Iron and magnesium isotope fractionation in oceanic lithosphere and sub-arc mantle: perspectives from ophiolites

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Abstract

We present high-precision Fe and Mg isotopic data for the Purang ophiolite, southwestern Tibet, representing the first combined Fe and Mg isotopic study of the oceanic lithosphere hitherto. The $\delta^{56}\text{Fe}$ and $\delta^{26}\text{Mg}$ values of the ophiolitic peridotite,

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dunite and gabbro vary from -0.209 to 0.187‰ and from -0.28 to -0.14‰, respectively.

The average $\delta^{56}\text{Fe}$ of the peridotites is $-0.030 \pm 0.143\text{‰}$ (2SD, $n = 17$), a value

indistinguishable from abyssal peridotites and chondrites, and lower than oceanic basalts.

The average $\delta^{26}\text{Mg}$ value of the peridotites is $-0.20 \pm 0.10\text{‰}$, a value slightly higher than

both chondrites and oceanic basalts. Correlations between $\delta^{56}\text{Fe}$ and indices of partial

melting indicate fractionation of 0.323‰ in $\delta^{56}\text{Fe}$ between the oceanic lithospheric

mantle and the overlying mafic crust during an early episode of partial melting,

presumably beneath a spreading centre. Subsequent metasomatism in a supra-subduction

zone caused elevated oxygen fugacity and heavy Fe isotopic compositions in the oceanic

lithospheric mantle. The dunite with high Ba/La, a proxy for oxygen fugacity, and high

$\delta^{56}\text{Fe}$ values was likely formed during this process of sub-arc mantle-melt interaction.

The negatively coupled Fe-Mg isotopic variations of the Purang ophiolite indicate that

Mg isotope fractionation may also occur during high-temperature mantle processes. The

observed isotopic variations among different lithologies in the ophiolite may

satisfactorily account for the isotopic differences between arc lavas and mantle

peridotites with respect to oceanic basalts, thus providing implications for crust-mantle

differentiation.

Keywords: ophiolite; Fe isotopes; Mg isotopes; partial melting; metasomatism

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48 **1 Introduction**

49 Studies of peridotite xenoliths and komatiites indicate that the Earth's mantle might
50 have chondritic Fe and Mg isotopic compositions (e.g., [Weyer et al., 2005](#); [Weyer and](#)
51 [Ionov, 2007](#); [Dauphas et al., 2009a, 2010](#); [Handler et al., 2009](#); [Yang et al., 2009](#); [Teng et](#)
52 [al., 2010a](#)). By contrast, oceanic basalts (MORBs and OIBs), partial melting products of
53 the mantle, have chondritic Mg ([Teng et al., 2007, 2010a](#); [Bourdon et al., 2010](#)) and non-
54 chondritic, heavier Fe isotopic compositions ([Beard et al., 2003](#); [Weyer and Ionov, 2007](#);
55 [Teng et al., 2008; 2013](#); [Schuessler et al., 2009](#)). The difference in Fe isotopic
56 composition between oceanic basalts and mantle peridotites has been considered as
57 indicative of Fe isotope fractionation during partial melting. As ferric iron (Fe^{3+}) is more
58 incompatible (e.g., [Canil et al., 1994](#)) than ferrous iron (Fe^{2+}) (e.g., [Polyakov and Mineev,](#)
59 [2000](#); [Polyakov et al., 2007](#); [Dauphas et al., 2009a](#)), heavy Fe isotopes are preferentially
60 concentrated in the melts, leaving an isotopically light residue ([Williams et al., 2005](#);
61 [Weyer and Ionov, 2007](#); [Nebel et al., 2013](#)). However, a lack of measurable Fe isotope
62 fractionation in abyssal peridotites ([Craddock et al., 2013](#)) implies that Fe isotope
63 fractionation of peridotite residues is limited during partial melting and requires further
64 constraint. It also conflicts with the isotopic feature of boninites and island arc basalts,

which originate from a more oxidized environment compared to the lithospheric mantle beneath the mid-ocean ridges whereas have lighter Fe isotopic compositions (Dauphas et al., 2009a). This discrepancy is not yet fully understood, although some studies suggested that redox variations and fractional crystallization might play an important role (Beard and Johnson, 2004; Williams et al., 2005; Dauphas et al., 2009a, 2014; Nebel et al., 2013).

