Enstatite chondrites EL3 as building blocks for the Earth: the debate 1 over the 146 Sm-142 Nd systematics
Maud Boyet, A. Bouvier, P. Frossard, Tahar Hammouda, M. Garçon, A. Gannoun

To cite this version:
Maud Boyet, A. Bouvier, P. Frossard, Tahar Hammouda, M. Garçon, et al.. Enstatite chondrites EL3 as building blocks for the Earth: the debate 1 over the 146 Sm-142 Nd systematics. Earth and Planetary Science Letters, Elsevier, 2018, 488, pp.68-78. <10.1016/j.epsl.2018.02.004>. <hal-01723716>

HAL Id: hal-01723716
https://hal-clermont-univ.archives-ouvertes.fr/hal-01723716
Submitted on 5 Mar 2018

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
Enstatite chondrites EL3 as building blocks for the Earth: the debate over the $^{146}$Sm-$^{142}$Nd systematics

M. Boyet$^1$, A. Bouvier$^2$, P. Frossard$^1$, T. Hammouda$^1$, M. Garçon$^3$, A. Gannoun$^1$

1 Laboratoire Magmas et Volcans, Université Clermont Auvergne, France
2 Department of Earth Sciences, Centre for Planetary Science and Exploration, University of Western Ontario, London, Canada
3 Department of Earth Sciences, ETH Zurich, Switzerland


Keywords: $^{146}$Sm-$^{142}$Nd systematics, enstatite chondrites, Earth’s evolution, Earth’s building blocks

Abstract

The $^{146}$Sm-$^{142}$Nd extinct decay scheme ($^{146}$Sm half-life of 103 My) is a powerful tool to trace early Earth silicate differentiation. Differences in $^{142}$Nd abundance measured between different chondrite meteorite groups and the modern Earth challenges the interpretation of the $^{142}$Nd isotopic variations found in terrestrial samples because the origin of the Earth and the nature of its building blocks is still an ongoing debate. As bulk meteorites, the enstatite chondrites (EC) have isotope signatures that are the closest to the Earth value with an average small deficit of ~10 ppm in $^{142}$Nd relative to modern terrestrial samples. Here we review all the Nd isotope data measured on EC so far, and present the first measurements on an observed meteorite fall Almahata
Sitta containing pristine fragments of an unmetamorphosed enstatite chondrite belonging to the EL3 subgroup. Once $^{142}\text{Nd}/^{144}\text{Nd}$ ratios are normalized to a common chondritic evolution, samples from the EC group (both EL and EH) have a deficit in $^{142}\text{Nd}$ but the dispersion is important ($\mu^{142}\text{Nd} = -10 \pm 12$ (2SD) ppm). This scatter reflects their unique mineralogy associated to their formation in reduced conditions (low fO$_2$ or high C/O). Rare-earth elements are mainly carried by the sulfide phase oldhamite (CaS) that is more easily altered than silicates by weathering since most of the EC meteorites are desert finds. The EL6 have fractionated rare-earth element patterns with depletion in the most incompatible elements. Deviations in Nd mass independent stable isotope ratios in enstatite chondrites relative to terrestrial standard are not resolved with the level of analytical precision achieved by modern mass spectrometry techniques. Here we show that enstatite chondrites from the EL3 and EL6 subgroups may come from different parent bodies. Samples from the EL3 subgroup have Nd ($\mu^{142}\text{Nd} = -0.8 \pm 7.0$, 2SD) and Ru isotope ratios undistinguishable from that of the Bulk Silicate Earth. EL3 samples have never been analyzed for Mo isotopes. Because these enstatite chondrites are relatively small in size and number, they are usually not available for destructive isotopic measurements. Average values based on the measurement of EL6 samples should not be considered as representative of the whole EL group because of melting and thermal metamorphism events affecting the Sm/Nd ratios and prolonged open-system history. The EL3 chondrites are the best candidates as the Earth’s building blocks. These new results remove the need to change the composition of refractory incompatible elements early in Earth’s history.

