teng et al., 2013).

Discussion

The high-grade metamorphic terranes and granulite xenoliths from eastern China, which are considered to be representative of the middle and lower continental crust (Gao et al., 1998a; Liu et al., 2001; 2004; Teng et al., 2008), display up to 0.9‰ variation in Mg isotopic composition. Based on studies of peridotites, oceanic basalts and granites (Teng et al., 2007; 2010a; Handler et al., 2009; Yang et al., 2009; Bourdon et al., 2010; Liu et al., 2010), no more than 0.07% Mg isotope fractionation will occur during closed-system partial melting of the mantle and fractional crystallization of basaltic or granitic magma. Therefore, the large variation in the Mg isotopic composition of the deep crustal rocks must be due to source heterogeneity and/or other processes. Below, we first evaluate each of these mechanisms for both terrane and xenolith samples, and then estimate the average Mg composition of the continental crust.

Magnesium isotopic systematics of the Archean high-grade metamorphic terranes from the eastern China

The 30 samples from Archean high-grade metamorphic terranes yield a δ^{26}Mg variation ranging from -0.40 to +0.12‰, among which 28 samples fall within the range of the mantle rocks (-0.48 to -0.06 ‰) (Teng et al., 2007; 2010a; Handler et al., 2009; Yang et al., 2009; Bourdon et al., 2010; Dauphas et al., 2010; Bizzarro et al., 2011; Pogge von Strandmann et al., 2011). These samples also have a mantle-like Li isotopic composition with δ^{7}Li ranging from +1.7 to +7.5‰ (Teng et al., 2008). The mantle-like Li and Mg isotopic compositions of the Archean high-grade metamorphic terranes indicate that they inherited Li and Mg isotopic compositions of their protoliths, which have not been modified by subsequent processes.

The other two samples (14R110 and D148) yield relatively high δ^{26}Mg values of $+0.12 \pm 0.06$‰ and -0.05 ± 0.09‰, respectively (Fig. 5). Three potential mechanisms could result in the heavy Mg isotopic compositions: (1) surface weathering; (2) metamorphism; and/or (3) protolith heterogeneity.

Surface weathering can significantly modify Mg isotopic compositions of rocks towards heavy values (Pogge von Strandmann et al., 2008a; Teng et al., 2010b; Huang et al., 2012; Liu et al., 2014). However, the high-grade metamorphic samples studied here were carefully collected and are very fresh as indicated by petrographic studies (Gao et al., 1998a). Therefore, the effects of weathering are considered to be negligible. This is also supported by the Li isotopic composition of these samples. Surface weathering can significantly fractionate Li isotopes and result in large variations in both Li isotopic composition and Li concentration in the weathered products (Teng et al., 2004). However, all these high-grade metamorphic samples have similar and mantle-like Li isotopic compositions (Teng et al., 2008).

The relatively heavy Mg isotopic composition of these two samples may thus reflect either metamorphism or protolith heterogeneity. Most progressive metamorphic reactions are accompanied by dehydration, leading to the depletion of fluid-mobile elements in the high-grade metamorphic rocks (Rudnick et al., 1985). Lithium and Mg both are fluid mobile. During dehydration, ^{7}Li and ^{24}Mg preferentially enters fluids over most minerals, leading to lower δ^7Li and higher δ^{26}Mg in rocks. The extent of isotope fractionation is determined by the fraction of Li and Mg released from rocks into fluids and the fractionation factor ($\alpha_{fluid\text{-}rock}$). Theoretically, this process may not affect Mg isotopic systematics. Since Mg contents in the silicate rocks are at least 10-100 times higher than those in the fluid (Brenot et al., 2008), Mg isotopic composition of rocks is expected to change little with such a small loss of Mg. This was demonstrated by previous studies on eclogites, granulites and metapelites (Li et al., 2011; 2014; Teng et al., 2013; Wang et al., 2014b). Though significant (up to 3%) amount of fluid was lost during metamorphic dehydration, eclogites, granulites and metapelites still have similar Mg isotopic composition to their protoliths, suggesting limited Mg isotope fractionation during metamorphic dehydration (Li et al., 2011; 2014; Teng et al., 2013; Wang et al., 2014b).

Therefore, the most likely mechanism for the heavy δ^{26}Mg values of these two samples (14R110 and D148) is the involvement of upper crustal sediments in their protoliths. Sedimentary rocks have highly heterogeneous and, in most cases, heavy Mg isotopic compositions, with δ^{26}Mg ranging from -0.52 to +0.92‰ (Li et al., 2010). These two samples also have evolved compositions with SiO_2 = 70.84 and 71.84%. However, other samples (D139, D140, D142 and D143) with even higher SiO_2 contents (72.03 – 72.54%) do not exhibit high δ^{26}Mg values (-0.24 – -0.35%). Possible explanation is that the sedimentary materials have a very large range of Mg isotopic composition (δ^{26}Mg = -0.52 to +0.92‰) and only a part of them has heavy Mg isotopic composition (δ^{26}Mg > 0‰). Nevertheless, the majority of the terrane samples have a mantle-like Mg isotopic composition (δ^{26}Mg = -0.40 to -0.15‰), and samples from different localities do not display systematic variation (Fig. 6b), indicating that the Mg isotopic compositions of the middle crust beneath both the North China craton and the South China craton are quite similar.

Magnesium isotopic systematics of the Hannuoba granulite xenoliths from the Eastern China

The high and low Mg granulite xenoliths from Hannuoba exhibit quite distinct Mg isotopic compositions (Fig. 5). The high Mg granulite xenoliths falls within the mantle range (δ^{26}Mg = -0.48 to -0.06) (Teng et al., 2007; 2010a; Handler et al., 2009; Yang et al., 2009; Bourdon et al., 2010; Dauphas et al., 2010; Bizzarro et al., 2011; Pogge von Strandmann et al., 2011; Xiao et al., 2013), indicating an inheritance of Mg isotopic compositions of their protoliths, because their high MgO contents make it difficult to modify by other processes.

However, the low Mg granulite xenoliths display an obviously lighter Mg isotopic composition with large variations in δ^{26}Mg ranging from -0.76 to -0.24‰. Since Mg isotope fractionation during the progressive metamorphic reactions is limited (Li et al., 2011; 2014; Teng et al., 2013; Wang et al., 2014b), the light Mg isotopic composition of the Hannuoba granulite xenoliths may result from one or a combination of the following processes: (1) unrepresentative sampling, (2) protolith heterogeneity, (3) interaction with host magma, (4) interaction with fluid (fluid metasomatism).

For small-size xenoliths with coarse-grained minerals, a mineralogically unrepresentative sampling could result in bias in the Mg isotopic composition of the whole rock (Teng et al., 2013). For example, garnet is isotopically lighter than clinopyroxene in eclogite (Li et al., 2011; Wang et al., 2012; 2014a; 2014b), and its preferential enrichment could result in light isotopic composition of the whole rock (Teng et al., 2013). The McBride xenolith (83-162), which contains an overabundance of metamorphic garnet, has a very low $\delta^{26}Mg$ value of -0.72‰ (Teng et al., 2013). The light Mg isotopic composition of the low Mg granulite xenoliths in Hannuoba could not result from unrepresentative sampling, because these samples are garnet absent (Table 3). The only two garnet-bearing mafic granulites (DMP-08 and DMP-15) are high Mg granulite xenoliths. They have low Ni/Ho (308 ~ 309) and Cr/Ho (438 ~ 538) ratios (Fig. 7) relative to the other high Mg granulites, probably indicating preferential sampling of garnet (Teng et al., 2013). However, their Ni/Ho and Cr/Ho ratios are obviously higher than those of the two unrepresentative samples reported by Teng et al. (2013), which have extremely low Ni/Ho (< 60) and Cr/Ho (< 60) ratios (Fig. 7). In addition, their $\delta^{26}Mg$ values (-0.37‰ and -0.31‰) are still in the mantle range. Thus, the unrepresentative sampling does not play a significant role in controlling Mg isotopic variation in the Hannuoba xenoliths.