Unlike Fe isotopes, Mg isotopes appear to be insensitive to the partial melting processes described above. However, recent studies have revealed that some cratonic eclogites and subduction-related basalts have lighter Mg isotopic compositions relative to the lithospheric mantle (Wang et al., 2012, 2014; Yang et al., 2012). These abnormal $\delta^{26}\text{Mg}$ signals thus raise an important question on the mechanisms responsible for transferring the light $\delta^{26}\text{Mg}$ in the recycled eclogitic slab to the overlying mantle wedge, and subsequent manifestation on Earth's surface as continental volcanic rocks. Oceanic lithosphere represented by ophiolites that commonly involve recycled materials during subduction may record recycling process of light Mg isotopes. However, the lack of relevant data on oceanic lithosphere impedes our understanding of the above question.

In order to constrain the behavior of Fe and Mg isotope fractionation in oceanic and sub-arc lithosphere, we present, to our knowledge, the first combined Fe and Mg isotopic study on the well-studied Purang ophiolite complex in southwestern Tibet. This ophiolitic

massif was considered to have formed after melt extraction at a mid-ocean ridge setting, followed by percolation of arc magmas in a supra-subduction zone (e.g., [Liu et al., 2014](#); [Li et al., 2015](#)), thus being the ideal samples to study Fe and Mg isotope fractionation during the evolution of the upper mantle. Specifically, we aim at constraining the Fe and Mg isotopic compositions of the oceanic lithospheric mantle and revealing the influence of various mantle processes on fractionating Fe and Mg isotopes. Our results show that the Purang ophiolite has variable, but negatively correlated Mg and Fe isotopic compositions, which suggests that partial melting and melt-peridotite interaction may have played an important role in fractionating Fe and Mg isotopes in the upper mantle.

2 Geological background and sample descriptions

The Tibetan Plateau formed by collision between different blocks. The Yarlung Zangbo Suture Zone is the youngest suture, which marks the final collision of the Indian with Eurasian plates ([Fig. 1](#)). This suture zone is marked by Cretaceous ophiolites that represent remnants of the Neo-Tethyan oceanic lithosphere ([Hébert et al., 2012](#) and references therein). The Purang ophiolite complex in the western segment of the Yarlung Zangbo Suture Zone consists mainly of a large ultramafic massif with limited exposures of crustal rocks ([Fig. 1](#)). The outcrops are dominated by harzburgites, with local

occurrence of lherzolites and subordinate dunites. Chromitites are occasionally observed in dunites in places (Liu et al., 2015). Mafic rocks commonly occur as dykes or veins in the peridotites with MORB-like composition (Miller et al., 2003; Liu et al., 2014, 2015).

Lherzolites and harzburgites in the Purang ophiolite commonly display coarse equigranular to porphyroclastic textures and have a mineral assemblage of olivine, orthopyroxene, clinopyroxene and spinel (Fig. 2a). Olivine is subhedral to euhedral and displays kink bands. Some olivine grains are locally replaced with serpentine (Fig. 2b). Olivine grains in harzburgites are commonly large and fresh (Fig. 2c). Orthopyroxene occurs as coarse porphyroclasts and is surrounded by fine-grained orthopyroxene, olivine and spinel (Fig. 2d). Clinopyroxene generally displays irregular forms with curvilinear grain boundaries. Both orthopyroxene and clinopyroxene show exsolution lamellae. Anorthitic plagioclase and pargasitic amphibole are also observed in Purang peridotites (Liu et al., 2010). Dunites in the Purang ophiolite occur as pods and lenses in the harzburgite bodies (Fig. 1), and occasionally host small banded or disseminated chromites (Fig. 2e). Minor orthopyroxene and clinopyroxene are also observed in dunites. Gabbroic dykes or veins consist of plagioclase, clinopyroxene and ilmenite (Fig. 2f).

Nine lherzolites, eight harzburgites, one dunite and one gabbro were selected for Fe and Mg isotope analyses. They are typically fresh with traces of serpentine along

fractures of silicate minerals (Fig. 2). Major elemental compositions of minerals in the Purang peridotites and dunite (Table A1) overlap those in literatures and are analogous to those in abyssal peridotites and MORB-like ophiolites (e.g., Liu et al., 2014; Li et al., 2015). Clinopyroxene has extremely depleted Nd isotopic compositions (Miller et al., 2003) and variable trace element patterns overlapping the fields of abyssal and supra-subduction zone peridotites (Liu et al., 2010, 2012). In terms of bulk trace elemental compositions, the gabbro and the majority of lherzolites display similar rare earth element patterns and multiple trace element patterns to those of MORBs, whereas the dunite and most harzburgites are depleted in light rare earth elements and high field strength elements (Fig. 3). Furthermore, platinum group element and osmium isotope studies of the Purang peridotite reveal their compositional transition from abyssal peridotites to mantle wedge xenoliths (Liu et al., 2012). These features suggest that the Purang ophiolite sequence was initially formed at a mid-ocean ridge environment and was subsequently modified at a supra-subduction zone setting.

