Magnesium isotopic composition of achondrites

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Abstract

Magnesium isotopic compositions of 22 well-characterized differentiated meteorites including 7 types of achondrites and pallasite meteorites were measured to estimate the average Mg isotopic composition of their parent bodies and evaluate Mg isotopic heterogeneity of the solar system. The $\delta^{26}\text{Mg}$ values are -0.236‰ and -0.190‰ for acapulcoite-lodranite and angrite meteorites, respectively and vary from -0.267‰ to -0.222‰ in the winonaite-IAB-iron silicate group, -0.369‰ to -0.292‰ in aubrites, -0.269‰ to -0.158‰ in HEDs, -0.299‰ to -0.209‰ in ureilites, -0.307‰ to -0.237‰ in mesosiderites, and -0.303‰ to -0.238‰ in pallasites. Magnesium isotopic compositions of most achondrites and pallasite meteorites analyzed here are similar and reveal no significant isotopic fractionation. However, Mg isotopic compositions of D'Orbigny (angrite) and some HEDs are slightly heavier than chondrites and the other achondrites studied here. The slightly heavier Mg isotopic compositions of angrites and some HEDs most likely resulted from either impact-induced evaporation or higher abundance of clinopyroxene with the Mg isotopic composition slightly heavier than olivine and orthopyroxene. The average Mg isotopic composition of achondrites ($\delta^{26}\text{Mg} = -0.246 \pm 0.082\text{‰}$, 2SD, $n = 22$) estimated here is indistinguishable from those of the Earth ($\delta^{26}\text{Mg} = -0.25 \pm 0.07\text{‰}$; 2SD, $n = 139$), chondrites ($\delta^{26}\text{Mg} = -0.28 \pm 0.06\text{‰}$; 2SD, $n = 38$), and the Moon ($\delta^{26}\text{Mg} = -0.26 \pm 0.16\text{‰}$) reported from the same laboratory. The chondritic Mg isotopic composition of achondrites, the Moon, and the Earth further reflects homogeneity of Mg isotopes in the solar system and the lack of Mg isotope fractionation during the planetary accretion process and impact events.

Keywords: magnesium isotopes; achondrites; pallasites; meteorites; isotope fractionation.

1. Introduction

Achondrites and pallasites are stony and stony-iron meteorites with parent bodies that have gone through different magmatic processes because of different conditions such as distinct gravitational fields, source region compositions, heat sources, and time scales of magmatic evolution (e.g., McSween, 1989). Therefore, studies of these meteorites can help to investigate the general planetary differentiation and constrain the degree of isotopic heterogeneity of the solar system. For example, chemical and O isotopic compositions of these different groups of meteorites reflect the origins of different parent bodies and their different differentiation processes (e.g., Clayton and Mayeda, 1996; Mittlefehldt et al., 1998; Mittlefehldt, 2014). Improvements in analytical techniques allowed measurement of mass-dependent isotope fractionation of non-traditional stable isotopes (e.g., Fe, Si, and Zn) caused by protoplanetary disk processes or phase separation during planetary formation and evolution (e.g., Fitoussi et al., 2009; Georg et al., 2007; Kato et al., 2015; Paniello et al., 2012a, b; Polyakov 2009; Wang K. et al., 2012, 2014; Williams et al., 2012) as well as mass-independent fractionation (e.g., Anderson and Sharma, 2006; Dauphas et al., 2014; Regelous et al., 2008) in bulk meteorites.

Magnesium is a major element in all terrestrial planetary objects, stony, and stony-iron meteorites. Large relative mass differences (~8%) between its three isotopes (^{24}Mg , ^{25}Mg , and ^{26}Mg) can potentially produce large mass-dependent isotope fractionation during low-temperature (e.g., Li et al., 2010; Teng et al., 2010a) and high-temperature processes (e.g., Li et al., 2011; Liu et al., 2011; Richter et al., 2007; Wang S.-J. et al., 2014, 2015). Therefore, it can be an excellent tracer for planetary formation and geological processes. In addition, ^{26}Mg is also the decay product of ^{26}Al ($t_{1/2} = 0.72$ Ma) (Lee et al., 1976; 1977) causing mass-independent anomalies, which can be used as a high-precision chronometer and a tracer of isotopic heterogeneity in the early solar system (e.g., Gray and Compston, 1974; Jacobsen et al., 2008; Lee et al., 1976; 1977).

Volatility of Mg (condensation temperature = ~1400 K, Lodders, 2003) is lower than that of moderately volatile elements such as Zn, K and Li (~700 K, 1006 K, and 1140 K, respectively, Lodders, 2003). Studies of Zn isotopes indicate that, compared to the Earth and other HEDs, the Moon and eucrites are enriched in heavy Zn isotopes (Paniello et al., 2012a, b). Paniello et al. (2012a, b) suggested that the Zn isotopic fractionation is due to evaporation during the giant impact and accretion (Paniello et al., 2012a, b). However, studies of Li isotopes found a lack of

isotope fractionation by volatilization in lunar samples and HEDs, similar to the K isotopic homogeneity in the Solar System (Humayun and Clayton, 1995; Magna et al., 2006; 2014). On the other hand, isotopic studies of Si and Fe with volatilities similar to Mg (~1310 K and 1330, respectively, Lodders, 2003) reveal heavier Si and Fe isotopic compositions of some planetary bodies compared to chondrites (Dauphas et al., 2015; Fitoussi et al., 2009; Polyakov 2009; Poitrasson et al., 2004; Pringle et al., 2013, 2014; Wang K. et al., 2012; Weyer et al., 2007; Williams et al., 2012). These isotopic fractionations could have been controlled by metal/silicate segregation during planetary core formation (e.g., Georg et al., 2007; Polyakov 2009; Williams et al., 2012), impact-induced evaporation (Poitrasson et al., 2004; Pringle et al., 2014), planetary differentiation (Weyer et al., 2007), nebular fractionation (Dauphas et al., 2015) or preferential re-melting of isotopically heavy ilmenite during the formation of Stannern-trend eucrites (Wang K. et al., 2012).