1. Introduction
Variations in mass independent stable isotope ratios, also called nucleosynthetic anomalies, of refractory elements have become extremely useful to trace the origin of the different planetary bodies and their relative contribution into the building process of terrestrial planets (e.g., Warren, 2011). Enstatite chondrites appear to be the closest in isotope composition to terrestrial samples and carbonaceous chondrites the furthest. Enstatite chondrites and the Earth are undistinguishable for many isotope systems, i.e. Cr, Ti, O, Ni (see Dauphas, 2017 for a recent review), meaning that with the current level of precision no resolvable difference is observed between both materials. A small difference in Si isotopes does exist and this has been used against the Enstatite Chondrite Earth model (Fitoussi and Bourdon, 2012) but Si isotopes may have been fractionated during nebular processes and their isotope ratios may not faithfully reflect the initial composition of the starting material (Savage and Moynier, 2013; Zambardi et al., 2013; Dauphas et al., 2015). The only resolvable differences between enstatite chondrites and the Earth were reported for Mo and Ru isotope compositions (Burkhardt et al., 2011; Fischer-Gödde and Kleine 2017). The enrichment in s-process Mo for the Earth relative to chondrites suggests that the composition of the Earth cannot be reconstructed by any known combination of meteorites because it is itself an end-member in the mixing trend (Burkhardt et al., 2011). The initial composition of the Earth has become an issue for the use and interpretation of the $^{146}$Sm-$^{142}$Nd short-lived isotopic systematic in meteorites and Earth rocks. An early fractionation of the Sm/Nd ratio during the lifetime of $^{146}$Sm ($T_{1/2}=103$ Ma) would modify the $^{142}$Nd/$^{144}$Nd signatures by radiogenic production. In 2005, the first high-precision $^{142}$Nd measurements in chondrites revealed a difference of about 20 ppm with the modern terrestrial samples (Boyet and Carlson, 2005). Because both Sm and Nd are
lithophile elements under terrestrial redox conditions (Bouhifd et al., 2015), a
differentiation event of the silicate Earth in the first 30 Ma of Solar System history
has been proposed to explain this offset. One type of model considers that the bulk
Silicate Earth (BSE) preserves a relative chondritic abundance of refractory lithophile
elements. The early-depleted mantle reservoir (high Sm/Nd) served as the source for
magmatism since its formation whereas the enriched reservoir remains isolated from
mantle convection and is still untapped (Boyet and Carlson, 2005). Other scenarios
proposed that the complementary enriched reservoir (low Sm/Nd) has been lost to
space by collisional erosion of planetesimals (Caro et al., 2008; O’Neill and Palme,
2008) leaving the Bulk Earth depleted in incompatible elements and with a higher
Sm/Nd ratio relative to the well-constrained average composition of whole-rock
chondrites from the carbonaceous (CC), ordinary (OC) and enstatite (EC) chondrite
groups (Bouvier et al., 2008).

These models have been challenged by the discovery of variations in mass
independent stable Sm and Nd isotope ratios in whole-rock chondrites and their
components (Andreasen and Sharma, 2006; Carlson et al., 2007; Gannoun et al.,
2011; Brennecka et al., 2013, Bouvier and Boyet, 2016; Burkhardt et al., 2016). The
isotope $^{142}$Nd is mainly produced by $s$-process but has a small contribution of $p$-
process, estimated to be lower than a few percent (Arlandini et al., 1999; Bisterzo et
al., 2011). The parent isotope, $^{146}$Sm is a pure $p$-process nuclide so any variation in
the $p$ production would affect the abundance of its daughter isotope $^{142}$Nd as well.
Although we have undisputable evidences for different contribution of $s$-process
isotopes in Solar System materials and measurable variations in the $^{142}$Nd/$^{144}$Nd
ratios, the origin of the terrestrial $^{142}$Nd signature compared to the chondrite
meteorites is still unclear. The isotope signature of some ocean island basalts is
difficult to interpret without an early event that would fractionate the Sm/Nd ratio of
the bulk silicate Earth very early on in its history (Jackson et al., 2010). Here we re-
evaluate the small isotopic variations measured in enstatite chondrites. Such
meteorites were formed under reducing conditions and have a particular mineral
assemblage. Importantly for interpreting their Sm-Nd systematics, half of the rare
earth element (REE) budget is hosted in oldhamite (CaS), which is easily altered to
Ca-sulphate by weathering (Crozaz and Lundberg, 1995; Gannoun et al., 2011).