3 Analytical methods

3.1 Iron isotopic analysis

Iron isotope analyses were performed at the Isotope Laboratory of China University of Geosciences, Beijing, following the established methods of [Dauphas et al. \(2009b\)](#), which was reported in detail in [He et al. \(2015\)](#). Pulverized samples (~5 mg) were dissolved in Teflon beakers in HF-HNO₃-HClO₄ mixture. Sample solutions were evaporated to dryness at 180 °C and the residues were then refluxed with aqua regia (HCl:HNO₃ = 3:1) and excess HNO₃ aqua regia (HCl:HNO₃ = 2:1) successively at 130 °C, and subsequently evaporated to dryness at 100 °C. All dried residues were finally dissolved in 6N HCl for chromatographic separation. Sample solutions were checked at each step for complete dissolution.

After dissolution, Fe was separated from matrix elements by anion-exchange chromatography in HCl following the procedures by [Dauphas et al. \(2009b\)](#). Iron isotopic compositions were analyzed by sample-standard bracketing method using a Thermo-Finnigan Neptune Plus MC-ICPMS. The bracketing sequence was repeated for four times to enhance reproducibility and accuracy. The long-term external reproducibility and accuracy on $\delta^{56}\text{Fe}$ are better than ± 0.05 (2SD), based on replicate analyses of natural and synthetic standard solutions ([He et al., 2015](#)). Two international standards of basaltic compositions, BHVO-2 and BCR-2, were analyzed for multiple times to monitor accuracy and yielded $\delta^{56}\text{Fe}$ values of $0.101 \pm 0.050\text{‰}$ and $0.101 \pm 0.045\text{‰}$ with $\delta^{57}\text{Fe}$ of

0.161 $\pm$ 0.063‰ and 0.131 $\pm$ 0.063‰, respectively (Table 1), identical to those reported by Weyer et al. (2005), Craddock and Dauphas (2011), Millet et al. (2012) and Sossi et al. (2015).

3.2 Magnesium isotopic analysis

Magnesium isotopic analyses were performed at the Isotope Laboratory of the University of Washington, Seattle, following the method described in previous studies (Teng et al., 2007, 2010a, 2015; Yang et al., 2009; Li et al., 2010). Sample dissolution and column chemistry were carried out in a clean laboratory. About ~1 to 3 mg of sample powder was digested in Optima-grade concentrated HF-HNO₃-HCl mixture in sealed 7 ml Teflon beakers on a hot plate in a laminar flow exhaust hood. After complete dissolution, the samples were dried and redissolved in 1N HNO₃ for chromatographic separation. Magnesium was purified on a cation exchange resin (Bio-Rad AG50W-X8, 200-400 mesh) in 1N HNO₃. The same procedure was repeated twice in order to effectively remove matrix elements.

Magnesium isotopic compositions were analyzed on a Nu Plasma MC-ICPMS using the standard-sample bracketing method (Teng and Yang, 2014). At least two standards (JB-1 basalt, PCC-1 peridotite and Hawaiian seawater) were analyzed with each group of samples during the course of chemical separation and instrumental analysis to enhance

accuracy and reproducibility. One analysis of the standard JB-1 yielded a $\delta^{26}\text{Mg}$ value of -0.22‰ and two analyses of the standard PPC-1 yielded $\delta^{26}\text{Mg}$ values of -0.22 and -0.20‰, and four analyses of the Hawaiian seawater standard gave $\delta^{26}\text{Mg}$ values of -0.81‰, -0.77‰, -0.81‰ and -0.80‰, consistent with published data ($-0.276 \pm 0.098\text{‰}$ for JB-1, $-0.29 \pm 0.055\text{‰}$ for PCC-1 and $-0.83 \pm 0.09\text{‰}$ for seawater, [Teng et al., 2015](#); $0.22 \pm 0.10\text{‰}$ for PCC-1, [Huang et al., 2011](#)). Repeated analyses of samples indicate data reproducibility of better than 0.07‰.