Magnesium has a similar volatility to Fe and Si but is not siderophile, hence does not reside in the core, which could result in different behaviors of Mg isotopes during planetary differentiation and accretion processes. Early studies found large Mg isotopic fractionation by volatilization during the formation of calcium-aluminum-rich inclusions (CAIs) and chondrules, the building blocks of asteroids and planets (e.g. Clayton and Mayeda, 1977; Clayton et al., 1988; Galy et al., 2000; Wasserburg et al., 1977; Young et al., 2002). Nevertheless; whether these isotopic variations are preserved during accretion process, and whether Mg isotopes could be fractionated during planetary differentiation at different conditions are still not well-constrained. Though early studies suggest a non-chondritic Mg isotopic composition of the Earth (Wiechert and Halliday, 2007), the more recent comprehensive studies have found similar Mg isotopic compositions for the Earth, the Moon, and chondrites (Bourdon et al., 2010; Chakrabarti and Jacobsen, 2010; Pogge von Strandmann et al., 2011; Schiller et al., 2010b; Sedaghatpour et al., 2013; Teng et al., 2010b). Compared to the Earth, the Moon, and chondrites, our understanding of mass-dependent Mg isotopic behavior in achondrites is limited (Norman et al., 2006; Wiechert and Halliday, 2007; Chakrabarti and Jacobsen, 2010). Wiechert and Halliday (2007) found Mg isotopic composition of eucrites and diogenites similar to those of the Earth and Martian meteorites, but different from chondrites. By contrast, Mg isotopic analysis of achondrites by others suggested similar homogenous chondritic Mg isotopic compositions for the Earth, Mars, Moon, and pallasite parent body (Norman et al., 2006; Chakrabarti and Jacobsen, 2010). In

addition, the average Mg isotopic compositions of terrestrial, lunar, achondrite and chondrite samples reported by Wiechert and Halliday (2007) and Chakrabarti and Jacobsen (2010) are different from those reported by other groups (e.g., Teng et al., 2007, 2010b; Handler et al., 2009; Bourdon et al., 2010; Schiller et al., 2010b; Pogge von Strandmann et al., 2011; Teng et al., 2015a). There are also reported Mg isotopic data for some achondrites that focused on mass-independent fractionation but these have never been discussed in terms of mass-dependent isotopic fractionation during planetary accretion processes (e.g., Baker et al., 2012; Larsen et al., 2011; Schiller et al., 2010; Spivak-Birndorf et al., 2009).

Here, we analyzed 22 achondrites and pallasite meteorites from different groups to estimate Mg isotopic composition of achondrites, to investigate Mg isotope fractionation during different magmatic processes and planetary formation, and to evaluate the extent of Mg isotopic heterogeneity in the solar system. Our results indicate small variations between different achondrites, reflecting mainly their mineralogical differences. Overall, achondrites have similar Mg isotopic compositions to those of the Earth, the Moon, and chondrites, reflecting the homogeneity of Mg isotopes in the inner solar system.

2. Samples

Based on the degree of differentiation, achondrites are classified into two main groups: 1) primitive achondrites with approximately chondritic bulk chemical compositions, but different textures; 2) differentiated achondrites with the parent bodies that underwent large degrees of partial melting and isotopic homogenization, with distinct chemical compositions that are fractionated from chondritic materials (e.g., Krot et al., 2004; Mittlefehldt, 2014). Based on the chemical and O isotopic compositions, primitive and differentiated achondrites are further divided into different subgroups, with each representing different parent bodies (e.g., Clayton and Mayeda, 1996; Mittlefehldt, 2014).

Twenty two meteorite samples including 7 types of achondrites and 2 pallasites were analyzed in this study. These samples cover the whole range of chemical compositions and oxidation states of parent bodies of achondrites (Fig. 1), with different O isotopic compositions. Samples' names, classifications, the fall and find conditions, and MgO contents are listed in Table. 1. The chemical compositions, mineralogy, and petrogenesis of these achondrites are

reviewed in previous studies (e.g., Mittlefehldt et al., 1998; Mittlefehldt, 2014). Nevertheless, a brief description is given below.

2.1. Primitive achondrites

Primitive achondrites are divided into acapulcoite-lodranite, winonaite-IAB-iron silicate inclusion, and Zag (b) groups. Meteorite samples from acapulcoite-lodranite and winonaite-IAB-iron silicate groups are investigated in this study.

2.1.1. Acapulcoite-Lodranite group

Acapulcoites and lodranites are meteorites with chondritic composition but achondritic texture. Oxygen isotopic compositions of acapulcoites and lodranites indicate a heterogeneous single parent body for these meteorites (Clayton and Mayeda, 1996; Greenwood et al., 2012). They are formed from chondritic material that went through different degrees of partial melting and high-temperature metamorphism (e.g., Mittlefehldt et al., 1996; McCoy et al., 1997). Acapulco from this group was analyzed in this study. The major minerals in Acapulco are olivine, orthopyroxene, sulfide and plagioclase with low abundances of chromite, phosphate and clinopyroxene (Palme et al., 1981; Zipfel et al., 1995).

2.1.2. Winonaite-IAB-iron silicate group

Winonaite-IAB-iron silicate group have chondritic compositions with metamorphic textures. The silicate minerals are composed of olivine, orthopyroxene, clinopyroxene, and plagioclase (e.g., Benedix et al., 1998; Mittlefehldt, 2014). The silicate inclusions in IAB iron meteorites are also in the same group with similar composition but with a more diverse mineralogy (e.g., Bild, 1977; Benedix et al., 2000). Meteorites from the winonaite-IAB-iron silicate group have the same chondritic parent body that has gone through limited partial melting, mixing and metamorphism (Bild, 1977; Benedix et al., 1998). Benedix et al. (2000) suggested the formation of IAB-iron silicate group by mixing of the near-surface silicate rocks with the metal from the parent body's interior during a catastrophic impact of a partially differentiated chondritic parent body. Campo del Cielo and Landes from IAB iron meteorites are studied here. Winona from winonaite-IAB-iron silicate group, which is extremely weathered and mainly composed of enstatite with some olivine and minor plagioclase and diopside, was also analyzed (Mason and Jarosewich, 1967; Benedix et al., 1998).

