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Homogeneous distribution of Fe isotopes in the early solar nebula

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Abstract—To examine the iron (Fe) isotopic heterogeneities of CI and ordinary chondrites, we have analyzed several large chips (approximately 1 g) from three CI chondrites and three ordinary chondrites (LL5, L5, and H5). The Fe isotope compositions of five different samples of Orgueil, one from Ivuna and one from Alais (CI chondrites), are highly homogeneous. This new dataset provides a $\delta^{56}\text{Fe}$ average of $0.02 \pm 0.04\text{‰}$ (2SE, $n = 7$), which represents the best available value for the Fe isotopic composition of CI chondrites and probably the best estimate of the bulk solar system. We conclude that the homogeneity of CI chondrites reflects the initial Fe isotopic homogeneity of the well-mixed solar nebula. In contrast, larger (up to 0.26‰ in $\delta^{56}\text{Fe}$) isotopic variations have been found between separate approximately 1 g pieces of the same ordinary chondrite sample. The Fe isotope heterogeneities in ordinary chondrites appear to be controlled by the abundances of chondritic components, specifically chondrules, whose Fe isotope compositions have been fractionated by evaporation and recondensation during multiple heating events.

INTRODUCTION

The CI chondrites are primitive samples of the solar system that display the closest chemical composition (with the exception of some volatile elements such as H, C, N, O, and noble gases) to the solar photosphere (Anders and Ebihara 1982; Anders and Grevesse 1989; Lodders 2003). As the Sun comprises 99.9% of the mass of our solar system, CI chondrites are the best available samples to estimate the bulk elemental and isotopic composition of our solar system. To date, there are only nine recognized specimens of CI chondrites and only five of them are observed falls: Orgueil (14,000 g), Alais (6,000 g), Ivuna (700 g), Tonk (10 g), and Revelstoke (1 g). Most of the chemical and isotopic compositions reported for CI chondrites come from the same single stone of Orgueil (see Lodders et al. [2009] and references therein). Barrat et al. (2012) have recently reported major and trace elements and Cu and Zn isotopic compositions from six large chips (approximately 1 g each) of five different stones of

Orgueil, from one stone of Alais and one Ivuna stone. This study provides the best available average chemical composition of CI chondrites today. Here, we report the Fe isotopic compositions of the same eight large chips studied by Barrat et al. (2012).

The first and main objective of this study was to precisely determine the average Fe isotope composition of CI chondrites to provide a bulk solar system value for interplanetary comparison. In the past decade, with the application of MC-ICP-MS (multiple collector-inductively coupled plasma-mass spectrometry), high precision Fe isotopic measurements have been routinely performed (e.g., Zhu et al. 2001; Poitrasson et al. 2004; Weyer et al. 2005; Anand et al. 2006; Schoenberg and von Blanckenburg 2006; Dauphas et al. 2009b; Millet et al. 2012) and it has been discovered that planetary materials from different parent bodies have distinct Fe isotope compositions (e.g., Poitrasson et al. 2004; Poitrasson and Freyrier 2005; Weyer et al. 2005; Schoenberg and von Blanckenburg 2006; Dauphas et al. 2009a; Wang et al. 2012a). In particular, terrestrial

oceanic and continental crusts, lunar rocks, and angrites ($\delta^{56}\text{Fe}$ approximately 0.1–0.2‰) are enriched in the heavier isotopes of Fe compared with chondrites, martian, and HED meteorites ($\delta^{56}\text{Fe}$ approximately 0.0‰). Whether these variations are due to distinct solar nebular reservoirs of these planetary bodies or fractionation processes (volatilization, core-mantle segregation, or magmatic differentiation) on these planetary bodies is still a debated issue (cf. Weyer 2008; Poitrasson 2009). The precise determination of the Fe isotopic composition of CI chondrites could help us fully understand where this planetary-scale variation within our solar system originally stems from.

Another goal of this study was to examine the possible heterogeneities of CI and ordinary chondrites. Chondrites are composed of four main components: chondrules, CAIs (calcium-aluminum-rich inclusions), metals, and matrix (Scott and Krot 2005, 2007) and it has been shown that these components have distinct Fe isotopic compositions (Mullane et al. 2005; Theis et al. 2008; Needham et al. 2009; Hezel et al. 2010). The proposed possible causes for the observed Fe isotopic fractionations between chondritic components are that the Fe of these components is (1) inherited from isotopically distinct solar nebular reservoirs; (2) fractionated as a result of nebular evaporation/condensation processing; or (3) fractionated as a result of asteroidal processing on parent bodies, such as aqueous alteration and thermal metamorphism. To better understand the origin of the Fe isotopic variability within chondritic meteorites, we focused on two groups of chondrites: CI and ordinary chondrites, which have very different modal proportions of chondritic components and have experienced contrasting secondary processing effects (aqueous alteration or thermal metamorphism) on their parent bodies.

To this end, we report high precision Fe isotopic compositions of seven individual stones of CI chondrites (five Orgueil, one Ivuna, and one Alais) and several large chips of three ordinary chondrites (L, LL, and H), and discuss the heterogeneities of these early solar system materials and their implications for the Fe isotopic reservoirs in the early solar nebula and on nebular/parent-body processing.