Another mechanism to generate the large Mg isotopic variation in the Hannuoba granulite xenoliths is protolith heterogeneity. The high Mg and low Mg granulites display differences not only in Mg isotopic composition, but also in Nd, Pb and Li isotopic compositions (Fig. 8 and Fig. 9). The large variations in Nd and Pb isotopic compositions are considered to reflect mixing between preexisting Precambrian deep crust with the underplated basalts (Liu et al., 2001; 2004). In order to produce the mixing trends observed in $\delta^{26}Mg$ vs. $^{206}Pb/^{204}Pb$ (Fig. 8a) and $\delta^{26}Mg$ vs. $\varepsilon_{Nd}(t)$ plots (Fig. 8b), the preexisting Precambrian deep crust with low $^{206}Pb/^{204}Pb$ and $\varepsilon_{Nd}(t)$ should have a mantle-like Mg isotopic composition, whereas the underplated basalt should have a light Mg isotopic composition ($\delta^{26}Mg$ < -0.76‰). However, such a light value (<-0.76‰) has not been observed in the Mesozoic and Cenozoic basalts from the North China craton, which have $\delta^{26}Mg$ of -0.60 ~ -0.42‰ (Yang et al., 2012). In addition, the low Mg concentrations (2.95 to 6.97%) of these samples indicate that the involvement of underplated basalts, if any, is limited. Hence, the protolith heterogeneity resulted from mixing with underplated basalts may not play a major role in lowering the $\delta^{26}Mg$ value of the xenoliths, and additional processes are required to produce the light Mg isotopic composition.

The third possibility to generate light Mg isotopic composition of the xenoliths is through interactions with host magmas. During ascent to the Earth's surface, granulite xenoliths and host magmas are generally not in thermal and chemical equilibrium. This process can result in interaction between xenoliths and host magmas. To generate a light Mg isotopic composition observed in the Hannuoba granulite xenoliths, the host magma should have an even lighter Mg isotopic composition ($\delta^{26}Mg < -0.76‰$). It also seems impossible that host magmas could have such light Mg isotopic composition, thus, interaction with host magmas may also not play a major role in lowering the $\delta^{26}Mg$ value of the xenoliths.

The most likely mechanism to produce such a light Mg isotopic composition in the Hannuoba granulite xenoliths is rock-fluid interaction during metamorphism. Since ^{7}Li and ^{24}Mg preferentially enters fluids, fluids could have light Mg isotopic composition and heavy Li isotopic composition. Interaction with fluids should generate a negative correlation between $\delta^{7}Li$ and $\delta^{26}Mg$ in rocks, which has been observed in the Hannuoba granulite xenoliths (Fig.9). In addition, Mg isotopic compositions correlate with Rb, Ba and H_2O contents (Fig. 10), suggesting that the granulites have been interacted with metamorphic fluids to various extents. Although dehydration process can not affect Mg isotopic systematics in silicate rocks (Li et al., 2011; 2014; Teng et al., 2013; Wang et al., 2014b), fluid metasomatism probably can, especially for low Mg rocks. This is because Mg loss during dehydration represents only a very small portion of bulk Mg in silicate rocks (Li et al., 2011; 2014; Teng et al., 2013; Wang et al., 2014b), however, Mg add-in during fluid metasomatism could be very large especially in an open system. This is consistent with the observation that the low Mg granulites were modified more significantly in Mg isotopic composition than the high Mg granulites (Fig. 5). It is difficult to do a mass balance calculation of the rock-fluid reaction, because there is no Mg concentration and isotopic data of deep fluids reported. The seawater contains 0.12% (53000 μmol/L) Mg (Tipper et al., 2006a) with a δ^{26}Mg value of 0.832 ± 0.068‰ (Ling et al., 2011). The river water displays large variations of δ^{26}Mg (-2.08 ~ -0.05‰) (Tipper et al., 2006a; 2006b; 2008). Deep fluids (especially carbonate fluids) can contain higher Mg than the seawater, e.g. fluid inclusions can have up to 9% Mg (McCaig et al., 2000). Assuming a pure carbonate fluid with ~10% of Mg and -2‰ of δ^{26}Mg, a fluid/rock ratio of 0.05 is required to produce the light Mg isotopic composition in the Hannuoba granulite xenoliths. This metasomatism by carbonate fluids may lead to formation of carbonate minerals. However, no carbonate mineral is observed in the Hannuoba granulite xenolith. Possible mechanism is that under T-P condition of 1 Gpa and 800-950 ^{o}C, carbonate-bearing granulites will experience complete decarbonation (Fig. 3). The decomposition of carbonate mineral released CO_2, but remained light Mg isotopes in the rocks.

To summarize, all of the above processes could potentially lower δ^{26}Mg values of granulite xenoliths to various extent. Nonetheless, the major mechanism to produce such a light Mg isotopic composition in the Hannuoba granulite xenoliths is rock-fluid interaction.

Magnesium isotopic composition of the deep and bulk continental crust

Magnesium content and isotopic compositions of the crustal samples are highly variable (Fig. 11), thus we estimate the average Mg isotopic composition of the middle and lower crusts from eastern China by using the concentration-weighted δ^{26}Mg for terrane and granulite samples, respectively. The δ^{26}Mg of the middle crust is estimated to be -0.21 ± 0.07‰, which is slightly higher than the average δ^{26}Mg of the mantle (-0.25 ± 0.07‰) (Teng et al., 2010a).

The weighted average δ^{26}Mg value of the lower continental crust underneath the Hannuoba, eastern China is estimated to be -0.36 ± 0.05‰, which is significantly lighter than that of Chudleigh and McBride, Australia (-0.18‰) (Teng et al., 2013), indicating distinct Mg isotopic compositions of the lower crust in different continents. Nevertheless, based on all available data, the average δ^{26}Mg of the lower continental crust is estimated to be -0.26 ± 0.06‰.

The average δ^{26}Mg of the bulk continental crust is estimated to be -0.24 ± 0.07‰ by combining the average δ^{26}Mg value and Mg concentration of the upper crust (-0.22 ± 0.10‰, 2.48 wt %), middle crust (-0.21 ± 0.07‰, 3.59 wt %), and lower crust (-0.26 ± 0.06‰, 7.24 wt %), with their respective weight proportions of 0.337: 0.347: 0.317 (Huang et al., 2013b; Teng et al., 2013). The overall Mg isotopic composition of the continental crust is similar to the average δ^{26}Mg of the mantle (-0.25 ± 0.07‰) (Teng et al., 2010a). The large Mg isotopic variation in the continental crust results from the combination of several processes, such as continental weathering, involvement of supracrustal materials in the deep crust, and fluid metasomatism. Because all these processes can only significantly modify Mg isotopic compositions of rocks with low MgO contents and fractionate Mg isotopes in opponent ways, the bulk continental crust still remains a mantle-like Mg isotopic composition.

Implications

Magnesium isotopic composition of the continental crust can provide not only important constraints on the behavior of Mg isotopes during deep crustal processes, but also necessary parameters for the global Mg isotopic mass-balance calculation. Our studies of high-grade metamorphic terrane samples and the Hannuoba granulite xenoliths from eastern China, as well as previous studies on granulite xenoliths from Queensland, Australia (Teng et al., 2013), reveal large Mg isotopic variation in the deep crust (δ^{26}Mg = -0.76 ~ +0.19‰), indicating that light Mg isotopic composition could be a common phenomenon in the lower continental crust. In addition, the deep continental crust beneath the eastern China was previously considered to be homogeneous in Mg isotopic composition because granites derived from partial melting of the deep continental crust from this region have a very small Mg isotopic variation with δ^{26}Mg ranging from -0.35 to -0.14‰ (Li et al., 2010; Liu et al., 2010). Our study suggests that although partial melting and granite differentiation do not fractionate Mg isotopes, these processes may homogenize Mg isotopic composition of the source rocks, and thus erase the detail information of the deep continental crust.