2.2. *Differentiated achondrites*

Differentiated achondrites include angrites, aubrites, brachinites, howardite-eucrite-diogenite group (HED), mesosiderite silicates, ureilites, Itqiy, Ibitira, and Northwest Africa 011 (Mittlefehldt et al., 2014). Meteorites from several groups are analyzed here.

2.2.1. Angrite group

Angrites are crustal igneous rocks, which have been less altered by post-crystallization modification compared to HEDs; therefore, they could potentially record asteroidal igneous processes (Mittlefehldt et al., 2002). This group contains fassaite (Al-Ti-diopside), Ca-rich olivine (kirschsteinite), and anorthite as major minerals (e.g., Mittlefehldt and Lindstrom, 1990; Mittlefehldt et al., 2002). They are formed under oxidizing conditions and are extremely depleted in moderately volatile elements such as Mn, but enriched in incompatible trace element such as REE (e.g., Brett et al., 1977; Mittlefehldt et al., 2002). These meteorites are believed to be partial melts of a differentiated asteroidal source formed within ~2Ma of CAI formation (Keil, 2012). D'orbigny, which is an unmetamorphosed vesicular basaltic lava, and contains 39.4% anorthite, 27.7% fassaite (Al-Ti-diopside-hedenbergite), 19.4% Mg-rich olivine, 11.9% Ca, Fe-rich olivine, 0.5% troilite, 0.5% Ca silicophosphate, and 0.6% spinel, mostly ulvöspinel, was analyzed (Mittlefehldt et al., 2002).

2.2.2. Aubrite group

Aubrites are generally reduced brecciated achondrites with a mineralogy and O isotopic composition similar to enstatite chondrites. They contain mostly (~75-98 vol%) FeO-free enstatite, variable amount of albitic plagioclase, FeO-free diopside, and forsterite (e.g., Keil, 2010). The earliest studies suggested aubrites as nebular materials, but recent studies indicate that they are igneous rocks originally formed during magmatic differentiation (Keil, 2010). Peña Blanca Spring and Bishopville from this group were analyzed for Mg isotopes. These meteorites contain mostly enstatite (75-98%), variable amounts of plagioclase (0.3-16.2%) and minor diopside, and forsterite (Watters and Prinz, 1979; Lonsdale, 1947).

2.2.3. Howardite-Eucrite-Diogenite group

Howardites, eucrites, and diogenites (HEDs) make the largest group of differentiated meteorites from the crust of asteroid 4 Vesta (McCord et al., 1970). The HED parent body has undergone a large scale of melting and extensive differentiation, producing a crust including the outer eucritic crust and the inner diogenitic crust, an ultramafic mantle, and a metallic core (e.g., Yamaguchi et al., 2009). The HEDs include mafic and ultramafic rocks, which are mostly breccias. Homogeneous oxygen isotopic compositions of HEDs reveal a global homogenization event on the parent body (Greenwood et al., 2014). On the basis of chemical compositions and textures, eucrites are divided into two subgroups of basaltic and cumulate eucrites (Mittlefehldt et al., 1998). Basaltic eucrites are breccias that consist of pigeonite and plagioclase with iron-rich pyroxenes. On the basis of major- and trace-element compositions, they are subdivided into Main Group (MG)-Nuevo Laredo (NL) Trend, Stannern Trend (ST), and residual eucrites (e.g., Barrat et al., 2000; Yamaguchi et al., 2009). Basaltic eucrites cooled faster than cumulate eucrites that are unbrecciated coarse-grained gabbros, with pigeonite and plagioclase, and with Mg#s between diogenites and basaltic eucrites. Diogenites are fragmental breccias that contain >90% orthopyroxene with minor chromite and olivine (Mittlefehldt et al., 2002). Howardites are polymict breccias including both monomict eucrites and monomict diogenites (Mittlefehldt et al., 1998). The HED meteorites investigated in this study include six eucrites and three diogenites (Table 1). Oxygen isotopic compositions of Ibitira and Pasamonte suggest Vesta-like asteroid parent-bodies for these two meteorites (e.g., Mittlefehldt, 2014; Scott et al., 2009; Wiechert et al., 2004). Ibitira is recently classified as a new group of differentiated achondrites (Mittlefehldt, 2014). Since Ibitira was originally classified as a basaltic eucrite, we still put this sample under the HED group in Table 1. Pasamonte, which is a polymict breccia with highly unequilibrated basaltic clasts, may record possible fluid-rock interactions on the parent-body (Schwartz and McCallum, 2005).

2.2.4. Mesosiderite silicate group

The silicates from mesosiderites consist of coarse- and fine-grained olivine, low-calcium pyroxene including orthopyroxene and pigeonite, and calcic plagioclase (Mittlefehldt et al., 1998; Mittlefehldt, 2014). Based on silicate composition, mesosiderites are classified into three groups: i) class A that is basaltic and composed of ~24% plagioclase, ii) class B is more ultramafic and consists of more orthopyroxene, and ~21% plagioclase, iii) class C that contains

the lowest amount of plagioclase (~0-5%) (Hewins, 1984; 1988). Based on silicate texture, they are also classified into four grades from lowest metamorphic, grade 1, to the highest metamorphic, grade 4 (Floran, 1978). Mesosiderites are the mixture of crustal and core materials, reflecting their complex forming processes. Oxygen isotopic composition of mesosiderites and their silicate fractions are similar to those of HED meteorites, which suggest asteroid 4 Vesta as the potential parent body of these two groups (Greenwood et al., 2006). Alternatively, mesosiderites are also considered as a result of impact of a naked molten core by a differentiated asteroid, followed by the accretion of this core to the mesosiderites parent body at low velocity (Wasson and Rubin, 1985; Hassanzadeh et al., 1990). In another scenario, the differentiated mesosiderite parent body was disrupted by an impact and then re-accreted (Haack et al., 1996; Scott et al., 2001). Estherville and Crab Orchard were two mesosiderites analyzed in this study (Table 1). Estherville has shown textural grades of 3 and 4 and compositional class of A. Crab Orchard is 1A (Hewins, 1984). Crab Orchard and Estherville contain mainly Opx and Pl with small amount of Cpx and Ol (Prinz et al., 1980).