We have compiled all the Nd isotope data published on EC so far. We also
present the first measurements on an observed fall Almahata Sitta containing
fragments of the EL3 subgroup, such as the fragment MS-177. The fireball of
Almahata Sitta fall was captured on camera in northern Sudan on October 7th 2008.
Its trajectory was back-tracked to a small asteroid called 2008 TC₃ (Bischoff et al.,
2010). Additionally, REE concentration measurements are presented for samples
belonging to the two subgroups of enstatite chondrites (EH and EL). Since the
identification of $^{142}$Nd variations attributed to nucleosynthetic anomalies requires a
comparison at the same Sm/Nd evolution, we present in detail two methods of
calculations used for normalization of all chondrites to a common Sm/Nd and the
difficulties inherent to the enstatite chondrite sub-groups. The $^{142}$Nd/$^{144}$Nd ratios are
then compared to the corresponding mass independent stable Nd isotope ratios for
each meteorite to establish their significance with respect to the distribution of
nucleosynthetic anomalies amongst the chondrite groups. Finally, their potential
contributions as Earth’s building blocks are discussed.
2. Samples and Methods

2.1. Trace element contents in EC

Rare earth element concentrations have been measured for 12 EC from EH (4) and EL (8) sub-groups. The meteorite names and sample subgroups are reported in Table 1. Two techniques of dissolution have been used: 1) samples were digested in PFA Savillex® beakers using a mixture of concentrated HF-HNO₃ acids in 3:1 proportion at 120°C on hot plate during 48h; 2) samples were digested in individual Parr® pressure vessels in concentrated HF-HNO₃ (10:1) for 5 days at 155°C. In both cases, after drying down the samples, perchloric acid was added and evaporated on a hot plate to break down the fluorides. After successive dissolution/evaporation cycles in concentrated HNO₃, samples were taken back into solution in PFA Savillex® beakers or in Parr® pressure vessels in 6M HCl. All measurements were performed using the Agilent 7500 quadrupole ICP-MS at the Laboratoire Magmas et Volcans (LMV) in Clermont-Ferrand, with the exception of Almahata Sitta MS-177 and Hvittis that were measured at the University Western Ontario using a Thermo iCAP Qc quadrupole ICP-MS. Both ICP-MS are equipped with He collision cells. Samples were measured in HNO₃ 0.5M- HF 0.05 M and HNO₃ 0.3M- HF 0.01 M in the two laboratories, respectively. Counts per second were calibrated using rock standards BCR-2, BHVO-2 or BIR 1a. The rock standards and a synthetic standard solution (CMS) were routinely analyzed to validate the efficiency of the digestion procedures and the accuracy and precision of the acquired data. Blank and standard solutions were measured every 4 samples. Both the external reproducibility estimated from
repeated measurements of the standards and the accuracy checked from certified rock standard analyses are lower than 5%.

2.2. $^{146,147}$Sm-$^{142,143}$Nd systematics and mass independent stable Nd isotopes

Here we report new results for MS-177 (Table 2). After the Parr bomb digestion of the samples at the University of Western Ontario, following the methods detailed in Bouvier and Boyet (2016), an aliquot of about ~80% was used for isotopic composition, ~10% fraction was spiked with an enriched $^{149}$Sm-$^{150}$Nd mixed spike to determine precisely the Sm/Nd ratios by isotopic dilution, and another 10% aliquot was used to determine Q-ICPMS REE concentrations. The Sm and Nd of the spiked fractions were purified and separated using a two-stage chemistry procedure (cationic AG50W-X8 and Ln-spec resins). This procedure was also used for a BCR-2 USGS basalt standard (aliquot of 1.1 mg) to ensure accuracy and reproducibility of our spiked and unspiked measurements. Both spiked Sm and Nd isotopic fractions were analyzed using a MC-ICPMS Neptune Plus at LMV.