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Table 1

Magnesium isotopic compositions of samples from high-grade metamorphic terranes in Eastern China

Sample	Terrane	n[a]	SiO_2[b]	MgO[b]	CIA[b]	δ^7Li[c]	$\delta^{26}Mg$	2SD	$\delta^{25}Mg$	2SD	$\Delta^{25}Mg'$
Tonalite gneisses											
D138	Dengfeng	1	62.82	2.62	47	2.8	-0.40	0.06	-0.20	0.04	0.00
D141	Taihau	4	61.38	3.16	41	3.2	-0.35	0.06	-0.16	0.04	0.02
14R109	Wutai	10	62.67	2.63	50	5.2	-0.22	0.06	-0.11	0.04	0.00
14R110	Wutai	10	70.84	1.89	52	2.6	0.12	0.06	0.06	0.04	0.00
14R118	Wutai	15	61.03	2.89	47	5.6	-0.31	0.06	-0.13	0.04	0.03
14R117	Wutai	15	61.34	2.58	48	5.2	-0.23	0.06	-0.13	0.04	-0.01
Trondhjemite gneisses											
14R116	Wutai	15	69.32	1.04	51	7.5	-0.30	0.06	-0.17	0.04	-0.01
D142	Taihau	1	72.54	0.40	51	3.9	-0.29	0.09	-0.17	0.07	-0.02
D139	Dengfeng	3	72.39	0.60	54	3.7	-0.31	0.09	-0.18	0.07	-0.02
D147	Kongling	7	67.60	2.08	49	2.9	-0.24	0.09	-0.13	0.07	-0.01
Granite gneisses											
D143	Taihau	3	72.03	0.58	50	4.0	-0.35	0.09	-0.18	0.07	-0.01
D140	Dengfeng	2	72.32	0.46	51	3.8	-0.24	0.09	-0.11	0.07	0.01
D148	Kongling	4	71.84	1.31	53	2.7	-0.05	0.09	-0.02	0.07	0.00
Amphibolites											
D149	Dengfeng	10	51.41	6.09	41	3.4	-0.20	0.09	-0.10	0.07	0.00
D153	Taihau	12	50.81	5.76	39	6.8	-0.25	0.09	-0.09	0.07	0.04
14R162	Fuping	2	47.05	7.73	38	3.9	-0.17	0.09	-0.07	0.07	0.02
14R167	Hengshan	8	50.71	5.71	40	3.7	-0.22	0.07	-0.14	0.05	-0.03
D171	Kongling	8	49.54	6.84	38	6.2	-0.26	0.07	-0.11	0.05	0.02
Mafic granulites											
D154	Taihau	2	50.08	5.91	38	4.4	-0.19	0.07	-0.09	0.05	0.01
14R161	Fuping	8	50.71	5.17	35	4.7	-0.16	0.07	-0.06	0.05	0.02
14R168	Hengshan	1	48.86	4.51	33	5.7	-0.20	0.07	-0.09	0.05	0.02
D368	Jinning	10	45.69	6.54	38	3.4	-0.16	0.07	-0.08	0.05	0.01
Intermediate granulites											
15R281	Wulashan	10	54.54	4.52	41	2.3	-0.21	0.07	-0.08	0.05	0.03
15R267	Wulashan	10	61.60	3.54	45	5.1	-0.22	0.07	-0.12	0.05	0.00
15R278	Wulashan	10	62.33	2.38	45	3.2	-0.21	0.07	-0.11	0.05	0.00
15R266	Wulashan	10	62.76	1.55	47	2.4	-0.17	0.07	-0.07	0.04	0.02
Felsic granulites											
D366	Jinning	3	59.00	3.07	47	2.8	-0.19	0.07	-0.11	0.04	-0.01
15R277	Wulashan	10	65.27	2.11	47	1.7	-0.19	0.07	-0.10	0.04	0.00
15R268	Wulashan	10	65.53	1.69	47	3.1	-0.15	0.07	-0.08	0.04	-0.01
15R263	Wulashan	10	70.36	0.66	50	2.8	-0.17	0.07	-0.12	0.04	-0.03

[a] n = number of individual samples comprising the composite.

[b] Data from Gao et al. (1998a), CIA = Chemical Indexes of Alteration, defined as = molecular ratios of $Al_2O_3/(Al_2O_3 + CaO + Na_2O + K_2O) \times 100$ (Nesbitt and Young, 1982)..

[c] Data from Teng et al. (2008).

Table 2

Magnesium isotopic compositions of the Hannuoba granulite xenoliths

Sample	SiO_2[a]	MgO[a]	Mg#[a]	δ^7Li[b]	δ^{26}Mg	2SD	δ^{25}Mg	2SD	Δ^{25}Mg'
Pyroxenite									
DMP-10	48.99	18.62	76	-4.2	-0.30	0.07	-0.18	0.04	-0.03
Two-pyroxene mafic granulites									
DMP-03	49.07	14.01	79	0.5	-0.35	0.07	-0.16	0.04	0.02
DMP-09	49.54	18.37	78	-1.9	-0.35	0.07	-0.20	0.04	-0.02
DMP-11	49.83	20.18	77	-1.9	-0.28	0.07	-0.15	0.04	0.00
DMP-28	50.30	13.85	70	-8.0	-0.26	0.03	-0.11	0.03	0.02
DMP-45	50.66	15.06	74	-9.6	-0.33	0.03	-0.14	0.03	0.02
DMP-66	50.51	13.58	78	-2.9	-0.30	0.03	-0.15	0.03	0.00
DMP-68	49.74	16.23	78	-3.3	-0.27	0.03	-0.11	0.03	0.03
Garnet -bearing mafic granulites									
DMP-08	45.27	12.42	72	0.2	-0.37	0.03	-0.18	0.03	0.01
DMP-15	44.21	14.69	70	4.3	-0.31	0.03	-0.16	0.03	0.00
Plagioclase-rich mafic granulites									
DMP-06	51.72	6.97	64	13.8	-0.65	0.03	-0.34	0.03	-0.01
DMP-07	45.71	5.07	61	2.2	-0.67	0.07	-0.34	0.05	0.00
DMP-62	52.92	5.81	72	6.7	-0.74	0.07	-0.38	0.05	0.00
DMP-75	52.43	5.18	56	1.3	-0.42	0.07	-0.21	0.05	0.00
Intermediate granulites									
DMP-01	60.30	4.96	72	12.1	-0.44	0.07	-0.23	0.05	-0.01
DMP-27	54.00	3.74	47	-5.1	-0.24	0.07	-0.12	0.05	0.00
DMP-61	57.48	2.95	70	3.8	-0.76	0.07	-0.39	0.05	0.00
DMP-70	56.82	5.34	65	7.1	-0.56	0.07	-0.29	0.05	-0.01

[a] Data from Liu et al. (2001).

[b] Data from Teng et al. (2008).

Figure captions:

Figure 1. Cratons of eastern China with composite sampling area (dashed line box) and the Hannuoba granulite xenoliths (black circle) outlined. Numbers 1 to 8 correspond to the eight Archean terranes.

Figure 2. (a) MgO vs. SiO_2, (b) δ^7Li vs. SiO_2 for the high-grade metamorphic terrane composites and the Hannuoba granulite xenoliths. Database: MgO and SiO_2 for the high-grade metamorphic terrane composites and the Hannuoba granulite xenoliths are from Gao et al. (1998a) and Liu et al. (2001), respectively. Lithium isotopic data are from Teng et al. (2008).

Figure 3. P-T estimates for the Hannuoba granulite xenoliths and experimentally constrained equilibria of decarbonation of carbonated basalts (modified from (Dasgupta, 2013). Under the P-T condition of the Hannuoba granulite xenoliths, they will experience complete decarbonation, which is consistent with the observation that carbonate mineral is absent in these rocks.

Figure 4. Magnesium three-isotope plot of the samples in this study. The solid line represents the equilibrium mass fractionation line with a slope equal to 0.521. Error bars represent 2SD uncertainties. Data are from Tables 1 and 2.

Figure 5. δ^{26}Mg vs. MgO (wt%) for the high-grade metamorphic terrane composites and the Hannuoba granulite xenoliths. The grey bar represents the average mantle (δ^{26}Mg = -0.25 ± 0.07) (Teng et al., 2010a). Error bars represent 2SD uncertainties. Data are from Tables 1 and 2.

Figure 6. (a) δ^{26}Mg vs. rock type, (b) δ^{26}Mg vs. sample locality for the high-grade metamorphic terrane composites. T = Tonalite gneiss, TR = Trondhjemite gneiss, G = Granite gneiss, A = Amphibolite, M = Mafic granulite, I = Intermediate granulite, F = Felsic granulite. The grey bar represents the average mantle (δ^{26}Mg = -0.25 ± 0.07) (Teng et al., 2010a). Error bars represent 2SD

uncertainties. Data are from Table 1.

Figure 7. (a) δ^{26}Mg vs. Cr/Ho, (b) δ^{26}Mg vs. Ni/Ho for the Hannuoba high Mg granulite xenoliths. Database: Cr/Ho and Ni/Ho are from Liu et al. (2001), δ^{26}Mg from Table 2, the two samples with preferential sampling of garnet from Teng et al. (2013). Error bars represent 2SD uncertainties.