2.2.5. Ureilite group

Ureilite is the second largest group of achondrites with characteristics of both primitive and differentiated achondrites. For example, the mineralogy and texture of ureilites are similar to ultramafic rocks, but their O isotopic compositions, high trace siderophile element abundances, and planetary type noble gases suggest an origin from primitive materials for this group (Clayton and Mayeda, 1996; Mittlefehldt et al., 1998). Olivine and pyroxene are major minerals in ureilites (Mittlefehldt et al., 1998). The most plausible model for the formation of ureilites is partial-melt residue (e.g., Scott et al., 1993; Warren et al., 2006). Goalpara and Novo-Urei are the ureilites analyzed in this study. Olivine abundances in Novo Urei (67.8%) and Goalpara (63.5%) are higher than pyroxene, which are 30.0% and 31.7%, respectively (Berkley et al., 1980).

2.3. Pallasites

Pallasites are also stony iron meteorites originated from a differentiated asteroid. On the basis of silicate mineralogy, metal compositions, and O isotopic compositions, pallasites are classified into i) main-group, ii) Eagle Station, and iii) pyroxene-pallasite (Mittlefehldt et al., 1998). The

main-group pallasites contain mainly olivine, with minor amount of chromite, low-Ca pyroxene and some phosphates. The Eagle Station pallasites are composed of ~70 to 80 vol% olivine and minor amount of clinopyroxene, orthopyroxene, and chromite. Olivines in this group are more ferroan than the main-group, with higher CaO and lower MnO contents. Meteorites from pyroxene-pallasite group contain ~55-63 vol% olivine, and 1-3 vol% pyroxene with 1-6 mm-sized grains distinguished from the other groups (Boesenberg et al., 1995). The most plausible hypothesis for the pallasite origin is that they are from the core-mantle boundary of their parent bodies (e.g., Mittlefehldt et al., 1998). Composition of the metal phases in main-group and Eagle Station are similar to IIIAB metal and IIF irons, respectively (Scott, 1977; Kracher et al., 1980). Silicates (olivine) from two pallasites, Brahin and Mount Vernon, were analyzed in this study. Both Brahin and Mount Vernon are main-group pallasites with 11.55 and 12% of olivine, respectively (Buseck, 1977).

3. Analytical methods

Magnesium isotopic analyses were performed at the Isotope Laboratory of the University of Arkansas, Fayetteville. Procedures for sample dissolution, column chemistry, and instrumental analyses have been reported in more detail in previous studies but are briefly described below (Teng et al., 2007, 2010b; Yang et al., 2009; Li et al., 2010; Sedaghatpour et al., 2013; Teng and Yang, 2014).

Based on the Mg contents of samples, about 2-4 mg of well-mixed powdered sample was dissolved in order to obtain ~50 μg Mg for high precision isotopic analysis. Each sample was dissolved in a Savillex screw-top beaker in three steps in the mixtures of HF, HCl, and HNO_3 at 160 $^\circ\text{C}$ (For more details see Teng et al., 2007, 2010b and Yang et al., 2009). Purification of Mg was achieved by cation exchange chromatography, using Bio-Rad 200-400 mesh AG50W-X8 resin in 1 N HNO_3 media following previously established procedures (Teng et al., 2007, 2010b; Yang et al., 2009). In order to obtain a pure Mg solution, each sample was processed through the column chemistry twice. Two reference samples were processed through the column chemistry with each batch of achondrites to evaluate the precision and accuracy of our data.

Magnesium isotope measurements were carried out on a Nu Plasma MC-ICPMS in a low-resolution mode, and analyzed by the standard bracketing method (Teng and Yang, 2014). Magnesium isotopes are reported in δ -notation relative to DSM3 in per mil, which is defined as

$\delta^x\text{Mg} = [({}^x\text{Mg}/{}^{24}\text{Mg})_{\text{sample}}/({}^x\text{Mg}/{}^{24}\text{Mg})_{\text{DSM3}} - 1] \times 1000$, where x = mass 25 or 26, and DSM3 is Mg solution made from pure Mg metal (Galy et al., 2003). Full procedural replicate analyses of seawater, Allende and Murchison meteorites as reference materials yielded Mg isotopic compositions that agree with the previously published values (Table S1) (Teng et al., 2010b; Ling et al., 2011). The internal precision on the measured ${}^{26}\text{Mg}/{}^{24}\text{Mg}$ ratio, based on 4 repeat runs of the same sample solution during a single analytical session, is $\leq \pm 0.09\text{‰}$ (2SD; Tables 1, S1, and S2). For replicate and duplicate analyses, weighted average is calculated based on inverse-variance weighted model using Isoplot 3.75-4.15 (Ludwig 2012). 2SD of the weighted average is twice the standard deviation errors propagated from the assigned errors (Tables S1 and S2). The mass-independent anomaly ($\delta^{26}\text{Mg}^*$) in achondrites was calculated by using a slope of 0.514 on a plot of $\phi^{25}\text{Mg}$ vs. $\phi^{26}\text{Mg}$, as explained in Davis et al. (2005). $\delta^{26}\text{Mg}^*$ calculated values for all the meteorites are small (~ 0.000 - 0.068‰) and within our current long term analytical precision.