METHODS

Sample Descriptions

Eight large (approximately 1 g each) pieces from three CI carbonaceous chondrites (six Orgueil, one Alais, and one Ivuna) were used in this study. Orgueil,

Alais, and Ivuna are the only three CI chondrite falls with a mass >10 g. The six samples of Orgueil are from five different stones. All the CI chondrites are fine-grained regolith breccias and are composed of more than 95% matrix, lacking chondrules or CAIs (Krot et al. 2007). They have been classified as petrologic type 1 due to almost complete aqueous alteration on their parent body (Scott and Krot 2007). Petrological descriptions as well as major and trace element compositions and Cu and Zn isotopic ratios of these CI samples are reported by Barrat et al. (2012).

In addition, large chips (approximately 3 g each) of three Antarctic ordinary chondrites have been disaggregated to study the possible Fe isotopic heterogeneity of these meteorites. Lewis Cliff (LEW) 85320 is an H ordinary chondrite, Grosvenor Mountains (GRO) 95540 is an L ordinary chondrite, and Dominion Range (DOM) 03194 is an LL ordinary chondrite. All three have been classified as petrologic type 5 due to high-level thermal metamorphism in their parent bodies. Even though the peak temperature could reach more than 700 °C (Huss et al. 2006) for type 5 thermal metamorphism, re-equilibration among large grains is prohibitively slow (e.g., Poitrasson et al. 2005) and will not be considered hereafter. Chondrites GRO 95540 and DOM (DOM) 03194 display “A” grade (minor) weathering. LEW 85320 is designated as having undergone “B” grade (moderate) weathering, as magnesium carbonates (terrestrial weathering products) have been observed (Jull et al. 1988; Grady et al. 1989; Velbel et al. 1991). Terrestrial weathering could affect the Fe isotopic composition of “find” meteorites from hot deserts (Saunier et al. 2010), while Antarctic “find” meteorites are usually considered less altered and better candidates for bulk isotopic cosmochemical studies (Croaz and Wadhwa 2001). We carefully removed all of the fusion crusts and only chose the “fresh” inner parts for analysis, to minimize terrestrial contamination.

Sample Preparation and Chemical Purification of Fe

For all carbonaceous chondrites, large (approximately 1 g) chips of samples were already powdered for the study of Barrat et al. (2012). About 10 mg of the well-mixed powders were used for Fe isotope analysis (see Table 1 for the mass of each sample). For the three ordinary chondrites, large fresh chips of samples were broken into four to seven large chips with a Parafilm-covered hand hammer (see Fig. 1). The mass of each individual chip ranges from several hundred mg to approximately 1 g (Table 1). Each chip was finely ground using an agate pestle and mortar, and no visible large metal grains were

Table 1. Fe isotopic compositions of CI and ordinary chondrites.

Sample name	Type	Fall/find (weathering) ^a	Mass (mg) ^b	Fe (wt%)	$\delta^{56}\text{Fe} \pm 2\text{SE}$	$\delta^{57}\text{Fe} \pm 2\text{SE}$	n ^d	Museum code ^e
Orgueil-1	CI1	Fall	23.2 (1000)	19.0 ^c	0.05 ± 0.06	0.00 ± 0.02	2	MNHN 219239
Orgueil-2	CI1	Fall	17.2 (620)	19.2 ^c	0.02 ± 0.04	0.00 ± 0.04	4	MNHN 222
Orgueil-3	CI1	Fall	17.5 (610)	20.9 ^c	-0.03 ± 0.03	-0.08 ± 0.04	5	MNHN 222
Orgueil-4	CI1	Fall	12.1 (840)	19.4 ^c	0.01 ± 0.07	0.08 ± 0.06	5	MNHN 234
Orgueil-5	CI1	Fall	12.2 (860)	19.2 ^c	0.01 ± 0.03	0.04 ± 0.05	5	MNHN 250
Orgueil-6	CI1	Fall	18.7 (1020)	18.7 ^c	0.07 ± 0.02	0.09 ± 0.05	5	MNHN 237
Orgueil (average) ^f				19.5	0.01 ± 0.04	0.01 ± 0.04		
Alais	CI1	Fall	16.8 (330)	18.6 ^c	0.02 ± 0.01	0.03 ± 0.07	2	MNHN 24
Ivuna	CI1	Fall	12.6 (710)	18.9 ^c	0.04 ± 0.02	0.10 ± 0.06	2	MNHN 3578
CI (average) ^f					0.02 ± 0.04	0.02 ± 0.05		
LEW 85320-1	H5	Find (Be)	14.7 (513)	10.7	-0.06 ± 0.00	-0.10 ± 0.03	3	JSC LEW 85320,45
LEW 85320-2	H5	Find (Be)	24.9 (584)	32.3	0.01 ± 0.05	-0.01 ± 0.03	3	JSC LEW 85320,45
LEW 85320-3	H5	Find (Be)	19.8 (267)	12.0	0.03 ± 0.01	0.09 ± 0.10	2	JSC LEW 85320,45
LEW 85320-4	H5	Find (Be)	17.6 (307)	21.7	0.00 ± 0.04	0.00 ± 0.04	3	JSC LEW 85320,45
LEW 85320-5	H5	Find (Be)	10.8 (410)	29.6	0.05 ± 0.04	0.08 ± 0.06	3	JSC LEW 85320,45
LEW 85320-6	H5	Find (Be)	17.8 (516)	24.6	0.01 ± 0.01	0.00 ± 0.02	3	JSC LEW 85320,45
LEW 85320-7	H5	Find (Be)	9.8 (498)	11.5	-0.01 ± 0.04	-0.01 ± 0.02	3	JSC LEW 85320,45
LEW 85320 (average)				20.3	0.00 ± 0.03	0.01 ± 0.05		
GRO 95540-1	L5	Find (A)	14.3 (657)	4.6	-0.09 ± 0.01	-0.10 ± 0.04	3	JSC GRO 95540,8
GRO 95540-2	L5	Find (A)	12.1 (387)	5.8	0.15 ± 0.01	0.20 ± 0.03	3	JSC GRO 95540,8
GRO 95540-3	L5	Find (A)	20.7 (652)	41.9	0.03 ± 0.02	0.04 ± 0.08	3	JSC GRO 95540,8
GRO 95540-4	L5	Find (A)	23.2 (961)	3.9	0.17 ± 0.03	0.25 ± 0.02	3	JSC GRO 95540,8
GRO 95540-5	L5	Find (A)	8.8 (492)	5.7	-0.06 ± 0.02	-0.05 ± 0.09	3	JSC GRO 95540,8
GRO 95540 (average)				12.4	0.04 ± 0.11	0.07 ± 0.14		
DOM 03194-1	LL5	Find (A)	14.2 (392)	10.4	-0.12 ± 0.02	-0.18 ± 0.03	3	JSC DOM 03194,5
DOM 03194-2	LL5	Find (A)	9.7 (563)	13.6	0.07 ± 0.03	0.11 ± 0.02	3	JSC DOM 03194,5
DOM 03194-3	LL5	Find (A)	13.1 (277)	9.5	-0.05 ± 0.02	-0.07 ± 0.04	3	JSC DOM 03194,5
DOM 03194-4	LL5	Find (A)	11.0 (450)	8.8	-0.06 ± 0.01	-0.11 ± 0.02	3	JSC DOM 03194,5
DOM 03194 (average)				10.6	-0.04 ± 0.08	-0.06 ± 0.12		
Ordinary chondrites (average)					0.00 ± 0.04	0.01 ± 0.06		
BCR-2	Basalt				0.12 ± 0.04	0.19 ± 0.05	4	
BHVO-2	Basalt				0.10 ± 0.02	0.12 ± 0.04	5	