4 Results

The Fe and Mg isotopic data are reported in [Table 1](#). The $\delta^{56}\text{Fe}$ values of the samples vary from -0.209 to 0.187‰, overlapping ranges of abyssal peridotites and peridotite xenoliths, and are systematically lower than that reported for oceanic basalts ([Fig. 4](#)). The gabbro and dunite have heavier Fe isotopic compositions ($\delta^{56}\text{Fe} = 0.114 \pm 0.035\text{‰}$ and $0.187 \pm 0.047\text{‰}$, respectively; [Table 1](#)) than the peridotites. The average $\delta^{56}\text{Fe}$ value for the peridotites ($-0.030 \pm 0.143\text{‰}$, $n = 17$) is indistinguishable from that of chondrites within error ($+0.005 \pm 0.006\text{‰}$; [Craddock and Dauphas, 2011](#)).

The Mg isotopic compositions of investigated lithologies display a limited range ($\delta^{26}\text{Mg} = -0.28$ to -0.14‰) ([Table 1](#)), and no systematic relation is found among various

rock types. The average $\delta^{26}\text{Mg}$ value of $-0.20 \pm 0.10\text{‰}$ is slightly higher than the average values of oceanic basalts and peridotite xenoliths (Fig. 4).

5 Discussion

Seafloor hydrothermal alteration is a common phenomenon observed in ophiolitic rocks, and redistribution of Fe and Mg isotopes might occur during this process. In this study, the majority of samples have undergone various extents of serpentinization, as indicated by the variable loss-on-ignition (LOI) (0 to 14 wt.%; Table A2). However, the lack of correlations between LOI and $\delta^{56}\text{Fe}$ or $\delta^{26}\text{Mg}$ (Fig. 5) indicates that serpentinization did not play an important role in controlling Fe and Mg isotopic compositions. This is consistent with studies of Fe and Mg isotopes in komatiites (Dauphas et al., 2010), abyssal peridotites (Craddock et al., 2013) and ultramafic complex (Beinlich et al., 2014). Hence, the isotopic data for the Purang ophiolite can be used to examine the Fe and Mg isotope fractionation from mid-ocean ridge to supra-subduction zone and to constrain the Fe and Mg isotopic composition of the oceanic lithosphere.

Ophiolitic and abyssal peridotites have average $\delta^{56}\text{Fe}$ values of $-0.030 \pm 0.143\text{‰}$ and $0.010 \pm 0.007\text{‰}$, respectively (Table 1; Craddock et al., 2013), both of which are

consistent with the $\delta^{56}\text{Fe}$ values of mantle peridotite xenoliths (e.g., Williams et al., 2005; Weyer and Ionov, 2007; Zhao et al., 2010) and are indistinguishable from that of chondrites (Fig. 4) (e.g., Craddock and Dauphas, 2011). This suggests that both oceanic and continental lithospheric mantle have a near-chondritic Fe isotopic composition. Nonetheless, both ophiolitic and abyssal peridotites display large Fe isotopic variation, which reflects isotope fractionation during partial melting and metasomatism.

Below, we first discuss the behaviors of Fe isotopes during partial melting of the mantle under mid-ocean ridge setting and mantle metasomatism during supra-subduction process, followed by Mg isotopes.

5.1 Iron isotope fractionation during partial melting of the mantle under mid-ocean ridge setting

Peridotites of the Purang ophiolite contain silicate minerals with high and variable Mg# [$100 \times \text{Mg}/(\text{Mg} + \text{Fe})$], $\text{Mg\#}_{\text{Ol}} = 90.5$ to 91.6 , $\text{Mg\#}_{\text{Opx}} = 89.7$ to 91.8 , $\text{Mg\#}_{\text{Cpx}} = 91.1$ to 94.4]; spinel with high Cr# [$100 \times \text{Cr}/(\text{Cr} + \text{Al})$], 15.8 to 50.1] (Table S1) and depletion in rare earth elements, which can be modeled by extraction of ~5 to 25% partial melts under an isobaric melting condition (Figs. 3, 6a). The variation in MgO content of these peridotites suggests increasing degree of partial melting from lherzolites to harzburgites

(Fig. 6f; Table A2). Their $\delta^{56}\text{Fe}$ values correlate with various indices of melting (i.e. Cr# of clinopyroxene, MnO content of spinel and whole-rock TFe_2O_3 and Sc) except for sample PL1212-1 and PL1203-5 that might have experienced Fe-Mg inter-diffusion (Fig. 6b-e), implying that Fe isotopes were fractionated during mantle melting at a spreading centre. The partial melt, as represented by the Purang gabbro, has similar geochemical features and $\delta^{56}\text{Fe}$ value to MORBs (Figs. 3, 6g), which provides further evidence that partial melting of the upper mantle can produce melts enriched in heavy Fe isotopes (Williams et al., 2005; Weyer and Ionov, 2007; Dauphas et al., 2009a, 2014; Nebel et al., 2013; Teng et al., 2013).