4. Results

Magnesium isotopic compositions of reference samples, achondrites, and pallasite meteorites are reported in supplementary tables and Table 1. All samples analyzed in this study along with the bulk Earth, chondrites, seawater, and the Moon from the same laboratory fall on a single mass-dependent fractionation line with a best-fit slope of 0.509 (Fig. 2) (Teng et al., 2010b; Ling et al., 2011; Sedaghatpour et al., 2013), which is consistent with previous studies (Young and Galy, 2004; Teng et al., 2010b).

$\delta^{26}\text{Mg}$ values range from -0.267‰ to -0.222‰ in winonaite-IAB-iron silicate group, from -0.369‰ to -0.292‰ in aubrites, from -0.269‰ to -0.158‰ in HEDs, from -0.299‰ to -0.209‰ in ureilites, from -0.307‰ to -0.237‰ in mesosiderites, and from -0.303‰ to -0.238‰ in pallasites (Table S2 and Fig. 3). $\delta^{26}\text{Mg}$ values of acapulcoite-lodranite and angrite meteorites are -0.236‰ and -0.190‰ , respectively (Table S2 and Fig. 3). Allende, Murchison, and seawater samples yielded weighted average $\delta^{26}\text{Mg}$ values of $-0.298 \pm 0.032\text{‰}$ (2SD, $n = 5$, Table S1), $-0.347 \pm 0.032\text{‰}$ (2SD, $n = 3$, Table S1), and $-0.861 \pm 0.036\text{‰}$ (2SD, $n = 4$, Table S1), respectively, which are in agreement with the data reported by Teng et al. (2015a) and Ling et al. (2011).

5. Discussion

In this section, we first discuss Mg isotopic variation within and among different types of achondrites and evaluate the behavior of Mg isotopes during magmatic differentiation of the parent bodies for the groups from which we have analyzed a sufficient number of samples. We then estimate the average Mg isotopic composition of achondrites to evaluate their chondritic origins and implications.

5.1. Magnesium isotopic variation within individual group of achondrites

Isotopic studies of achondrites for some elements (e.g., O, Zn, Fe) indicate isotopic heterogeneity in some groups of these meteorites, which reflects rapid mixing of the interior sources, primary source heterogeneity and/or heterogeneities produced by magmatic differentiation of parent bodies (e.g., Wiechert et al., 2004; Paniello et al., 2012b; Wang K. et al., 2012). For example, heavier Fe isotopic compositions of Stannern-trend (ST) eucrites compared to the other types of eucrites and HEDs are suggested to be the result of magmatic differentiation of the parent body, asteroid 4 Vesta (Wang K. et al., 2012). In contrast to the heavy Fe isotopic compositions of ST eucrites, Magnesium isotopic composition of Bouvante, which is a basaltic ST eucrite, is similar to those of MG (Béréba, Juvinas, and Sioux county) and ungrouped (Pasamonte and Ibitira) eucrites, and diogenites (Bilanga, Johnstown, and Tatahouine). In addition, some eucrites have shown evidence of fluid-rock interactions and metasomatic events on 4 Vesta (e.g., Barrat et al., 2011). Two eucrites among our samples, Sioux County and Pasamonte, have secondary minerals, which may have resulted from pre-terrestrial fluid interactions (Barrat et al., 2011). However, none of these meteorites reveals Mg isotope fractionation compared to the other eucrites, which contrasts with the significant Mg isotope fractionations seen in terrestrial samples produced by chemical weathering and formation of the secondary phases (e.g., Brenot et al., 2008; Teng et al., 2010a; Tipper et al., 2010, 2012; Huang et al., 2012). Mg isotopic compositions of the meteorites from aubrites (Peña Blanca Spring and Bishopville), ureilites (Goalpara and Novo-Urei), mesosiderite silicates (Estherville and Crab Orchard), winonaite-IAB-iron group (Campo del Cielo, Landes, and Winona), and pallasites (Mount Vernon and Brahin) also display no significant Mg isotope fractionation within each individual group, different from the one observed for other isotopes (Wiechert et al., 2004; Wang K. et al., 2012; Paniello, 2012b). Since only two or three meteorites from each group were

analyzed for Mg isotopes, more studies need to be done to investigate Mg isotope fractionation during magmatic differentiation of achondrite parent bodies.

5.2. Magnesium isotopic variation among different groups of achondrites

Magnesium isotopic compositions of most achondrites and pallasite meteorites reveal no significant variations (Fig. 3). However, $\delta^{26}\text{Mg}$ of D'Orbigny (angrite) and some HEDs are slightly heavier than the average isotopic composition of chondrites ($-0.28 \pm 0.06\text{‰}$, 2SD; Teng et al., 2010b) (Fig.3). Magnesium isotopic compositions of different achondrites reported in some geochronological articles also show some variations between achondrites but the discussion of these variations were not in the scope of these studies (e.g., Baker et al., 2012; Larsen et al., 2011; Schiller et al., 2010a; Spivak-birndorf et al. 2009). Compared to the Earth, chondrites, and the other achondrites, slightly heavier Mg isotopic composition in these meteorites could possibly be due to weathering, volatilization during accretion process and/or magmatic differentiation of parent bodies.

5.2.1. Terrestrial weathering

The meteorite finds that have been on the surface of the Earth for several to thousands of years can be subjected to weathering at low temperature with metal and crystalline silicates being the least and the most resistant minerals, respectively (e.g., Weiss et al., 2010). Previous studies reveal Mg isotope fractionation of terrestrial rocks during surface weathering (e.g., Shen et al., 2009; Li et al., 2010; Teng et al., 2010a). Therefore, Mg isotopes in the meteorite finds could be fractionated under severe terrestrial weathering. Nevertheless, our result indicates that the enrichment of heavy Mg isotopes by terrestrial weathering in achondrites is unlikely because all meteorites from winonaite-IAB-iron silicate group including Winona are finds and weathered (e.g., Mason and Jarosewich, 1967) but have Mg isotopic compositions similar to those of achondrite falls (this study), the Earth, and chondrites (Teng et al., 2010b).