^aThe weathering conditions are from MetBase (version 7.1) and references therein. Weathering index A, B, or C represents “minor,” “moderate,” or “severe” rustiness, respectively. Letter “e” represents evaporite minerals visible.

^bThe numbers in the parentheses are the masses of the well-mixed powders.

^cFe concentration data for the same CI chondrite solutions have been measured and reported by Barrat et al. (2012).

^dNumber of measurements.

^eMNHN = Muséum National d’Histoire Naturelle, Paris. JSC = NASA Johnson Space Center, Houston.

^fOrgueil-6 has different elemental compositions from those of other Orgueil samples (Barrat et al. 2012). It is not included in the calculation of the average.

identified. About 10 mg of the well-mixed powders was digested with concentrated HF/HNO₃ (4:1 v/v) under heat lamps for 3 days. Fully dissolved samples were dried and redigested with double-distilled 6 M HCl.

Anion exchange chromatography for the purification of Fe was applied to all samples following the procedures described by Dauphas et al. (2004, 2009b); such procedures are routinely utilized in our



Fig. 1. Photograph of the Antarctic L5 ordinary chondrite GRO 95540 broken up into five pieces using a Parafilm sealed hand hammer.

laboratory (e.g., Wang et al. 2011, 2012). Samples were loaded onto columns filled with 1 mL AG1-X8 200-400 mesh anion-exchange resin. Matrix elements were eluted in 6 M HCl and Fe was subsequently eluted in 0.4 M HCl. The ion chromatography procedures were repeated for maximum purification. The yield of the procedure is close to 100% (Dauphas et al. 2009b). Two geostandards (BCR-2 and BHVO-2) were subjected to the same chemical treatment and analyzed as external standards to monitor for any fractionation induced due to the chemistry.

Fe Isotope Analyses

The Fe isotopic compositions of samples were measured on a Thermo Scientific NEPTUNE Plus MC-ICP-MS at Washington University in St. Louis. Samples were introduced into the plasma ion source via a 100 μ L/min PFA MicroFlow nebulizer and a cyclonic spray chamber. The intensities of ^{54}Fe , ^{56}Fe , ^{57}Fe were measured on Faraday cups L2, C, and H1, respectively. Isobaric interference of ^{54}Cr and ^{58}Ni was monitored by measuring the intensities of ^{53}Cr and ^{60}Ni on Faraday cups L3 and H4. Measurements were performed on the Fe peak shoulder to avoid polyatomic interferences from $^{40}\text{Ar}^{14}\text{N}^+$, $^{40}\text{Ar}^{16}\text{O}^+$, and $^{40}\text{Ar}^{16}\text{O}^1\text{H}^+$, running the instrument at medium resolution (resolving power $M/\Delta M$ approximately 8500; Weyer and Schwieters 2003). Instrumental mass bias was corrected for using sample-standard bracketing. All results are reported as $\delta^{56}\text{Fe}$ and $\delta^{57}\text{Fe}$, calculated by the following equation.

$$\delta^x\text{Fe} = \left[\frac{(^x\text{Fe}/^{54}\text{Fe})_{\text{sample}}}{(^x\text{Fe}/^{54}\text{Fe})_{\text{standard}}} - 1 \right] \times 1000 \quad (1)$$

In the above equation $x = 56$ or 57 , and IRMM-014 is the standard. Analytical uncertainties are reported as $2 \times$ standard errors (2SE) of repeated