Figure 8. (a) δ^{26}Mg vs. ^{206}Pb/^{204}Pb, (b) δ^{26}Mg vs. ε_{Nd}(t) for the Hannuoba granulite xenoliths. Database: ^{206}Pb/^{204}Pb and ε_{Nd}(t) from Liu et al. (2004) and δ^{26}Mg from Table 3. Error bars represent 2SD uncertainties. The grey bar represents mixing between preexisting Precambrian deep crust with the underplated basalts. If the mixing is the major mechanism to generate the large Mg isotopic variation in the Hannuoba granulite xenoliths, the underplated basalt should have a very light Mg isotopic composition (δ^{26}Mg < -0.76‰).

Figure 9. δ^{26}Mg vs. δ^{7}Li for the high-grade metamorphic terrane composites and the Hannuoba granulite xenoliths. Database: δ^{7}Li from Teng et al. (2008) and δ^{26}Mg from Tables 1 and 2. Error bars represent 2SD uncertainties.

Figure 10. (a) δ^{26}Mg vs. H_2O, (b) Rb vs. H_2O, (c) Ba vs. H_2O for the Hannuoba granulite xenoliths. Database: H_2O, Rb and Ba from Liu et al. (2001) and δ^{26}Mg from Table 2. Error bars represent 2SD uncertainties.

Figure 11. Magnesium isotopic composition of the crustal samples. Database: δ^{26}Mg data of upper crustal rocks are from Li et al. (2010), Liu et al. (2010), Ling et al. (2013) and Huang et al. (2013a); Middle crustal rocks are from Table 1; Lower crustal rocks are from Teng et al. (2013) and Table 2. The gray bar represents the average mantle (δ^{26}Mg = -0.25 ± 0.07) (Teng et al., 2010a).

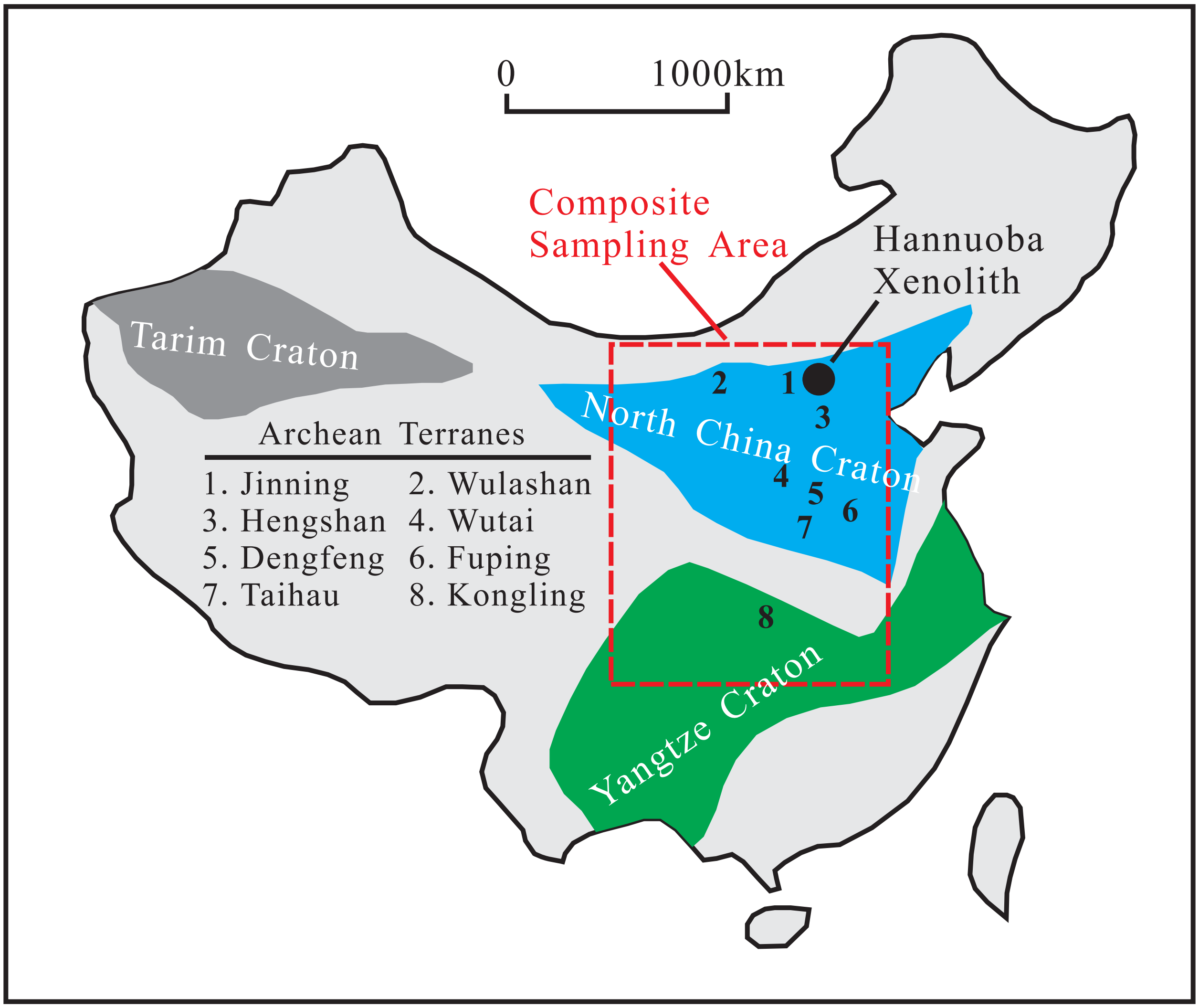
0
1000km
Composite
Sampling Area
Hannuoba
Xenolith
Tarim Craton
North China Craton
Yangtze Craton
1
2
3
4
5
6
7
8
Archean Terranes
1. Jinning
2. Wulashan
3. Hengshan
4. Wutai
5. Dengfeng
6. Fuping
7. Taihau
8. Kongling

Figure 1

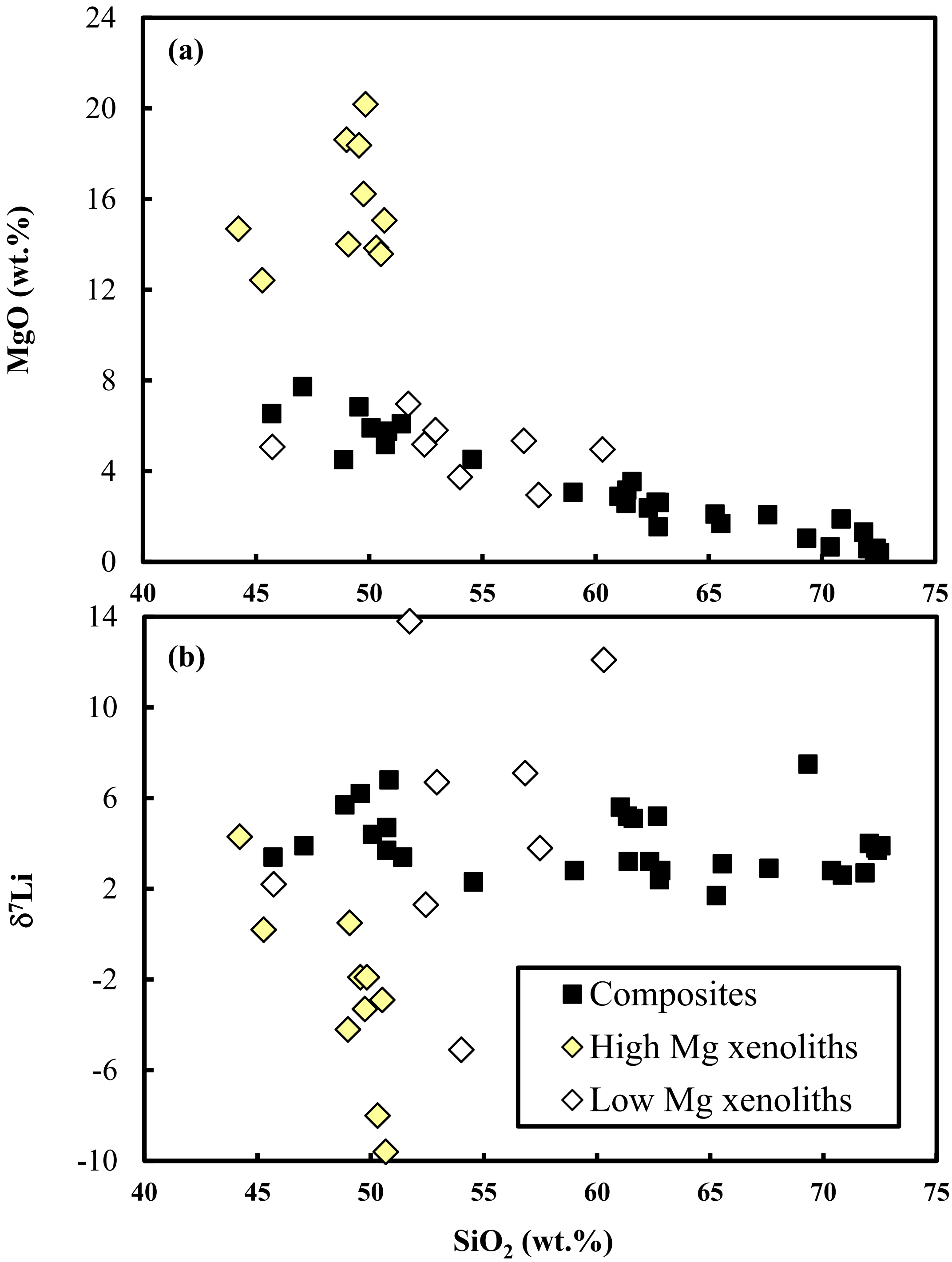
(a)
MgO (wt.%)
24
20
16
12
8
4
0
40
45
50
55
60
65
70
75
(b)
δ7Li
14
10
6
2
-2
-6
-10
Composites
High Mg xenoliths
Low Mg xenoliths
SiO2 (wt.%)