Williams et al. (2005) estimated a melt-residue fractionation of -0.13‰ in $\delta^{56}\text{Fe}$ at 20% degrees of partial melting. However, the value is higher than the observed fractionation of ~0.085‰ between typical MORBs ($\delta^{56}\text{Fe} = 0.105\text{‰}$; Teng et al., 2013) and mantle peridotite xenoliths ($\delta^{56}\text{Fe} = 0.02\text{‰}$; Weyer and Ionov, 2007) and predicated fractionation of <~0.04‰ (Dauphas et al., 2009a) at similar degrees of partial melting (Fig. 4). In terms of Fe isotope fractionation among peridotite residues, Craddock et al. (2013) proposed a small variation of only 0.01‰. This value stands in marked contrast to the results from Purang ophiolite rocks, where the $\delta^{56}\text{Fe}$ of the most refractory harzburgite (PL1212-1) is 0.231‰ lower than that of the most fertile lherzolite (PL1202-

2), and 0.323‰ lower relative to the gabbro (Table 1). The limited fractionation found in Craddock et al. (2013) likely reflect to the small compositional variation in their samples (i.e. $\text{Cr\#}_{\text{spinel}} = 15\text{-}25$).

5.2 Iron isotope fractionation in mantle wedge at supra-subduction zone setting

Mantle metasomatism is an important process that can potentially modify Fe isotopic compositions (Weyer and Ionov, 2007; Zhao et al., 2010, 2012; Huang et al., 2011; Nebel et al., 2013; Poitrasson et al., 2013). The trace element patterns of whole-rocks and clinopyroxene separates, combined with the presence of anorthitic plagioclase and pargasitic amphibole, demonstrate that the Purang peridotites underwent low degree (<2%) metasomatism by arc magmas (Fig. 3; Liu et al., 2010, 2014). The lherzolites preserve refractory chemical and isotopic compositions i.e. high Mg# and Cr# in constituent minerals (Table A1) and depleted Nd isotopic compositions in clinopyroxene (Miller et al., 2003), indicating relatively weak metasomatism than harzburgites (Fig. 3). These features preclude the possibility that the lherzolites studied here are refertilization products of the refractory harzburgites left behind by previous melting events (Fig. 6; Liu et al., 2014). The Fe isotopic composition increases by 0.076‰ from the most refractory harzburgites to the least refractory one, whose $\delta^{56}\text{Fe}$ value is close to that of MORBs and

back arc basin basalts (Table 1; Fig. 6a; Teng et al., 2013), indicating that the metasomatic melts are enriched in heavy Fe isotopes. Brounce et al. (2014) found that Ba/La ratio is positively correlated with oxygen fugacity and that arc lavas with Ba/La ratios of >10 are commonly more oxidized than that of MORBs (~2.5). They also demonstrated that $\text{Fe}^{3+}/\Sigma\text{Fe}$ ratios of island arc lavas range from 0.195 to 0.280, which are more oxidized than average MORB (~0.16) and fertile upper mantle (~0.036) (Cottrell and Kelley, 2011; Canil et al. 1994). The elevated Ba/La and $\text{Fe}^{3+}/\Sigma\text{Fe}$ ratios of the Purang peridotites, which is similar to those of arc-related basalts, suggest that the metasomatic melts should also be highly oxidized (Fig. 6g, i; Brounce et al., 2014). These features are consistent with the hypothesis that Fe^{3+} is usually enriched in heavy Fe isotopes relative to Fe^{2+} (e.g., Polyakov and Mineev, 2000). Among all the studied lithologies, the dunite has the heaviest $\delta^{56}\text{Fe}$ and high Ba/La ratio (Figs. 6g, 7), and thus was most likely formed as the reaction product between the residual mantle and arc-like infiltrating melts, an explanation consistent with previous petrological and geochemical studies (Liu et al., 2014; Li et al., 2015).