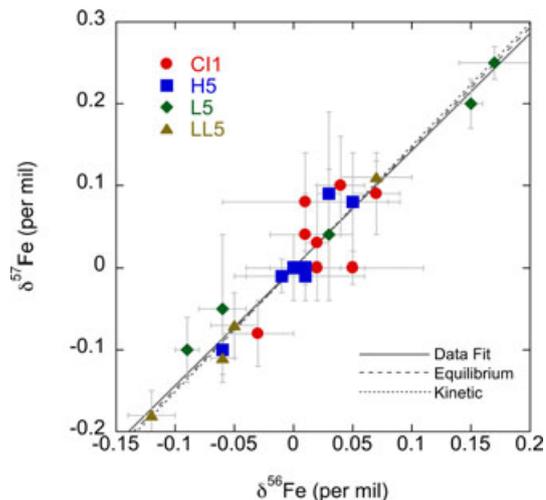


Fig. 2. The bulk Fe isotopic compositions of the CI and ordinary chondrites determined in this study (see also Table 1). The least squares regression of the data, theoretical equilibrium, and kinetic mass-dependent fractionation lines are shown in solid, dashed, and dotted lines, respectively (see Young et al. 2002).

measurements. As shown in Fig. 2, the $\delta^{56}\text{Fe}$ and $\delta^{57}\text{Fe}$ of all of the meteorite samples display an excellent linear correlation (slope approximately 1.41), which is in good agreement with the expected mass-dependent fractionation lines (see Fig. 2), and implies that our samples do not contain mass-independent Fe isotopic anomalies.

RESULTS

All isotopic data are listed in Table 1 and shown in Fig. 2. The Fe isotopic compositions of the two basaltic geostandards BCR-2 ($\delta^{56}\text{Fe} = 0.12 \pm 0.04\text{‰}$) and BHVO-2 ($\delta^{56}\text{Fe} = 0.10 \pm 0.02\text{‰}$) agree well with published data (approximately 0.1; e.g., Poitrasson et al. 2004; Weyer et al. 2005; Schoenberg and von Blanckenburg 2006; Dauphas et al. 2009a; Craddock and Dauphas 2011; Millet et al. 2012). Such analyses suggest that our analytical methods are robust, and provide a good evaluation of the accuracy of Fe isotope analysis in this study.

All of the Orgueil pieces have identical Fe isotopic compositions within error. Orgueil-6 is different from the five others based on its elemental abundances of Zn and REE, as previously reported by Barrat et al. (2012). This difference is probably due to local redistribution of these elements by aqueous fluids on CI parent body (Barrat et al. 2012). Following Barrat et al. (2012), who excluded Orgueil-6 from the calculation of averages, we did not use Orgueil-6 in the calculation of our average Fe isotopic composition. The five other chips from four

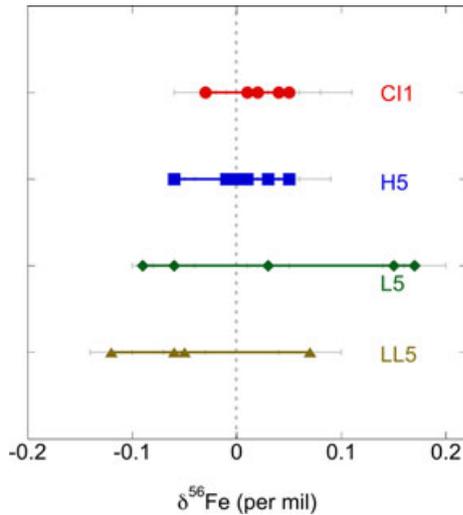


Fig. 3. The ranges of Fe isotopic compositions of CI and ordinary chondrite samples from this study. Error bars (2SE) are shown as gray lines.

different Orgueil stones define an average $\delta^{56}\text{Fe}$ of $0.01 \pm 0.04\text{‰}$ (2SE). This average is consistent with previous studies of one Orgueil stone (e.g., $-0.02 \pm 0.07\text{‰}$; Craddock and Dauphas 2011).

The two other carbonaceous chondrites, Alais and Ivuna, have the same Fe isotopic composition as Orgueil, and all of the CI carbonaceous chondrite data define a very limited range (average $\delta^{56}\text{Fe} = 0.02 \pm 0.04\text{‰}$; range from -0.03 to 0.05‰ ; see Fig. 3). The Fe isotopic composition of CI chondrites is well homogenized. In addition to this isotopic homogeneity, our study confirms that CI chondrites, on average, have $\delta^{56}\text{Fe}$ of approximately 0.00‰ , which agrees with previous reports (Zhu et al. 2001; Kehm et al. 2003; Poitrasson et al. 2004; Schoenberg and von Blanckenburg 2006; Craddock and Dauphas 2011). We propose that $\delta^{56}\text{Fe} = 0.02 \pm 0.04\text{‰}$ is the best value for the average Fe isotopic composition of CI chondrites. This new average for CI chondrites also supports the previous observations that all carbonaceous chondrites (CI, CM, CO, and CV) have indistinguishable Fe isotopic compositions (-0.01 ± 0.01 ; see Craddock and Dauphas [2011] and references therein) within current analytical precisions.

In contrast, the three ordinary chondrites show significantly larger Fe isotopic variations within the same stones (see Fig. 3). Specifically, the different chips of LEW 85320 (H5), GRO 95540 (L5), and DOM 03194 (LL5) define ranges in $\delta^{56}\text{Fe}$ of 0.11‰ , 0.26‰ , and 0.19‰ , respectively. However, the averages of the three ordinary chondrites ($\delta^{56}\text{Fe} = 0.00$) are indistinguishable from carbonaceous chondrites (see Fig. 4), and agree with previous reports (Theis et al.