5.2.2. Volatilization during planetary accretion or meteorite impact

Experimental and theoretical investigations, and studies of calcium-aluminum-rich inclusions (CAIs) and chondrules reveal significant Mg isotopic fractionations at high temperatures during condensation and evaporation processes (e.g., Clayton et al., 1988; Davis et al., 1990; Richter et

al., 2002; 2007; Young et al., 2002). Hence, the evaporation loss of lighter Mg isotopes during accretion processes or impact events may cause the heavier Mg isotopic composition of the D'Orbigny. However, comprehensive study of lunar samples has revealed similar Mg isotopic compositions for the Moon, Earth, and chondrites, which suggests no Mg isotopic fractionation induced by evaporation during the Moon-forming giant impact (Sedaghatpour et al., 2013). Nonetheless, impact-induced evaporation has been suggested for the heavier Zn isotopic composition of the Moon than the Earth and chondrites (Kato et al., 2015; Paniello et al., 2012a). In addition, volatilization induced by impacts are recorded in Zn isotope composition of enstatite chondrites, ureilites, and HEDs (Moynier et al., 2010, 2011; Paniello et al., 2012b), in Si isotope composition of angrites (Pringle et al., 2014), and probably in Fe isotope composition of angrites (Wang K. et al., 2012). Although Mg isotopes are not fractionated during the Moon-forming giant impact, the possibility of Mg isotope fractionation by volatilization during the accretion process of smaller parent bodies like APB cannot be ruled out. More systematic studies for samples from these groups are needed to further investigate whether or not the slightly heavier Mg isotopic compositions of these meteorites result from the accretion process of their parent bodies.

5.2.3. Magmatic differentiation

Magmatic evolution of parent bodies can be reflected in isotopic compositions of meteorites. Iron isotopic studies of achondrites indicate a chondritic Fe isotope composition for HED achondrites (Zhu et al., 2001; Poitrasson et al., 2004, 2005; Weyer et al., 2005; Wang K. et al., 2012), and heavier Fe isotopic composition for ureilites (Barrat et al., 2015) and angrites (Wang K. et al., 2012). The non-chondritic Fe and Si isotopic compositions of angrites reflect isotopic fractionation by either volatilization during accretion (see 5.1.2.), magmatic differentiation in APB, or nebular fractionation (Dauphas et al., 2015; Pringle et al., 2014; Wang K. et al., 2012). The slightly heavy Fe isotopic composition of ureilites relative to chondrites, together with S depletion in these meteorites, also suggests segregation of S-rich metallic melts in small terrestrial bodies (Barrat et al., 2015).

Magnesium isotopic composition of D'Orbigny (angrite) is slightly heavier than chondrites and is $\sim 0.1\%$ heavier than those of achondrites with the lightest Mg isotopic composition (Fig.3). Although this variation is small, barely beyond our analytical uncertainty, it may be an

indicator of isotope fractionation during magmatic differentiation. Petrological and mineralogical studies of angrites find abundant clinopyroxene (Cpx) in these meteorites (e.g., Boesenberg et al., 1995; Mittlefehldt et al., 2002). By contrast, olivine (Ol) and orthopyroxene (Opx) have high abundances in aubrites (e.g., Mittlefehldt, 2014). These mineral abundances are also reflected on CaO vs. MgO plots for achondrites (Fig. 1). D'Orbigny is plotted in the area with higher CaO content, which reflects more abundances of Cpx in these meteorites (Fig. 1), whereas aubrites are plotted in the area with Ol and Opx chemical compositions. Some HED meteorites with slightly heavier Mg isotopic compositions also have a higher abundance of CaO though they may reflect higher abundances of anorthite (Kitts and Lodders, 1998).

Theoretical and experimental studies of terrestrial rocks and minerals have shown that clinopyroxene is slightly heavier than orthopyroxene and olivine in Mg isotopes ($\sim 0.1\%$) (e.g., Young et al., 2009; Liu et al., 2011; Schauble, 2011; Xiao et al 2013). This slight enrichment of Cpx in $\delta^{26}\text{Mg}$ compared to Ol and Opx is discussed in detail by Liu et al. (2011), Schauble (2011) and Xiao et al (2013) and mainly reflects the slight difference in Mg-O bonding environments between Cpx, Opx and Ol. The higher Mg#s of aubrites reflect a higher abundance of Opx and Ol (consistent with mineral abundances, Watters and Prinz, 1979) with slightly lighter Mg isotopic composition, and lower Mg#s of D'Orbigny may reflect a higher abundance of Cpx with slightly heavier Mg isotopic composition in this meteorite (Fig. 3). This is in agreement with Mg isotopic composition of Ol ($\delta^{25}\text{Mg} = -0.180 \pm 0.210\%$) and Px ($\delta^{25}\text{Mg} = +0.164 \pm 0.064\%$) for D'Orbigny reported by Schiller et al. (2010a) (we use $\delta^{25}\text{Mg}$ for this comparison here to avoid any Mg isotopic anomaly effect but use $\delta^{26}\text{Mg}$ in the rest of paper to be consistent with the literature on mass-dependent Mg isotope fractionation). Clinopyroxene and orthopyroxene abundances in HED meteorites analyzed in this study do not correlate with Mg isotopic compositions of these meteorites (Fig. 4). Compared to D'Orbigny with ~ 27.7 wt.% fassaite (Cpx), these HEDs have lower amounts of Cpx, which may not be enough to affect Mg isotopic compositions of the bulk meteorites (Kitts and Lodders, 1998; Mikouchi and McKay, 2001).