The separation procedure for the Nd isotopic composition required 3 steps as described in Garçon et al. (2018). The Ce was separated from other trivalent REE using the oxidation of Ce to Ce$^{4+}$ on Ln-Spec resin (Tazoe et al., 2007; Li et al., 2015). Then the separation between Nd and Sm was achieved on Ln-Spec resin using 0.20N to 0.25N HCl. Total procedural blanks are generally lower than a few tens of pg for Nd. The Nd isotopic compositions were measured as Nd$^+$ using the LMV Triton Plus™ Multicollector Thermal Ionization Mass Spectrometer. For MS-177, Nd
isotope ratios were obtained in static mode following the method described in Gannoun et al. (2011a). Static measurements were preferred because the amount of Nd was small (~300 ng for MS-177) and dynamic routines require large sample size (e.g. 750 ng for the 4-line acquisition scheme developed by Garçon et al., 2018). In addition, we used a mass spectrometer equipped with new faraday collectors (6 month-old). No sign of deterioration of the faraday collectors was identified during the measurements. Ce and Sm contributions, monitored at masses 140 and 147 respectively, were lower than 3 ppm on masses 142 and 144 for both standard and sample measurements. Nd isotope ratios were corrected for mass fractionation to $^{146}\text{Nd}/^{144}\text{Nd}=0.7219$ using the exponential mass-fractionation law. The external reproducibility (2SD) calculated from 6 repeated measurements of the JNd1 standard is 5.6 ppm on $^{142}\text{Nd}/^{144}\text{Nd}$, 7.4 ppm on $^{143}\text{Nd}/^{144}\text{Nd}$, 4.4 ppm on $^{145}\text{Nd}/^{144}\text{Nd}$, 8.2 ppm on $^{148}\text{Nd}/^{144}\text{Nd}$, and 10.9 ppm on $^{150}\text{Nd}/^{144}\text{Nd}$ (Table 2).

3. Results

The new REE data are reported in Table 1 and compared to published values from Gannoun et al. (2011b) and Barrat et al. (2014) in Figure 1. Data from Kalleymen and Wasson (1986) is not shown in Figure 1 because they reported the concentrations of only 5 REEs (La, Sm, Eu, Yb, and Lu), which is insufficient to define proper REE patterns. All EH samples have flat REE patterns with concentrations very similar to those reported in CI, except Kota Kota (EH3) that is enriched in LREE (Figure 1A). This split is the most visibly weathered in color and texture of our collection, and the meteorite was described as a strongly weathered EC by Leitch and Smith (1982). The measured sub-chondritic $^{143}\text{Nd}/^{144}\text{Nd}$ ratio of Kota
Kota suggests that this sample has a complex history with several episodes of fractionation of the Sm/Nd ratio (Gannoun et al., 2011b). Two EH samples display unusual REE patterns: Galim (b) and LAP 0225 (Figure 1A, data from Barrat et al. 2014). They are classified as impact melt (IM) and their REE patterns are in agreement with the petrological description of the samples (Michel-Lévy and Bourot-Denise, 1988; Russel et al., 2004). EL samples present more variable REE distributions. Most of EL3 samples display flat REE patterns with concentrations ranging between 0.6 to 2 times the CI values. MAC 88180 is more enriched in REE (x2 CI) and has a negative Eu anomaly (Eu/Eu* = Eu_{CI}/(Sm_{CI}^{1/2} x Gd_{CI}^{1/2}) of 0.7 (Figure 1C). Similar REE patterns have been obtained following the two dissolution techniques. Almahata Sitta MS-177, which is the most depleted EL3 sample, has a positive Eu anomaly of 1.5. The REE pattern for MS-177 is more similar to Adrar Bous (EL5) with lower REE contents and large positive Eu anomalies (Eu/Eu* = 1.3).