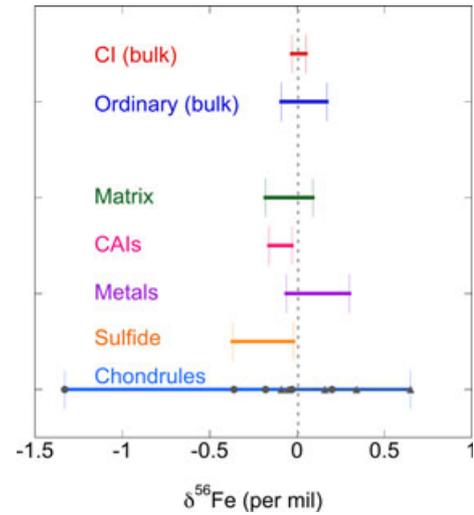


Fig. 4. The ranges of Fe isotopic compositions of chondritic components (chondrules, matrix, CAIs, and metals) and bulk chondrites. The identified type I and type II chondrules are highlighted as triangles and circles symbols, respectively. The data for bulk chondrites are from this study and the data for chondritic components are from literature sources: chondrules (Kehm et al. 2003; Mullane et al. 2005; Needham et al. 2009; Hezel et al. 2010), matrix (Mullane et al. 2005; Hezel et al. 2010), CAIs (Mullane et al. 2005), metals (Theis et al. 2008; Needham et al. 2009; Okabayashi et al. 2012), and sulfide (Needham et al. 2009).

2008; Needham et al. 2009; Craddock and Dauphas 2011). The Fe isotopic variations observed here between different chips of the same ordinary chondrites are as large as the Fe isotopic variation previously observed between different bulk ordinary chondrites (Theis et al. 2008; Needham et al. 2009). Therefore, this variation may be largely a matter of unrepresentative sampling of bulk meteorites and reflect the strongly Fe isotopic fractionated chondritic components (see discussion below). Caution should be taken when interpreting bulk ordinary chondrites when <1 g of homogeneous powder is used.

The three ordinary chondrites studied here are all Antarctic “finds”; however, the Fe isotopic variations between different chips of one sample are unlikely to have been caused by terrestrial weathering effects. First, the three samples used here are only minor to moderately weathered (see Table 1) and we carefully selected the “fresh” interior portions to avoid terrestrial contamination and weathering products. Second, terrestrial weathering effects have been observed only amongst the most weathered hot desert “finds.” Antarctic “finds” were preserved in ice during most of their terrestrial residence and have been less chemically altered compared to hot desert “finds” (Crozas and Wadhwa 2001; Saunier et al. 2010). Third, no Fe

isotopic fractionation due to weathering has been observed amongst Antarctic meteorites (Poitrasson et al. 2004; Saunier et al. 2010). Finally, Saunier et al. (2010) have studied the correlations between weathering condition and $\delta^{56}\text{Fe}$, and they show that even the most weathered (W4 or W5) hot desert meteorites would only be enriched in the heavy Fe isotope by up to 0.07‰ ($\delta^{56}\text{Fe}$) due to the preferential loss of the lighter Fe isotopes to water in the Earth's environment. For the reasons above, we exclude the possibilities of terrestrial weathering/contamination from our following discussion.

DISCUSSION

Fe Isotopic Homogeneity of the Early Solar Nebula

The four stable isotopes of Fe are formed by different nucleosynthesis processes in various stellar environments (Clayton 2003). However, the Fe isotopic compositions of terrestrial and extraterrestrial samples all fall on the same mass-dependent fractionation line in a three-isotope diagram and no nucleosynthetic anomalies remain (see Fig. 2; and Wang et al. 2011). This shows that the different stellar sources of Fe were well homogenized in the early solar nebula prior to the accretion of the parent bodies of the chondrites. Some presolar grains have survived such homogenization and have preserved large isotope anomalies for elements such as C, N, O, Si, and noble gases (e.g., Zinner 2007). Preliminary reports have shown that some presolar grains have non-solar Fe isotopic compositions (Floss et al. 2008; Marhas et al. 2008; Ong et al. 2012). However, except for a few FUN (fractionated and unidentified nuclear effects) CAIs from the Allende CV3 chondrite (Voelkening and Papanastassiou 1989), Fe isotopic anomalies have not been observed in either bulk or stepwise leaching of primitive and differentiated meteorites (Dauphas et al. 2008; Wang et al. 2011). Furthermore, all of the carbonaceous and ordinary chondrites, including the data from this study (Zhu et al. 2001; Kehm et al. 2003; Poitrasson et al. 2004; Schoenberg and von Blanckenburg 2006; Craddock and Dauphas 2011), have the same average bulk Fe isotope compositions ($\delta^{56}\text{Fe}$ approximately 0.00‰) within analytical error, even if they have different chemical compositions (CI, CM, CO, CV, LL, L, and H) and have experienced distinct secondary alteration processes on parent bodies (petrological types 1–7). All of these pieces of evidence indicate an efficient mixing of Fe in the starting materials in the early solar nebula, without large secondary isotopic fractionation effects on parent bodies such as aqueous alteration and thermal metamorphism. The average composition of CI

chondrites, calculated from six different chips from Orgueil, together with samples from Ivuna and Alais ($\delta^{56}\text{Fe} = 0.02 \pm 0.04\text{‰}$), represents the best estimate of the Fe isotopic composition of our solar system.