Figure 2

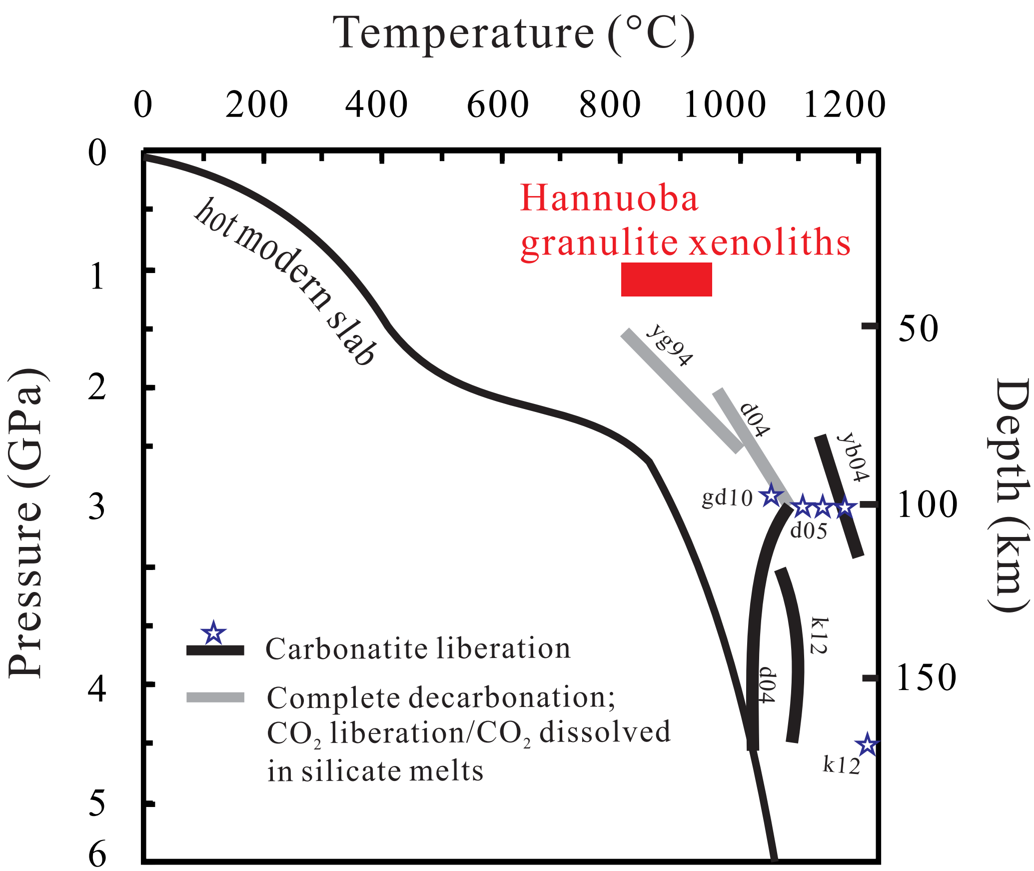
Temperature (°C)
0 200 400 600 800 1000 1200
Pressure (GPa)
0 1 2 3 4 5 6
Depth (km)
50 100 150
hot modern slab
Hannuoba granulite xenoliths
yg94
d04
yb04
gd10
d05
k12
d04
k12
Carbonatite liberation
Complete decarbonation; CO_2 liberation/CO_2 dissolved in silicate melts

Figure 3

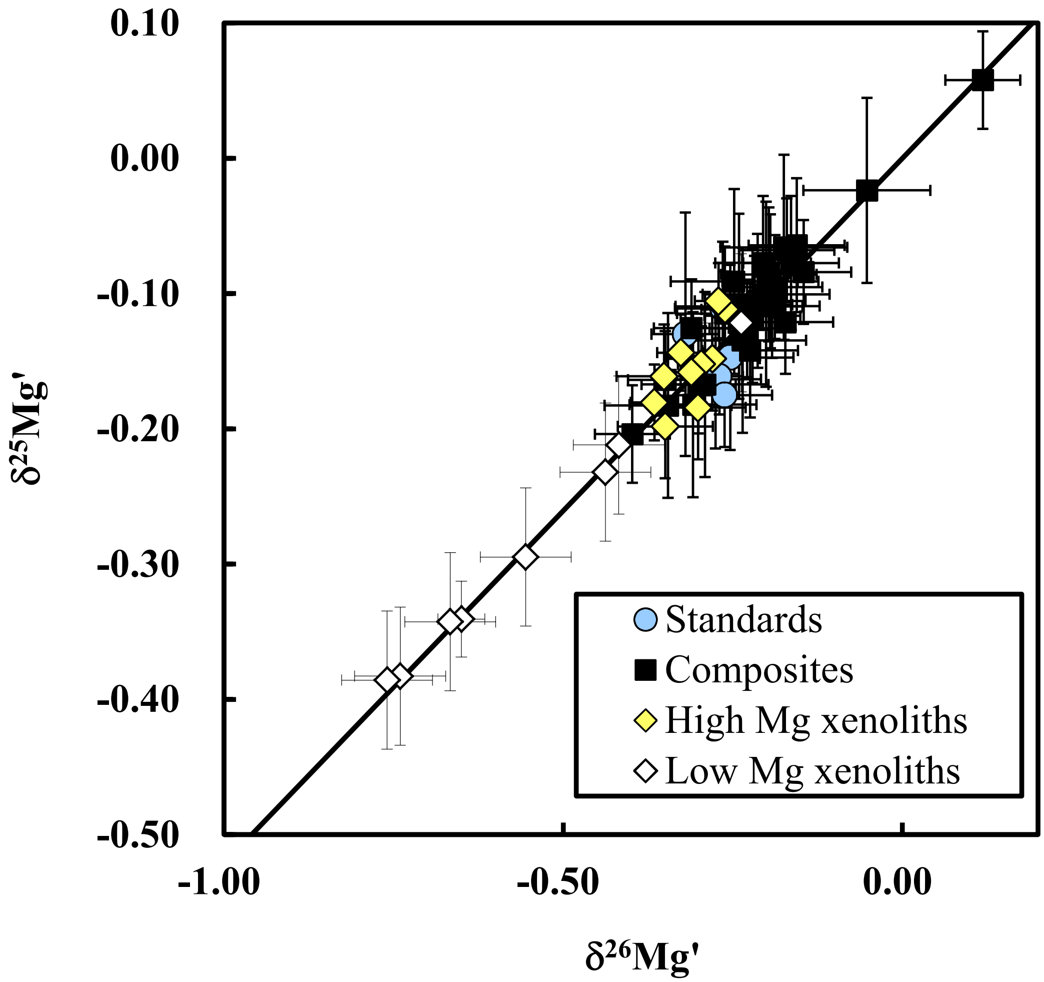
0.10
0.00
-0.10
-0.20
-0.30
-0.40
-0.50
δ25Mg'
-1.00
-0.50
0.00
δ26Mg'
Standards
Composites
High Mg xenoliths
Low Mg xenoliths

Figure 4

This is a preprint, the final version is subject to change, of the American Mineralogist (MSA)
Cite as Authors (Year) Title. American Mineralogist, in press.
(DOI will not work until issue is live.) DOI: http://dx.doi.org/10.2138/am-2015-5275