Our data also place strong constraints on the Fe isotopic compositions of arc lavas. The $\delta^{56}\text{Fe}$ values of arc lavas are different from those of MORBs and OIBs (Fig. 4), indicating that Fe isotopic composition of oceanic mafic crust varies with tectonic

settings. In general, mid-ocean ridges and oceanic islands have heavier $\delta^{56}\text{Fe}$ values than island arcs (Fig. 4; Beard et al., 2003; Weyer and Ionov, 2007; Dauphas et al., 2009a, 2014; Schuessler et al., 2009; Weyer and Seitz, 2012; Teng et al., 2013). Such differences are related to mantle redox conditions, compositional modification by slab melts/fluids and the degrees of partial melting (Beard and Johnson, 2004; Williams et al., 2005; Dauphas et al., 2009a; Nebel et al., 2013; Teng et al., 2013). Arc magmas commonly undergo composition modification as they pass through the overlying lithospheric mantle wedge, and preferentially leave the heavy Fe isotopes in the mantle en route (e.g., Nebel et al., 2013; Brounce et al., 2014). This process can explain the lighter Fe isotopic compositions of boninites and island arc basalts compared to MORBs and OIBs (Fig. 4) (e.g., Weyer and Ionov, 2007; Dauphas et al., 2009a; Teng et al., 2013). On the other hand, the mantle wedge modified by such slab melts/fluids must become more oxidized and have heavier $\delta^{56}\text{Fe}$ values than estimates from arc magmas. A simple mixing calculation demonstrates that the formation of mantle wedge requires 20 to 60% of Fe interchange between metasomatic agents (e.g., arc magmas) and the depleted mantle (Fig. 7).

5.3 Magnesium isotope fractionation during mantle melting and metasomatism

In contrast to Fe isotopic systematics, both oceanic basalts and peridotites have relatively uniform Mg isotopic compositions (Fig. 4) as high temperature processes (i.e. partial melting, metasomatism and fractional crystallization) are inefficient in fractionating Mg isotopes (e.g., Teng et al., 2007, 2010a, 2010b; Yang et al., 2009; Bourdon et al., 2010; Liu et al., 2011; Pogge von Strandmann et al., 2011). Similar to previous studies, the $\delta^{26}\text{Mg}$ values of the Purang ophiolite do not correlate with common indices for partial melting and metasomatism (Fig. 6f, h). However, $\delta^{26}\text{Mg}$ and $\delta^{56}\text{Fe}$ values are negatively coupled within individual rock types with the following features (Fig. 8): i) $\delta^{26}\text{Mg}$ and $\delta^{56}\text{Fe}$ are inversely correlated in lherzolites; ii) variably metasomatized harzburgites have slightly lighter Mg but heavier Fe isotopic compositions than lherzolites; iii) the gabbro has lighter $\delta^{26}\text{Mg}$ and heavier $\delta^{56}\text{Fe}$ relative to peridotites.

A few studies have reported negatively coupled Mg-Fe isotope fractionation in olivine xenocrysts and peridotite xenoliths, which were attributed to Fe-Mg inter-diffusion between olivine and evolving magma (e.g., Dauphas et al., 2010; Teng et al., 2011; Sio et al., 2013) and between peridotite and metasomatic melts (e.g., Huang et al., 2011). This mechanism may exert great effects on the formation of dunites and chromitites (sample PL1203-5 and harzburgite PL1212-1), governing the inter-mineral

Fe-Mg exchange in the sub-arc mantle (Zhou et al., 2014), but should not be the main mechanism producing the negative correlations observed in the studied samples. Instead, the coupled Fe-Mg isotopic signatures may reflect Mg isotope fractionation during metasomatism and/or partial melting in the sub-arc mantle processes. Future detailed Mg isotopic studies of arc magmas are needed in order to better constrain the exact mechanisms responsible for the coupled Fe and Mg isotopic variation.

6 Concluding remarks

The Purang ophiolitic rocks display a relatively wide range of Fe and Mg isotopic variations, which are related to different evolutionary stages of the Neo-Tethyan oceanic lithosphere. Coupled Fe and Mg isotope fractionations in the investigated rocks are thought to have resulted from mantle melting and subsequent metasomatism by highly oxidized arc-related magmas. Partial melting occurred at the mid-ocean ridge stage generated oceanic mafic crust with high $\delta^{56}\text{Fe}$ and low $\delta^{26}\text{Mg}$ values, leaving isotopically light Fe and heavy Mg refractory peridotites behind. By contrast, metasomatism by arc melts in a supra-subduction zone regime significantly elevated oxygen fugacity and consequentially enriched heavy Fe and light Mg isotopes in the mantle wedge above the subducted slab. The Fe and Mg isotopic variations measured in the Purang ophiolite can

explain the isotopic differences between MORBs, OIBs, arc lavas and peridotites, implying that the currently observed chondritic Fe and Mg isotopic compositions of the mantle is probably a mixed feature of multi-stage melting and metasomatism history of the oceanic lithospheric mantle.

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Figure Captions:

Fig. 1 Sketch geological map of the Purang ophiolite and its surrounding lithological units.