Fe Isotopic Fractionation Between Chondritic Components in a Solar Nebula Setting

Even though on average CI and ordinary chondrites have indistinguishable Fe isotopic compositions ($0.02 \pm 0.04\text{‰}$ versus $0.00 \pm 0.04\text{‰}$; see Table 1), ordinary chondrites show larger isotopic heterogeneity within one sample than CI. This is most likely due to variable contents of their different components (chondrules, metals, and matrix, etc.), which are isotopically distinct for Fe. Chondrules and matrix are the two main components of chondrites; together they make up more than 90% of chondrites by volume (Scott and Krot 2005) with CAIs, metals, and sulfides as minor components in most chondrite groups. In detail, different groups of chondrites have variable amounts of these components. For example, CI chondrites are composed of more than 95 vol% matrix and less than 5 vol% chondrules, while ordinary chondrites comprise 60–80 vol% chondrules, 10–15 vol% matrix, and approximately 10 vol% metals (Scott and Krot 2007). In early literature, due to the larger analytical uncertainties of ion microprobe techniques in comparison with MC-ICP-MS, no resolvable Fe isotopic variations between chondritic components were detectable (e.g., Alexander and Wang 2001). However, with the recent development of MC-ICP-MS and a large improvement in analytical precision, Fe isotopic variations between chondrules, matrix, CAIs, and metals have since been resolved (Kehm et al. 2003; Mullane et al. 2005; Theis et al. 2008; Needham et al. 2009; Hezel et al. 2010; Okabayashi et al. 2012). The isotopic ranges of such components are as follows (all data are ‰): chondrules $-1.33 < \delta^{56}\text{Fe} < +0.65$, matrix $-0.18 < \delta^{56}\text{Fe} < +0.09$, CAIs $-0.16 < \delta^{56}\text{Fe} < -0.03$, metals $-0.06 < \delta^{56}\text{Fe} < +0.30$, and sulfide $-0.37 < \delta^{56}\text{Fe} < +0.02$. As shown in Fig. 4, the degree of isotopic variation within each chondritic component is significant.

Of the chondritic components, chondrules display the largest variation in Fe isotope compositions. The refractory type I chondrules are isotopically heavier in Fe than the less refractory type II chondrules (Mullane et al. 2005): type I chondrules $\delta^{56}\text{Fe} = -0.05\text{‰}$ to $+0.65\text{‰}$; average $0.20 \pm 0.11\text{‰}$; type II chondrules $\delta^{56}\text{Fe} = -1.33\text{‰}$ to 0.20‰ ; average $-0.34 \pm 0.10\text{‰}$. Chondrules were formed by multiple stage heating events in the early solar nebula (Scott and Krot 2005), wherein it has been proposed that refractory type I

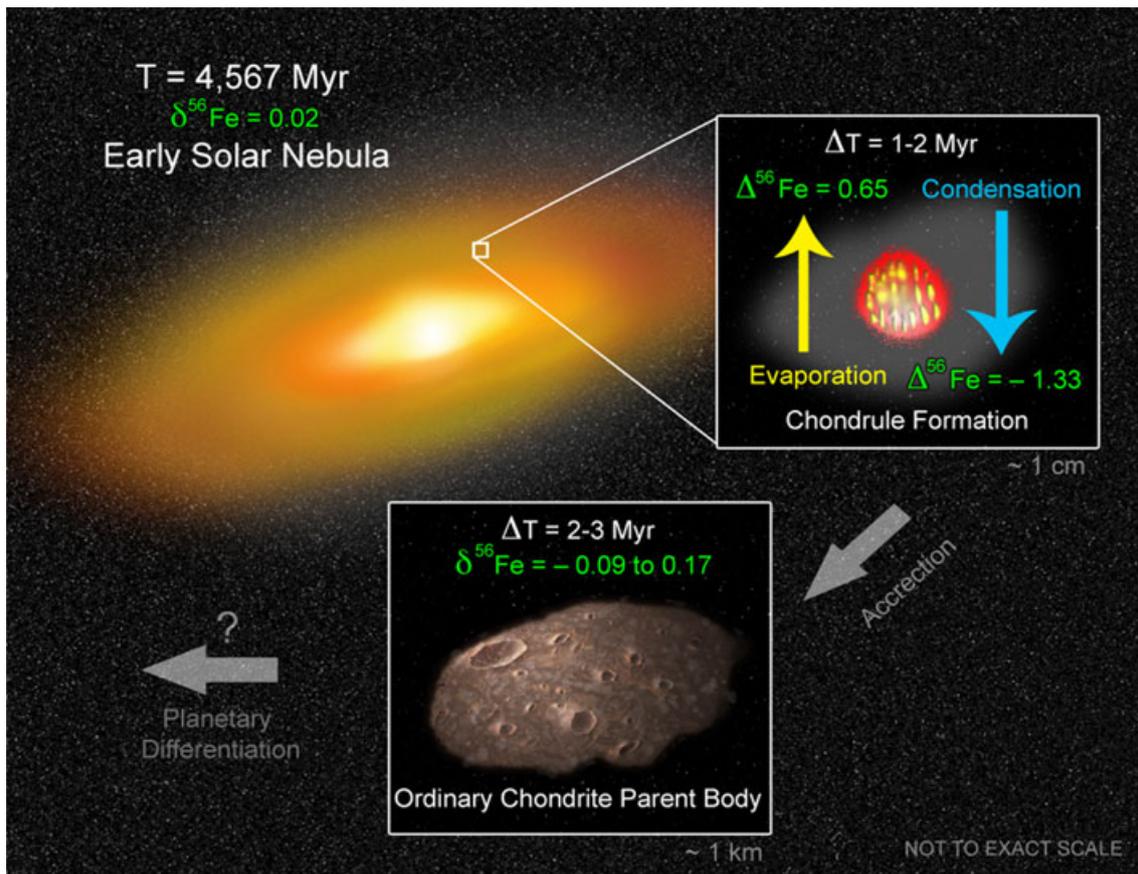


Fig. 5. Cartoon showing Fe isotopic composition ($\delta^{56}\text{Fe}$) evolution in the early solar system.