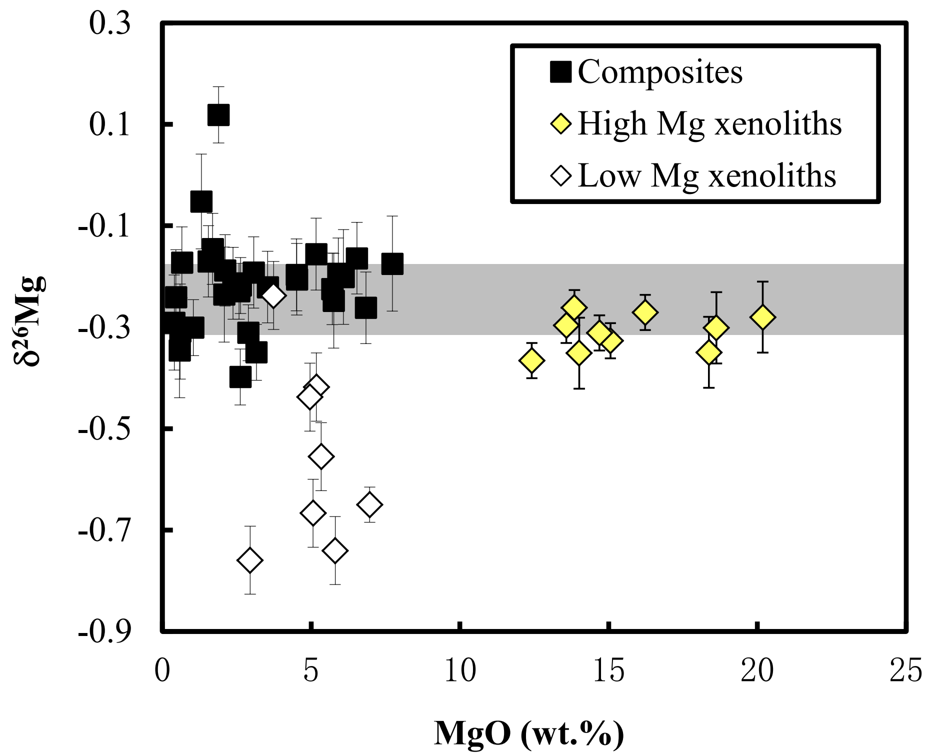

Figure 5

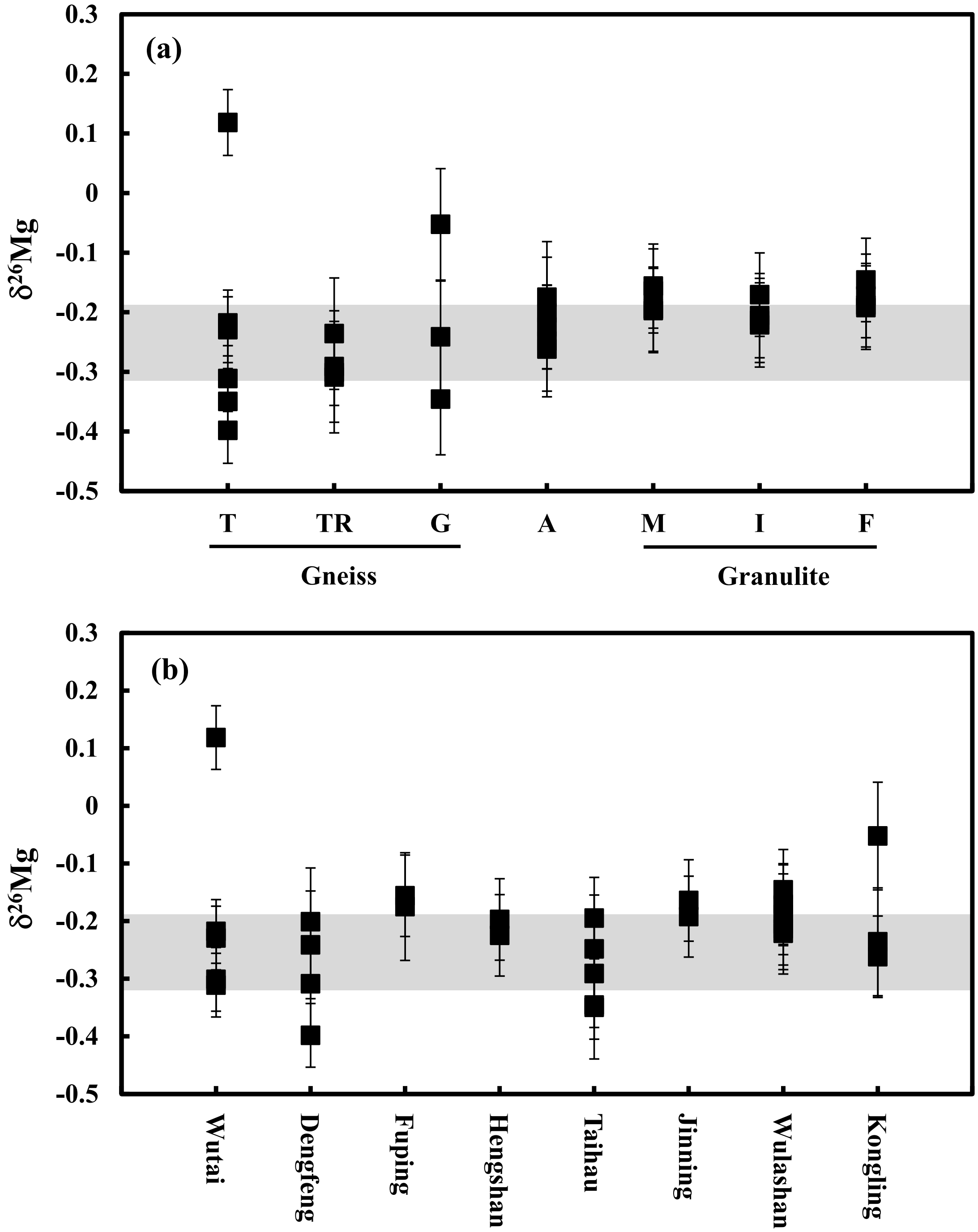
(a)
δ26Mg
0.3
0.2
0.1
0
-0.1
-0.2
-0.3
-0.4
-0.5
T
TR
G
A
M
I
F
Gneiss
Granulite
(b)
δ26Mg
Wutai
Dengfeng
Fuping
Hengshan
Taihau
Jinning
Wulashan
Kongling