Overall, these chemical and isotopic data may suggest that the small Mg isotopic variations between these achondrite groups could be caused by mineralogical differences produced during magmatic differentiation of their parent bodies. Further work is needed to characterize inter-mineral Mg isotope fractionation during magmatic differentiation of achondrite parent bodies.

5.3. Magnesium isotopic composition of achondrites and its implication

We provide a rough estimate for average Mg isotopic compositions of asteroid 4 Vesta and the other parent bodies of achondrites using the average $\delta^{26}\text{Mg}$ value of samples from each group because we have a limited number of samples from each group. The average $\delta^{26}\text{Mg}$ values are $-0.318 \pm 0.000\text{‰}$ (2SD, $n = 2$) for aubrites, $-0.213 \pm 0.045\text{‰}$ (2SD, $n = 9$) for HEDs, $-0.261 \pm 0.068\text{‰}$ (2SD, $n = 2$) for ureilites, $-0.287 \pm 0.021\text{‰}$ (2SD, $n = 2$) for mesosiderite silicates, $-0.252 \pm 0.018\text{‰}$ (2SD, $n = 3$) for winonaite-IAB-iron silicates, and $-0.285 \pm 0.024\text{‰}$ (2SD, $n = 2$) for pallasites. The average $\delta^{26}\text{Mg}$ value of all achondrites from different groups is $-0.246 \pm 0.082\text{‰}$ (2SD, $n = 22$). These estimated Mg isotopic compositions are between those reported by Wiechert and Halliday (2007) ($\delta^{26}\text{Mg}_{\text{HEDs}} = -0.148$) and Chakrabarti and Jacobsen (2010) ($\delta^{26}\text{Mg}_{\text{Pallasites}} = -0.541 \pm 0.038\text{‰}$ (2SE, $n = 7$)). Although, the cause of this discrepancy is not clear, the most comprehensive data for achondrites in this study, and terrestrial and extraterrestrial samples from the same laboratory (Teng et al., 2010b; Sedaghatpour et al., 2013) allow us to compare Mg isotopic compositions of these materials without being affected by inter-laboratory bias (Teng et al. 2015b).

The average Mg isotopic composition of achondrites ($\delta^{26}\text{Mg} = -0.246 \pm 0.082\text{‰}$, 2SD, $n = 22$) estimated here is indistinguishable from those of the Earth ($\delta^{26}\text{Mg} = -0.25 \pm 0.07\text{‰}$; 2SD, $n = 139$), chondrites ($\delta^{26}\text{Mg} = -0.28 \pm 0.06\text{‰}$; 2SD, $n = 38$), and the Moon ($\delta^{26}\text{Mg} = -0.26 \pm 0.16\text{‰}$) measured in the same laboratory (Teng et al., 2010b; Sedaghatpour et al., 2013), suggesting a homogenous distribution of Mg isotopes in the solar system (Fig. 5). In addition, primitive achondrites that have not undergone well-mixing processes are good records of the early isotopic and chemical heterogeneity in the solar system. These meteorites also have similar Mg isotopic compositions to the differentiated achondrites and chondrites, further supporting the homogeneity of Mg isotopes in the early solar system. This homogeneity weakens the possibility of physical separation and sorting processes of isotopically differentiated chondrules and CAIs in planetary accretion disk processes (Wiechert and Halliday 2007).

Finally, our results can also shed light on Mg isotopic composition of the deep Earth. Magnesium isotopic composition of the Earth is mainly estimated based on samples from the upper mantle (e.g., Teng et al., 2007, 2010b; Yang et al., 2009; Handler et al., 2009; Bourdon et al., 2010; Pogge von Strandmann et al., 2011; Xiao et al. 2013). Pallasites are from the mantle-

core boundary of a planetary object, hence can provide insights into the deep mantle composition of the planetary body. Our results show that these meteorites (which include mainly olivine) have also similar Mg isotopic compositions to those of the Earth's upper mantle and chondrites. This similarity may indicate a similar Mg isotopic composition of the deeper interior portions of the Earth and the upper mantle, and further supports the lack of Mg isotope fractionation during the terrestrial magmatic differentiation processes. Nevertheless, it still needs to consider the possible Mg isotope fractionation induced by phase transformation of olivine to wadsleyite, ringwoodite, bridgmanite and ferropericlase at higher pressure in the Earth's deep interior compared to the smaller parent body of pallasites (Wu et al., 2015).

6. Conclusion

The most comprehensive high precision Mg isotopic data are reported for achondrites including meteorites from 7 different groups and pallasites. The main conclusions are:

1. The $\delta^{26}\text{Mg}$ values range from -0.369‰ to -0.158‰ in all achondrites. In detail, $\delta^{26}\text{Mg}$ values vary from -0.267‰ to -0.222‰ in the winonaite-IAB-iron silicate group, -0.369‰ to -0.292‰ in aubrites, -0.269‰ to -0.158‰ in HEDs, -0.299‰ to -0.209‰ in ureilites, -0.307‰ to -0.237‰ in mesosiderites, and -0.303‰ to -0.238‰ in pallasites. $\delta^{26}\text{Mg}$ values of acapulcoite-lodranite and angrite meteorites are -0.236‰ and -0.190‰, respectively.
2. Overall, the small Mg isotopic variation among achondrites is likely caused by mineralogical differences produced during magmatic differentiation of their parent bodies at different conditions or volatilization during the accretion processes and/or impact events.
3. The average $\delta^{26}\text{Mg}$ value of all chondrites is $-0.246 \pm 0.082\text{‰}$ (2 SD), which is identical to those of chondrites ($\delta^{26}\text{Mg} = -0.28 \pm 0.06\text{‰}$), the Earth ($\delta^{26}\text{Mg} = -0.25 \pm 0.07\text{‰}$), and the Moon ($\delta^{26}\text{Mg} = -0.26 \pm 0.16\text{‰}$) (Teng et al., 2010b; Sedaghatpour et al., 2013).
4. The identical chondritic Mg isotopic compositions of achondrites, the Earth, and the Moon suggest homogenous Mg isotopic distribution in the solar system and the lack of Mg isotope fractionation during accretion disk processes.