chondrules represent partial evaporation residues of volatile type II chondrules (Jones 1990; Alexander 1994); alternatively, type I and type II chondrules were formed as their precursors interacted (through evaporation/condensation) with surrounding nebular gas (Hewins and Zanda 2012). Partial loss of Fe during a high-temperature heating event should enrich the surrounding gas in the lighter isotopes and enrich the residue in the heavier isotopes. Recondensation of these Fe isotopically light vapors could therefore produce isotopically light chondrules (rims). Even though the degree of fractionation observed is still smaller than the expectation of free evaporation/condensation under traditional Rayleigh conditions (Alexander and Wang 2001), nebular processing, evaporation, and/or recondensation could modify the Fe isotopic composition of chondrules. The heterogeneous distribution of chondrules between chondrite groups should therefore result in larger isotopic heterogeneities within individual ordinary chondrites (which are chondrule rich), compared with CI chondrites (which are chondrule poor; see Figs. 4 and 5).

The Fe isotopic variations in both metals and sulfides are not as large as those in chondrules; however, because metal and sulfide phases have the highest Fe concentrations in chondrites, the abundance of metals and sulfides has an important control on the bulk Fe isotope compositions. Metals in chondrites are generally enriched in the heavier Fe isotopes (Theis et al. 2008; Needham et al. 2009; Okabayashi et al. 2012), while typically, sulfides in chondrites are lighter (Needham et al. 2009), which agrees well with the observation of metals and sulfides in iron meteorites and pallasites (Poitrasson et al. 2005; Williams et al. 2006) and also the theoretical prediction of preferential partitioning of heavy/light Fe isotopes into metal/sulfide phases (Polyakov and Mineev 2000; Polyakov and Soultanov 2011). The iron isotope compositions ($\delta^{56}\text{Fe}$) of metals separated from H, L, and LL ordinary chondrites have been reported as 0.09‰, 0.20‰, and 0.28‰, respectively (Theis et al. 2008), while no systematic variation between metals in H, L, and LL ordinary chondrites have been found in Needham et al. (2009).

Using mixing calculations of the various chondritic components, we can model the bulk Fe isotopic

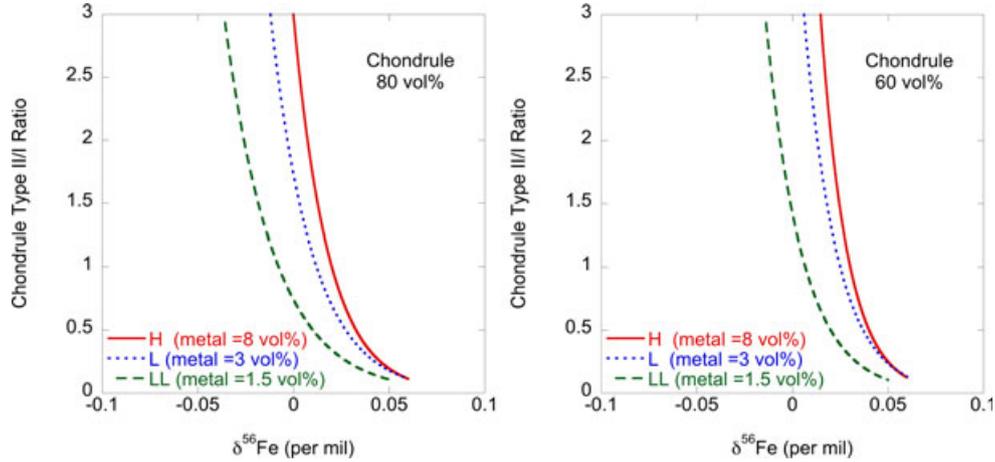


Fig. 6. Modeling of the bulk Fe isotopic compositions of ordinary chondrites by mixing chondritic components with varying chondrule type II/I ratios.

Table 2. Parameters and values used in the modeling of the bulk Fe isotopic compositions of ordinary chondrites by mixing chondritic components.

Parameter	Value	Reference
Type I chondrule $\delta^{56}\text{Fe}$ (‰)	+0.20	(Mullane et al. 2005)
Type II chondrule $\delta^{56}\text{Fe}$ (‰)	-0.34	(Mullane et al. 2005)
Metal $\delta^{56}\text{Fe}$ (‰) for H, L, and LL	0.09, 0.20, 0.28	(Theis et al. 2008)
Matrix $\delta^{56}\text{Fe}$ (‰)	~0	(Mullane et al. 2005; Hezel et al. 2010)
Chondrule density (g cm^{-3})	~3	(Hughes 1980)
Pure iron density (g cm^{-3})	7.875	(Lodders and Fegley 1998)
Bulk density (g cm^{-3}) for H, L and LL	3.44, 3.40, 3.29	(Wilkison and Robinson 2000)
Type I and II chondrule Fe concentration (wt%)	0.91, 9.87	(Jones and Scott 1989)
Metal Fe concentration (wt%)	~100%	Pure iron metal for simplification
Bulk Fe concentration (wt%) for H, L, and LL	27.2, 21.8, 19.8	(Lodders and Fegley 1998)