EET 92063 has a REE pattern with similar characteristics but REE contents are even lower and the Eu positive anomaly is higher (Figure 1D). This sample is described as highly weathered and classified in an anomalous EL6 group by Barrat et al. (2014). By contrast, all other EL6 samples display negative Eu anomalies and are called “normal EL6” by the same authors. EL6 samples are all depleted in LREE relative to HREE ((La/Lu)_N) up to 0.62. Chemical heterogeneities are observed within the same sample. For example Hvittis has been measured 4 times and Eu/Eu* varies from 0.8 to 1.0 depending on the split analyzed. Petrological observations show that all these samples are strongly heterogeneous at millimeter to centimeter scale. Rare earth element distribution in the different EC subgroups is presented in Figure 2. This time, the database for REE in enstatite chondrites from Kallemyen and Wasson (1986) is included. All subgroups have similar Yb concentrations but different (La/Sm)_N ratios.
Strong depletions in incompatible elements, hence low La/Sm ratios, are measured in EL6 samples and the two samples characterized by an unusual mineralogical assemblage (Galim (b) and LAP 0225). Then EL6 samples are clearly different from other EL samples. We note that the La/Sm ratios in EH are more variable and in average higher than those measured in EL3-5 samples.

Measured Nd isotope ratios for MS-177 are expressed in ppm deviation relative to the reference JNd-1 standard (µ-value) in Table 2 and all details on measurements (isotope ratios, intensity, interferences, etc) are available in Supplementary information (Table S2). This sample presents a small deficit of -11 ppm in $^{143}\text{Nd}/^{144}\text{Nd}$ whereas all stable isotope ratios are identical within errors to the JNd-1 standard values. The $^{147}\text{Sm}/^{144}\text{Nd}$ ratios calculated using the isotope dilution method are $^{147}\text{Sm}/^{144}\text{Nd} = 0.1381$ for the BCR-2 standard and 0.1897 for MS-177. Our repeated spiked Sm-Nd measurements of BCR-2 standard at LMV give an average of $^{147}\text{Sm}/^{144}\text{Nd} = 0.1381 \pm 0.0003$ (2SD, n=4), thus a precision of 0.2%. This value is within literature values which range from $^{147}\text{Sm}/^{144}\text{Nd} = 0.1357$ to 0.1396 and identical within errors with the most recent determination by ID TIMS at 0.1379 ±0.0002 (2SD; Li et al., 2012 and references therein). The initial $^{143}\text{Nd}/^{144}\text{Nd}$ ratio for MS-177 calculated at 4.568 Ga is $\varepsilon^{143}\text{Nd} = +0.06 \pm 0.25$ when normalized to the initial for the Solar System composition using the chondrite average (CHUR) $^{147}\text{Sm}/^{144}\text{Nd}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ ratio values (Bouvier et al., 2008).