Fig. 2 Petrography of the Purang ophiolite. a) Lherzolite sample PL1204-11 shows a mineral assemblage of olivine, orthopyroxene, clinopyroxene and spinel; b) Lherzolite sample PL1205-6 shows partially serpentinizing feature; c) Harzburgite sample PL1211-3 contains fresh and coarse olivine grain; d) Harzburgite sample PL1212-1 shows porphyroclastic texture; e) Dunite sample PL1203-5 contains spinel band; f) Gabbro sample PL1212-4 contains plagioclase, clinopyroxene and ilmenite. Cpx, clinopyroxene; Ilme, ilmenite; Ol, olivine; Opx, Orthopyroxene; Sp, spinel.

Fig. 3 Primitive mantle-normalized rare earth element patterns (a, c) and trace element patterns (b, d) for the Purang ophiolite. Fractional melting models (a, c) follow [Niu \(2004\)](#) and use both primitive mantle ([Sun and McDonough, 1989](#)) and a depleted mantle source ([Niu and Hekinian, 1997](#)).

Fig. 4 Fe and Mg isotope frequency distributions of boninites, island arc basalts, mid-ocean ridge basalts (MORBs), oceanic island basalts (OIBs), peridotite xenoliths, abyssal peridotites, and ophiolites. Iron isotope data sources: boninites, [Dauphas et al. \(2009a\)](#); island arc basalts, [Weyer and Ionov \(2007\)](#), [Dauphas et al. \(2009a\)](#), [Teng et al. \(2013\)](#); MORBs, [Beard et al. \(2003\)](#), [Schuessler et al. \(2009\)](#), [Teng et al. \(2013\)](#); OIBs, [Beard et al. \(2003\)](#), [Weyer and Ionov \(2007\)](#), [Weyer and Seitz \(2012\)](#), [Teng et al. \(2013\)](#); abyssal peridotites, [Craddock et al. \(2013\)](#); peridotite xenoliths, [Beard et al. \(2003\)](#), [Weyer and Ionov \(2007\)](#), [Zhao et al. \(2010, 2012\)](#), [Huang et al. \(2011\)](#), [Poitrasson et al. \(2013\)](#);

ophiolites, this study. Magnesium isotope data sources: MORBs, [Bourdon et al. \(2010\)](#), [Teng et al. \(2010a\)](#); OIBs, [Teng et al. \(2007, 2010a\)](#), [Bourdon et al. \(2010\)](#); peridotite xenoliths, [Yang et al. \(2009\)](#), [Young et al. \(2009\)](#), [Bourdon et al. \(2010\)](#), [Teng et al. \(2010b\)](#), [Huang et al. \(2011\)](#), [Liu et al. \(2011\)](#), [Pogge von Strandmann et al. \(2011\)](#), [Xiao et al. \(2013\)](#); ophiolites, this study. Dashed lines represent average $\delta^{56}\text{Fe}$ (0.005‰, [Craddock and Dauphas, 2011](#)) and $\delta^{26}\text{Mg}$ (-0.28‰, [Teng et al., 2010b](#)) values of chondrites.

Fig. 5 Correlations of $\delta^{56}\text{Fe}$ and $\delta^{26}\text{Mg}$ with whole-rock loss on ignition for the Purang ophiolite. Abyssal peridotite data are compiled from [Craddock et al. \(2013\)](#).

Fig. 6 Correlations of (a) Mg# in olivine (Ol) and Cr# in spinel (Sp) and (b-j) $\delta^{56}\text{Fe}$ and $\delta^{26}\text{Mg}$ with mineral and whole-rock compositions for the Purang ophiolite. The olivine-spinel mantle array (OSMA) and partial melting trend are from [Arai \(1994\)](#). The Ba/La ratio of MORB and arc trend are from [Brounce et al. \(2014\)](#). The $\text{Fe}^{3+}/\Sigma\text{Fe}$ ratios of MORB (~0.16) and fertile upper mantle (~0.036) are from [Cottrell and Kelley \(2011\)](#) and [Canil et al. \(1994\)](#), respectively.

Fig. 7 Modeling calculation of $\delta^{56}\text{Fe}$ and $\text{Fe}^{3+}/\Sigma\text{Fe}$ variations in sub-arc mantle metasomatism. Initial arc magma is assumed to have $\delta^{56}\text{Fe}$ of 0.075‰ by assuming 20% partial melting of a non-buffered mantle source that had previously been depleted by 10% non-buffered melt extraction ([Dauphas et al., 2009](#)), and $\text{Fe}^{3+}/\Sigma\text{Fe}$ of 0.595 (sample NMNH 116852-12 in [Dauphas et al. \(2009\)](#) that might experience the least modification during ascent). Mean values for boninites, island arc basalts and MORBs are from the same references as in Fig. 4. Depleted mantle is assumed to have $\delta^{56}\text{Fe}$ of 0.075‰ and $\text{Fe}^{3+}/\Sigma\text{Fe}$ of 0.019 as estimated by [Dauphas et al. \(2009\)](#). The mantle wedge field is defined according to studies of [Nebel et al. \(2013\)](#) and [Brounce et al. \(2014\)](#).