Figure 6

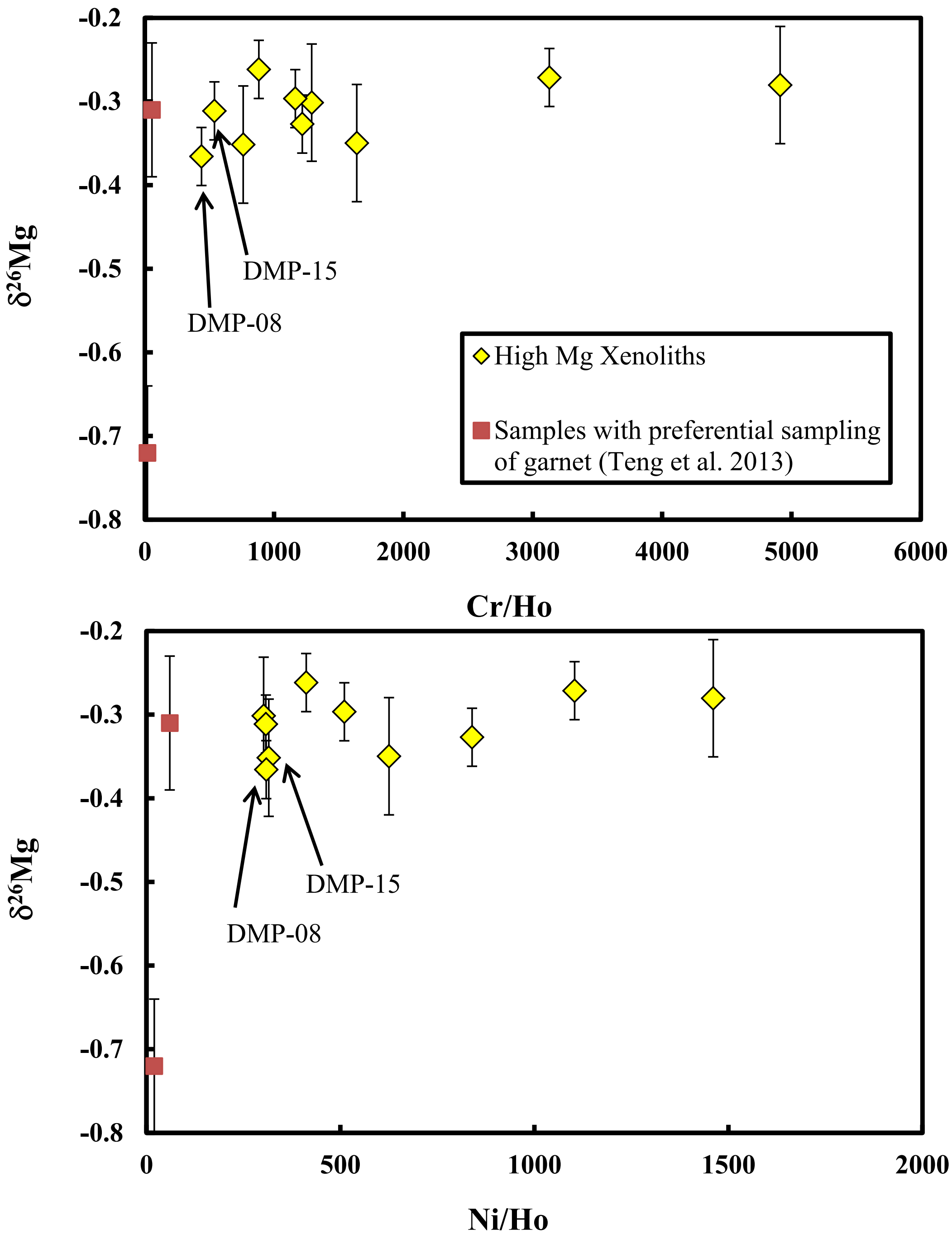
-0.2
-0.3
-0.4
-0.5
-0.6
-0.7
-0.8
δ26Mg
DMP-15
DMP-08
High Mg Xenoliths
Samples with preferential sampling of garnet (Teng et al. 2013)
0
1000
2000
3000
4000
5000
6000
Cr/Ho
-0.2
-0.3
-0.4
-0.5
-0.6
-0.7
-0.8
δ26Mg
DMP-15
DMP-08
0
500
1000
1500
2000
Ni/Ho

Figure 7

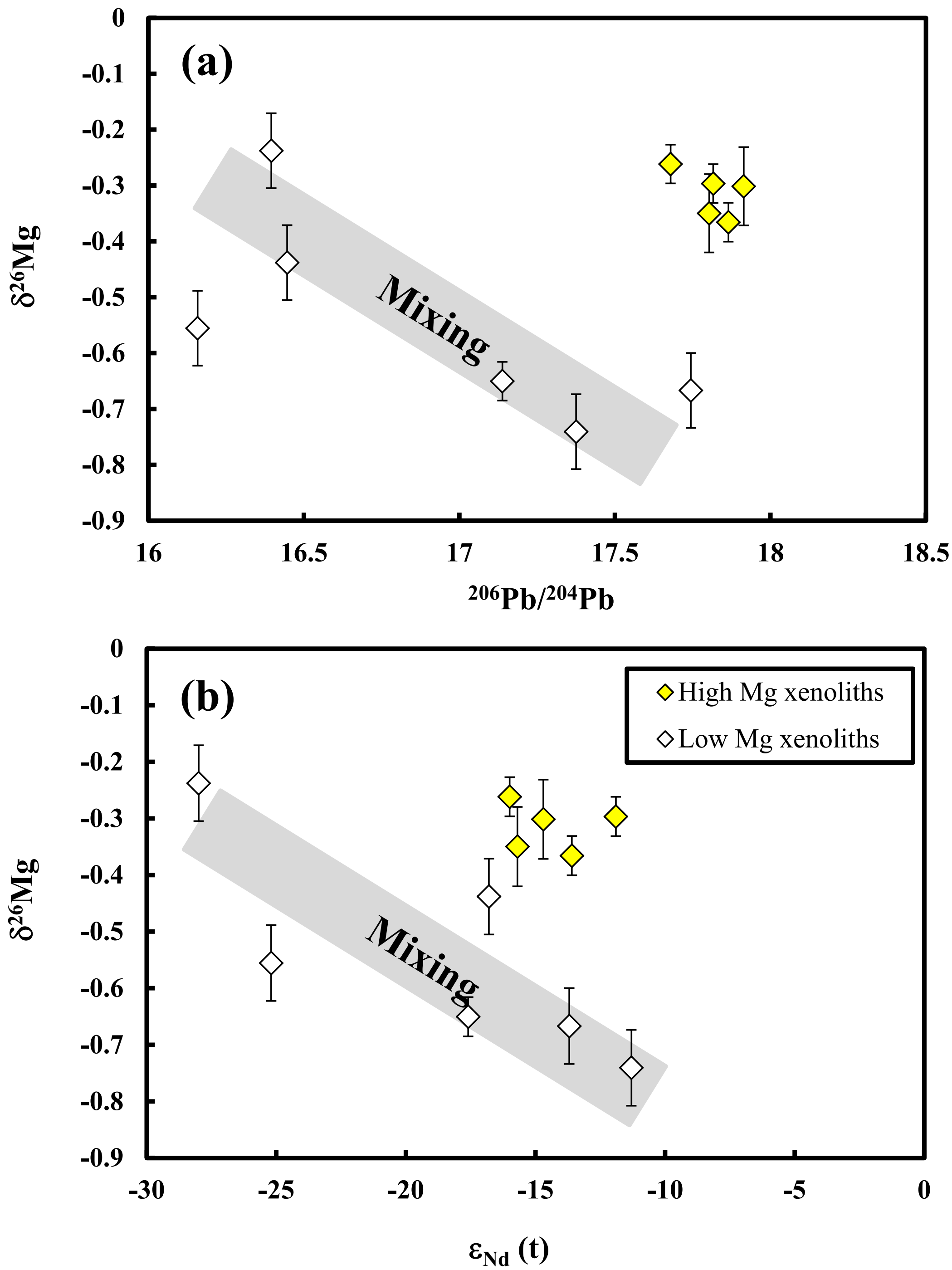
(a)
Mixing
δ26Mg
206Pb/204Pb
(b)
High Mg xenoliths
Low Mg xenoliths
Mixing
δ26Mg
εNd (t)

Figure 8

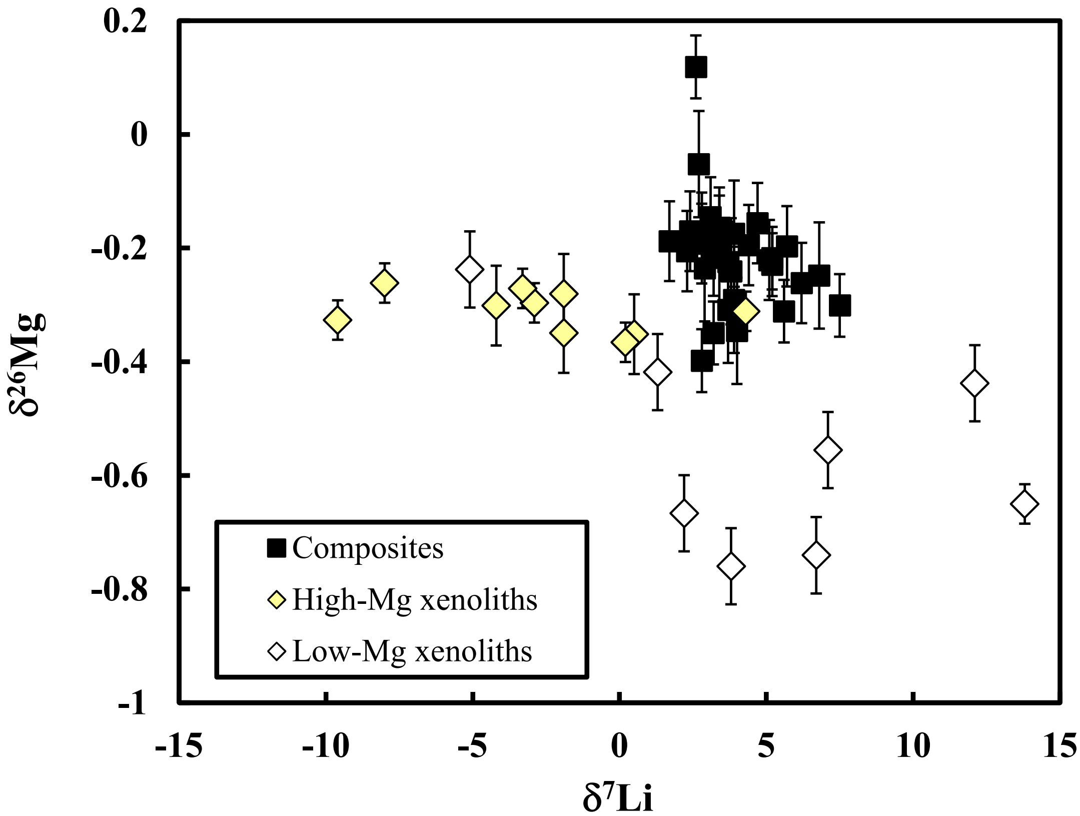
0.2
0
-0.2
-0.4
-0.6
-0.8
-1
δ26Mg
Composites
High-Mg xenoliths
Low-Mg xenoliths
-15
-10
-5
0
5
10
15
δ7Li