4. Discussion

4.1. $^{146,147}\text{Sm-142,143}\text{Nd}$ systematics in enstatite chondrites
The purpose of this paper is to take a more rigorous look at enstatite chondrites and their genetic relationship to the Earth. To compare the compositions of enstatite chondrites to other chondrite groups, we compiled all Nd data published on chondrites from this group so far, as well as new data. This compilation is provided in supplementary materials. Available data comes from 7 different publications: Boyet and Carlson (2005); Rankenburg et al. (2006); Carlson et al. (2007); Gannoun et al. (2011); Bouvier and Boyet (2016); Burkhardt et al. (2016); and Fukai and Yokohama (2017). For the studies using La Jolla Nd standard for the calculation of deviation in ppm relative to the terrestrial value (\(\mu\)Nd), the data were recalculated relative to the JNd1 terrestrial standard. The two synthetic standards La Jolla and AMES record significant mass-dependent variability and then cannot be used as a reference either for the BSE or the modern mantle (O’Neil et al., 2008; Gautam et al., 2017; Saji et al., 2016). All the measurements have been obtained on large-size samples (1-2 g). The Nd isotope compositions obtained for the Allende CV3 chondrite using different methods of digestion (table-top acid-digested, Parr bomb acid-digested and alkali-fused acid-digested samples) are similar (Boyet and Carlson, 2005; Carlson et al., 2007). The complete digestion of refractory components may be an issue for some elements but not for Nd. Presolar SiC grains are a primary carrier of large Ba isotope anomalies and the destruction of these grains during the digestion is required (Bermingham et al., 2016). When measuring the Nd isotope composition of extra-terrestrial materials, an additional consideration is to take into account the potential neutron capture effects due to the interaction of materials with galactic cosmic rays. However previous studies have measured very small deviations in the \(^{149}\)Sm abundance of chondrites which is one of the Sm isotopes having the largest thermal neutron capture cross section. This suggests that chondrites have not been
significantly affected by neutron fluence effects and that the correction on $^{142}\text{Nd}/^{144}\text{Nd}$ ratios is negligible or within the measurement precision (e.g. Boyet and Carlson, 2005; Bouvier and Boyet, 2016). McCoy-West et al. (2017) have determined the mass-dependent stable Nd isotope composition using a double spike technique. Chondrites and terrestrial samples have identical isotopic compositions with $\delta^{146}\text{Nd}/^{144}\text{Nd} = -0.025 \pm 0.025\%$ (2SD; n=39) and $\delta^{146}\text{Nd}/^{144}\text{Nd} = -0.022 \pm 0.034\%$ (2SD; n=30) respectively. These results imply that no bias is produced by the correction of instrumental mass fractionation effects when processing the raw data.

The first high-precision $^{142}\text{Nd}$ measurements on chondrites have shown that differences in $^{142}\text{Nd}/^{144}\text{Nd}$ ratios existed among chondrite groups and that these variations were not correlated with changes of Sm/Nd ratios (Boyet and Carlson 2005). The new compilation combining all data measured on chondrites since 2005 leads to the same conclusion. The different groups of chondrites have different measured $^{142}\text{Nd}/^{144}\text{Nd}$ values (Figure 3A). We note that results obtained from different chondrite fragments which were processed in different labs using different analytical techniques are in good agreement. On average carbonaceous chondrites (CC) have a $\mu^{142}\text{Nd}$ of -31 ± 20 (2SD) ppm, ordinary chondrites (OC) of -16 ± 9 (2SD) ppm and EC of -12 ± 21 (2SD) ppm. Although standard deviations overlap, two sample t-tests strongly suggest that the three populations of chondrites are distinct from each other with p-values lower than 0.05 (p-value CC-OC = 1.5 x $10^{-8}$; p-value CC-EC = 8.9 x $10^{-10}$; p-value OC-EC = 0.004). Five samples have $^{147}\text{Sm}/^{144}\text{Nd}$ ratios far from the CHUR value of 0.1960 (dashed line in Figure 4A and 4B; Bouvier et al., 2008): NWA 2090 (CO3) and Tagish Lake (C2-ungrouped, not represented in the figure: $^{147}\text{Sm}/^{144}\text{Nd}$=0.148) for CC, Adrian (H4) for OC (not represented in the figure:
Sm/\(^{144}\)Nd = 0.137), and Kota Kota and Blithfield for EC. Moreover one EC sample displays an extreme \(^{142}\)Nd value and this measurement has a large error bar (Abee: -44 \pm 17 (2SE) ppm (Boyet and Carlson 2005). When all these samples are excluded from the averaging calculation, CC have a \(^{142}\)Nd of -33 \pm 16 (2SD) ppm, OC of -16 \pm 9 (2SD) ppm and EC of -11 \pm 12 (2SD) ppm. All these groups have similar measured Sm/Nd ratios even if a larger range is observed for EC (Figure 4A, B).