Fig. 8 $\delta^{56}\text{Fe}$ and $\delta^{26}\text{Mg}$ of the rocks from the Purang ophiolite. The error bars on the individual data points are the 2SD errors. The negative correlation between $\delta^{56}\text{Fe}$ and $\delta^{26}\text{Mg}$ of bulk rocks (except for the dunite sample PL1203-5 and harzburgite PL1212-1, which might have experienced Fe-Mg inter-diffusion) was linearly fitted by a least squares method using the Isoplot program.

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Table 1 Fe and Mg isotopic compositions of the Purang ophiolite in southwestern Tibetan Plateau.

Sample	Rock type	$\delta^{56}\text{Fe}$	2SE	$\delta^{57}\text{Fe}$	2SE	$\delta^{26}\text{Mg}$	2SD	$\delta^{25}\text{Mg}$	2SD
Standard									
BHVO-2		0.101	0.050	0.161	0.063				
BCR-2		0.101	0.045	0.131	0.063				
PCC-1						-0.22	0.05	-0.10	0.04
PCC-1						-0.20	0.07	-0.10	0.05
JB-1						-0.22	0.07	-0.10	0.06
Seawater						-0.77	0.07	-0.40	0.06
Seawater						-0.81	0.07	-0.38	0.06
Seawater						-0.81	0.07	-0.39	0.05
Seawater						-0.80	0.05	-0.44	0.04
PL1212-4	Gabbro	0.114	0.035	0.166	0.083	-0.27	0.07	-0.15	0.06
PL1202-2	Lherzolite	0.014	0.018	0.009	0.083	-0.26	0.05	-0.13	0.04
PL1204-11	Lherzolite	0.022	0.066	0.042	0.083	-0.24	0.05	-0.13	0.04
PL1205-1	Lherzolite	-0.033	0.052	-0.032	0.083	-0.16	0.07	-0.08	0.05
PL1205-3	Lherzolite	-0.100	0.015	-0.169	0.083	-0.16	0.07	-0.08	0.05
PL1205-4	Lherzolite	-0.131	0.062	-0.162	0.083	-0.15	0.07	-0.07	0.05
PL1205-5	Lherzolite	-0.103	0.048	-0.136	0.083	-0.17	0.07	-0.08	0.05
PL1205-6	Lherzolite	-0.028	0.018	-0.034	0.083	-0.17	0.07	-0.12	0.05
PL1206-4	Lherzolite	-0.036	0.039	-0.062	0.083	-0.15	0.07	-0.07	0.05
PL1206-7	Lherzolite	-0.006	0.031	-0.044	0.083	-0.15	0.07	-0.04	0.05
PL1204-9	Harzburgite	0.000	0.056	0.006	0.083	-0.21	0.05	-0.14	0.04
PL1207-2	Harzburgite	0.076	0.019	0.117	0.083	-0.23	0.07	-0.11	0.05
PL1207-3	Harzburgite	-0.032	0.031	-0.080	0.083	-0.21	0.07	-0.10	0.05
PL1207-6	Harzburgite	-0.013	0.045	-0.013	0.083	-0.14	0.07	-0.06	0.05
PL1209-2	Harzburgite	-0.023	0.055	-0.021	0.083	-0.17	0.07	-0.11	0.05
PL1211-3	Harzburgite	0.050	0.044	0.066	0.083	-0.28	0.07	-0.15	0.05
PL1212-1	Harzburgite	-0.209	0.014	-0.342	0.083	-0.27	0.07	-0.13	0.05
PL1212-7	Harzburgite	0.042	0.042	0.035	0.083	-0.28	0.07	-0.16	0.06
PL1203-5	Dunite	0.187	0.047	0.246	0.083	-0.17	0.05	-0.07	0.04
	Replicate	0.180	0.033	0.276	0.083				

Note: $2\text{SE}=[(2\text{SD}_{\text{standard}}/n)^{1/2}]^2+2\text{SE}_{\text{chemical process}}^2]^{1/2}$, where n is the number of replicate analyses.