compositions of ordinary chondrites (see Fig. 6). Chondrules, metals, and matrix are the only three components considered in ordinary chondrites for the purpose of simplification. CAIs are minor components for ordinary chondrites (<0.1 vol%; Scott and Krot 2007), and are poor in Fe; therefore, they were neglected in our calculations. All the parameters and

values used in our models are listed in Table 2 and the results are shown in Fig. 6. Three major factors control the chondrite bulk Fe isotopic compositions of ordinary chondrites, which agrees with the discussion above, which are: (1) chondrule abundance, (2) chondrule type, and (3) metal abundance. Our calculations indicate that up to 0.1‰ $\delta^{56}\text{Fe}$ variations (between -0.05‰ and +0.05‰; see Fig. 6) in bulk meteorites could be explained simply by these three factors. Chondrule abundance in ordinary chondrites can vary between 60 and 80 vol% (Scott and Krot 2007). Higher chondrule abundance is usually correlated with lower bulk $\delta^{56}\text{Fe}$ when other factors are unchanged.

Chondrule type proportion is another important variable controlling bulk isotopic compositions. Refractory type I chondrules are enriched in Fe heavy isotopes, while volatile type II chondrules are depleted (Mullane et al. 2005). Type I chondrules are Fe-poor, whereas type II chondrules are very Fe-rich (Jones and Scott 1989), although it has been recently found that the Fe content in chondrules is possibly a continuum rather than a bimodality (Berlin 2009). A high proportion of Fe-rich type II chondrules in chondrites could strongly affect bulk composition and efficiently drag bulk $\delta^{56}\text{Fe}$ to negative values (see high type II/I ratio scenarios in Fig. 6). The amounts of type I and II chondrules are highly variable between H, L, and LL ordinary chondrites. On average, the type II/I ratios (v/v) are 0.8, 2.1, and 2.8 for H, L, and LL ordinary chondrites respectively, but there is considerable overlap between each group (Zanda et al. 2006). By using the average ratios of type II/I, we can predict the bulk Fe isotopic compositions for H (0.02‰ to 0.03‰), L (-0.01‰ to 0.01‰), and LL (-0.04‰ to -0.02‰). These small differences between the three ordinary chondrite groups predicted by our model are at the

limit of our current analysis precision, and have not yet been observed (i.e., Needham et al. 2009).

In addition, metal abundance appears to play an important role in controlling bulk Fe isotope composition. From LL to L and H ordinary chondrites, metal abundance increases from 1.5 to 3 and 8 vol% (Scott and Krot 2007). Because the metal abundances appear to correlate with chondrule type II/I ratios (Zanda et al. 2006), these two factors are coupled in controlling bulk Fe isotope composition. For example, H ordinary chondrites have high metal abundances (high $\delta^{56}\text{Fe}$) and also high type II/I chondrule ratios (low $\delta^{56}\text{Fe}$). This antithetic effect shown in our model is very important to the bulk Fe isotope composition and could explain why, on average, no resolvable systematic variations of bulk $\delta^{56}\text{Fe}$ between H, L, and LL ordinary chondrites have been observed in previous studies (i.e., Needham et al. 2009) and also why on average ordinary chondrites have the same bulk Fe isotopic composition as CIs.

Given that the abundances of chondritic components can be highly variable between different samples and can significantly control bulk Fe isotope compositions, care should be taken when interpreting isotopic data acquired using small mass samples, as sampling bias could become an issue (isotopically light or heavy chondrules, or metal grains, etc.). Even large chips (approximately 1 g) from the same ordinary chondrites have $\delta^{56}\text{Fe}$ which can vary up to 0.26‰ in $\delta^{56}\text{Fe}$. These isotopic variations within one individual sample are comparable to the bulk variations measured within the whole ordinary chondrite group (e.g., Needham et al. 2009; Craddock and Dauphas 2011). Nonetheless, this will not change the fact that on average ordinary and CIs have indistinguishable bulk Fe isotopic composition as observed in this and previous studies, which suggests that Fe isotopes were well homogenized in the early solar system.

CONCLUSIONS

In this study, we report the most complete Fe isotope data set of CI chondrites using large sample masses. Thus, we provide the best estimate of Fe isotopic composition for CI chondrites ($\delta^{56}\text{Fe} = 0.02 \pm 0.04\text{‰}$). We propose that this average represents the bulk Fe isotopic composition of our solar system.

Different approximately 1 g chips of CI chondrites display a very limited range of Fe isotope compositions, while several approximately 1 g chips from the same ordinary chondrites show significantly larger variations. This appears to be the result of different modal proportions of chondritic components between CI and ordinary chondrites. Amongst all of the main chondritic

components, chondrules have the largest Fe isotopic variation because of fractionation during evaporation and recondensation in the solar nebular setting. By employing mass balance calculations, we have shown that chondrule abundance and type, in addition to metal abundance, are the main parameters controlling the bulk Fe isotopic compositions of the different ordinary chondrites. CI chondrites do not show large isotopic heterogeneity because they are composed mainly of matrix, with only a small (<5%) proportion of chondrules; this is not the case for ordinary chondrites, which have greater modal abundances of chondrules.

Ordinary chondrites exhibit large Fe isotopic heterogeneity within 1 g chips of individual samples (up to 0.26‰). This suggests that large samples are needed to obtain accurate bulk compositions, to avoid sampling bias (caused by natural, uneven distributions of large chondrules or metal grains, and/or by incomplete mixing during sample preparation). For CI chondrites, which contain dominantly matrix and lack chondrules and other chondritic components, small masses of samples should be enough to obtain a homogeneous bulk composition. This is serendipitous, given the low abundance of CI chondrites as a group.

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