When the same samples are selected, the mean \(^{147}\)Sm/\(^{144}\)Nd ratios of each group is 0.1959 \pm 0.0046 (2SD) for CC, 0.1946 \pm 0.0033 (2SD) for OC and 0.1950 \pm 0.0084 (2SD) for EC. The median values are equal to 0.1953, 0.1951 and 0.1952 for these 3 groups, respectively. CC and OC have normal distributions whereas EC present a larger scatter. These values are slightly lower than the CHUR value of 0.1960 \pm 0.0004 (2SD) defined from the average of unequilibrated chondrites (Bouvier et al., 2008) but are similar within uncertainties. Two sample t-tests further confirm that the mean \(^{147}\)Sm/\(^{144}\)Nd ratios of the three populations are undistinguishable from each other since p-values are much higher than 0.05 (p-value CC-OC = 0.9; p-value CC-EC = 0.3; p-value OC-EC = 0.3).

The measured abundance of \(^{142}\)Nd comes from two distinct contributions: its initial abundance and the quantity produced with time by the decay of \(^{146}\)Sm. Since \(^{146}\)Sm was alive only during the first 500 Ma of Solar System history, we can write the following equations:

\[
^{142}\text{Nd}_{\text{meas}} = ^{142}\text{Nd}_{T0} + ^{146}\text{Sm}_{T0} \quad (1)
\]

\[
(^{142}\text{Nd}/^{144}\text{Nd})_{\text{meas}} = (^{142}\text{Nd}/^{144}\text{Nd})_{T0} + (^{146}\text{Sm}/^{144}\text{Nd})_{T0} \quad (2)
\]
where \( m_{\text{eas}} \) denotes the abundance or isotopic composition measured today in chondrites and \( t_0 \) the initial abundance or isotopic composition at the time of formation.

These equations show that any fractionation of the Sm/Nd ratio during the \(^{146}\text{Sm}\) lifetime will significantly affect the measured \(^{142}\text{Nd}/^{144}\text{Nd}\) ratios. In order to compare initial \(^{142}\text{Nd}/^{144}\text{Nd}\) ratios between chondrites, the data are thus corrected from the \(^{142}\text{Nd}\) radiogenic production since the formation of the Solar System and normalized to a common chondritic evolution following the CHUR parameters defined by Bouvier et al. (2008). Two different methods of calculation can be used assuming a modification of the Sm/Nd ratio either early in the Solar system history or once the \(^{146}\text{Sm}-^{142}\text{Nd}\) systematics is extinct.

**Method A.** \(^{142}\text{Nd}/^{144}\text{Nd}\) corrected using measured \(^{147}\text{Sm}/^{144}\text{Nd}\) ratios

This correction is illustrated in Figure 5A. The \(^{142}\text{Nd}/^{144}\text{Nd}\) ratio of the sample is calculated back in time to the age of the Solar System formation, i.e. 4.568 Ga (Bouvier and Wadhwa, 2010) using the measured \(^{147}\text{Sm}/^{144}\text{Nd}\) ratio. From this initial \(^{142}\text{Nd}/^{144}\text{Nd}\) ratio, a modern value is recalculated using the CHUR \(^{147}\text{Sm}/^{144}\text{Nd}\) ratio of 0.1960 (Bouvier et al., 2008). Such a correction assumes that the modification of the Sm/Nd ratio occurred at the beginning of Solar System history i.e. when the chondrites form.

**Method B.** \(^{142}\text{Nd}/^{144}\text{Nd}\) corrected using measured \(^{143}\text{Nd}/^{144}\text{Nd}\) ratios

This correction, illustrated in Figure 5B, does not consider the measured \(^{147}\text{Sm}/^{144}\text{Nd}\) ratio but instead the integrated \(^{147}\text{Sm}/^{144}\text{Nd}\) ratio calculated from the \(^{147}\text{Sm}-^{143}\text{Nd}\) long-lived radiogenic systematics. Since recent changes of Sm/Nd ratios, for example
due to weathering, are not recorded in the evolution of $^{143}\text{Nd}/^{144}\text{Nd}$ ratios, the $^{147}\text{Sm}/^{144}\text{Nd}$ ratio is deduced from the measured $^{143}\text{Nd}/^{144}\text{Nd}$ ratios considering that all samples evolved from the same initial $^{143}\text{Nd}/^{144}\text{Nd}$ value of 0.506686 which corresponds to the composition of the CHUR at 4.568 Ga (Bouvier et al., 2008).