Figure 9

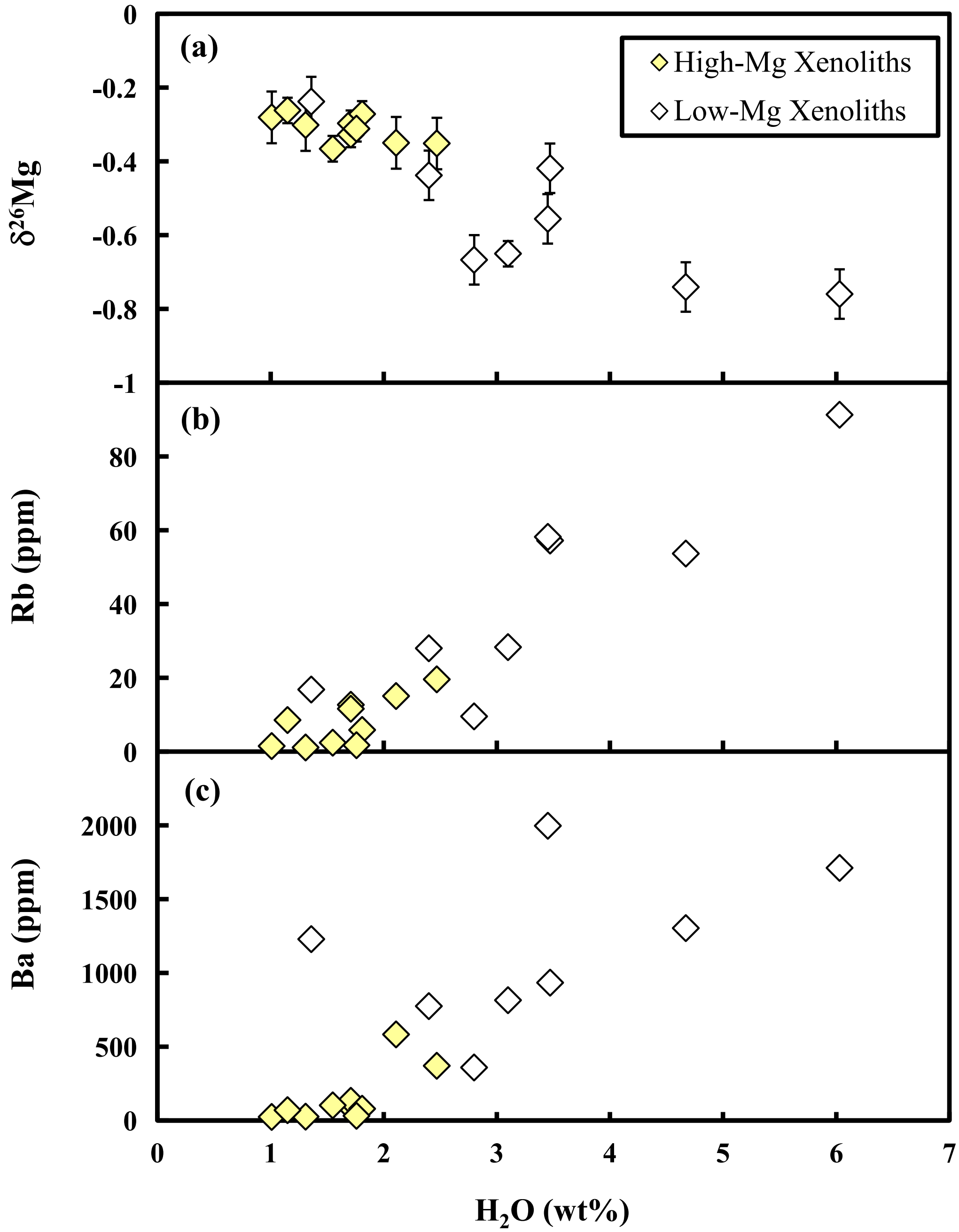
(a)
(b)
(c)
High-Mg Xenoliths
Low-Mg Xenoliths
$\delta^{26}Mg$
Rb (ppm)
Ba (ppm)
H_2O (wt%)
0
-0.2
-0.4
-0.6
-0.8
-1
80
60
40
20
0
2000
1500
1000
500
0
0
1
2
3
4
5
6
7

Figure 10

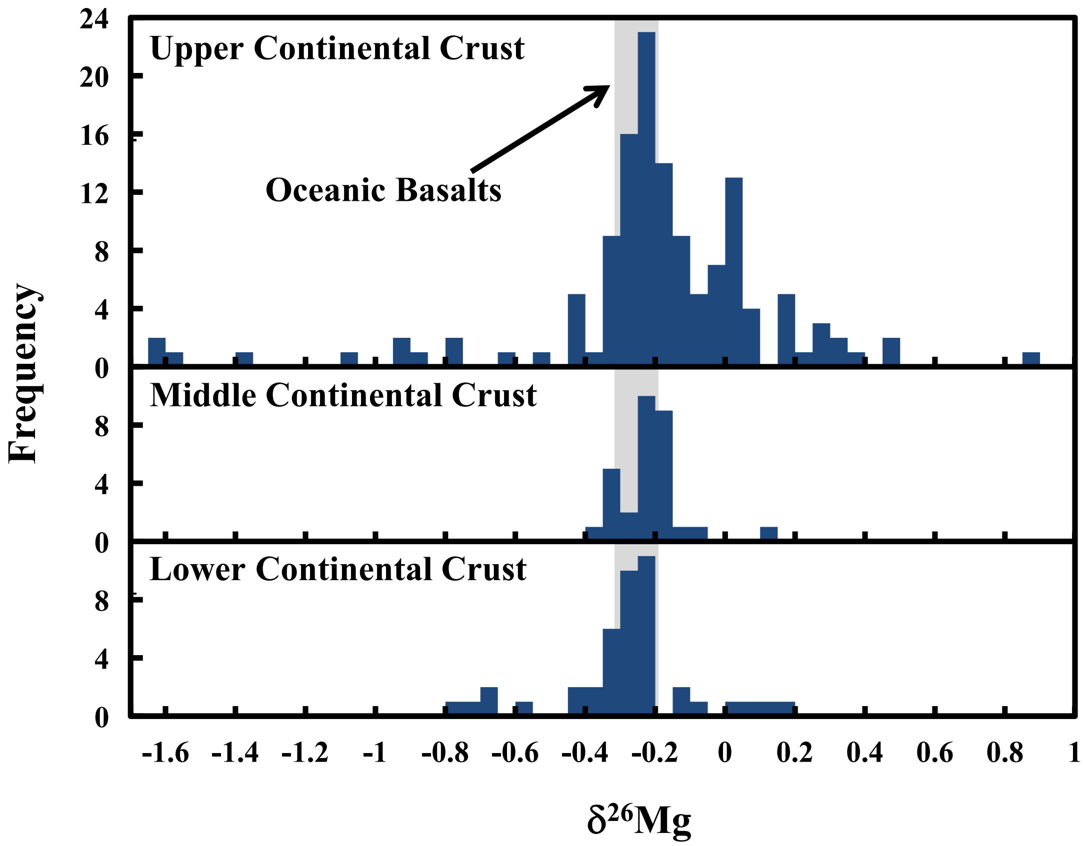
Upper Continental Crust
Oceanic Basalts
Middle Continental Crust
Lower Continental Crust
Frequency
24
20
16
12
8
4
0
-1.6 -1.4 -1.2 -1 -0.8 -0.6 -0.4 -0.2 0 0.2 0.4 0.6 0.8 1
δ26Mg

Figure 11