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Table 1. Magnesium isotopic compositions of achondrites

Sample	Type	Fall /find	MgO (wt%)	$\delta^{26}\text{Mg}^a$	2SD ^b	$\delta^{25}\text{Mg}^a$	2SD ^b	$\delta^{26}\text{Mg}^*$	2SD ^b
Primitive Achondrites									
Acapulcoite-lodranite									
Acapulco		Fall	26.8	-0.236	0.045	-0.122	0.041	0.002	0.061
Winonaite-IAB-iron silicate									
Campo del Cielo	IAB	Find	15.9	-0.254	0.035	-0.140	0.036	0.037	0.053
Landes	IAB	Find	15.3	-0.260	0.041	-0.105	0.039	-0.057	0.057
Winona	Winonaite	Find	26.7	-0.242	0.051	-0.145	0.038	0.044	0.064
Differentiated Achondrites									
D'Orbigny	Angrite	Find	6.5	-0.190	0.035	-0.100	0.030	0.007	0.046
Peña Blanca Spring	Aubrite	Fall	37.2	-0.318	0.042	-0.164	0.033	0.003	0.054
Bishopville	Aubrite	Fall	34.8	-0.318	0.055	-0.158	0.051	-0.011	0.075
HED									
Bilanga	Diogenite		29.7	-0.230	0.051	-0.112	0.038	-0.008	0.064
Johnstown	Diogenite	Fall	15.3	-0.211	0.033	-0.116	0.021	0.022	0.040
Tatahouine	Diogenite	Fall	27.4	-0.219	0.045	-0.095	0.041	-0.036	0.061
Béréba	Eucrite (MG ^c)	Fall	7.1	-0.212	0.037	-0.121	0.033	0.024	0.051
Sioux county	Eucrite (MG ^c)	Fall	7.2	-0.192	0.040	-0.134	0.034	0.069	0.053
Juvinas	Eucrite (MG ^c)	Fall	7.1	-0.189	0.029	-0.105	0.030	0.022	0.042
Bouvante	Eucrite (ST ^d)	Find	6.4	-0.230	0.042	-0.118	0.034	0.012	0.055
Ibitira	Eucrite (ungrouped)	Fall	7.2	-0.253	0.043	-0.163	0.034	0.044	0.032

Pasamonte	Eucrite (ungrouped)	Fall	6.5	-0.183	0.037	-0.129	0.024	0.066	0.046
Goalpara	Ureilite	Find	36.5	-0.285	0.045	-0.141	0.035	-0.016	0.048
Novo-Urei	Ureilite	Fall	34.4	-0.237	0.039	-0.109	0.025	-0.016	0.048
Estherville	Mesosiderite	Fall	18.8	-0.279	0.065	-0.157	0.077	0.030	0.100
Crab Orchard	Mesosiderite	Find	13.8	-0.294	0.045	-0.163	0.053	0.022	
Mount Vernon	Pallasites (MG ^c)	Find	29.3	-0.293	0.030	-0.122	0.034	-0.052	0.046
Brahin	Pallasites (MG ^c)	Find	25.8	-0.276	0.031	-0.127	0.043	-0.002	0.057

^a Average isotopic composition of replicate and/or duplicate analyses, see supplementary tables.

^b 2SD = 2 times the standard deviation of the population of n (n >20) repeat measurements of the standards during an analytical session.

^c MG = Main group.

^d ST = Stannern Trend.

Reference data: MetBase (version 7.1) and Juvinas, D'Orbigny, Campo del Cielo, Sioux county, Ibitira, Landes, Estherville, Brahmin and Acapulco (Mittlefehldt, 2014; Mittlefehldt et al., 1998); Johnstown, Bilanga and Tatahouine (Barrat et al., 2008); Crab Orchard (Edgerley and Rowe, 1979); Bishopville, Béréba, Pasamonte and Peña Blanca Spring (Urey and Craig, 1953); Novo-Urei (Ringwood, 1960).

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Figure captions

Fig. 1 a) Na/Al ratio, an index of moderately volatile element depletion of parent bodies vs. FeO/MnO ratio, an index of oxidation state of parent bodies in meteorites investigated in this study. b) Variation of CaO versus MgO contents of meteorites studied here as well as olivine (Ol), orthopyroxene (Opx) and clinopyroxene (Cpx) separated from some achondrites. Whole rock data are from references in Table 1 and supplementary Tables. Mineral data are from Mittlefehldt et al. (1998).

Fig. 2 Magnesium three-isotope plot of all achondrites and pallasites analyzed in this study (Table 1), the Earth and chondrites (Teng et al., 2010b), the Moon (Sedaghatpour et al., 2013), and seawater (Ling et al., 2011). The solid line represents a fraction line with a slope of 0.509.

Fig. 3 Variation of $\delta^{26}\text{Mg}$ with Mg# and FeO/MnO in meteorites studied here. The solid line and grey bar represent the average $\delta^{26}\text{Mg}$ of -0.28‰ and two standard deviation of $\pm 0.06\text{‰}$ for chondrites (Teng et al., 2010b). Data are reported in Table 1.

Fig. 4 Orthopyroxene (Opx) and clinopyroxene (Cpx) abundances vs. $\delta^{26}\text{Mg}$ in HED meteorites. Data are from Table 1 and Kitts and Lodders, (1998).

Fig. 5 A summary of Mg isotopic composition of the Earth, Moon, chondrites and achondrites. Data are from Table 1, Sedaghatpour et al. (2013), and Teng et al. (2010b). The vertical dashed line and yellow bar represent the average $\delta^{26}\text{Mg}$ of -0.28‰ and two standard deviation of $\pm 0.06\text{‰}$ for chondrites.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5