Then, the normalized-CHUR $^{142}\text{Nd}/^{144}\text{Nd}$ ratio is calculated following the steps described in Method A using the $^{147}\text{Sm}/^{144}\text{Nd}$ ratio of the CHUR from 4.568 Ga to present-day.

To evaluate the influence of the chosen method on the CHUR-normalized $\mu^{142}\text{Nd}$ values, we re-corrected all published $^{142}\text{Nd}/^{144}\text{Nd}$ ratios with the two methods and compared the results in Figure 3B and C. Note that the Sm/Nd ratios of OC and CC from Fukai and Yokoyama (2017) were measured by quadrupole ICPMS and are thus reported with a much lower precision (1-1.5%) than that typically obtained (about 0.2%) by isotope dilution with MC-ICPMS measurements of the spiked Sm and Nd isotopic compositions. Such a large uncertainty on Sm/Nd ratios propagates to errors of about 10 ppm on the CHUR-normalized $\mu^{142}\text{Nd}$ values. Fukai and Yokoyama (2017) did not need to determine the precise $^{147}\text{Sm}/^{144}\text{Nd}$ ratios of their samples because they used the method B to calculate their normalized $\mu^{142}\text{Nd}$ values. However, we encourage future studies to systematically measure both the Nd isotope composition and the corresponding Sm/Nd ratio at high precision using the isotope dilution method.

Weighted averages for CC, OC and EC are $-32 \pm 13$ (2SD), $-15 \pm 9$ (2SD), $-8 \pm 15$ (2SD) for the method A and $-32 \pm 13$ (2SD), $-16 \pm 9$ (2SD), and $-10 \pm 12$ (2SD) for the method B. Mean values are identical within errors but when looking at the data
in further details, significant differences are identified within the EC group. This is illustrated in Figure 4C where both methods of correction are compared. For CC and OC, all $^{142}\text{Nd}/^{144}\text{Nd}$ ratios plot along the 1:1 line whereas the EC show a larger scatter. This scatter indicates that changes in the Sm/Nd ratios of EC have occurred at different times, probably early in the Solar System history and recently by weathering. The larger dispersion of the Sm/Nd ratios observed in EC may be explained by their unique mineralogy as they formed under extreme reducing conditions. Larimer and Bartholomay (1979) suggested that EC formed within a region in the solar nebula with a high C/O ratio (>1). Although rare earth elements are mainly carried by silicate minerals in other chondrite groups, about half of the Sm and Nd elemental budget of EC is hosted in oldhamite, which is a Ca-sulfide (CaS). In-situ measurements indicate that CaS are very rich in REE relative to CI chondrites (10 to 100 times the CI values; Crozaz and Lundberg, 1995; Gannoun et al., 2011b). In EH3, the CaS have fractionated REE patterns with an enrichment in LREE relative to HREE ($(\text{La}/\text{Yb})_N = 1-15$) and positive anomalies in Eu and Yb (Crozaz and Lundberg, 1995; Gannoun et al., 2011b). Although in-situ REE measurements of CaS were conducted only in EH3 samples, the measurements carried out in other groups of EC and in aubrites (reduced enstatite achondrites) show diverse REE patterns which may be associated with the weathering of CaS (Floss et al., 1990; Floss and Crozaz, 1993; Crozaz and Lundberg, 1995). Petrological observations have shown that CaS are not uniformly distributed in enstatite chondrites from EH and EL subgroups. In EH, CaS is mainly included in metal-sulfide nodules even if it can be present in the matrix or in chondrules dominated by silicate phases (El Goresy et al., 1988; Gannoun et al., 2011b; Piani et al., 2016). Our observations of EL3 samples suggest that CaS is mostly found in chondrules where it is intimately intergrown with enstatite and
occasionally with diopside (El Goresy et al., 2017). Its occurrence in metal-dominated nodules is less common. Although metal-sulfide nodules are more subject to oxidation/weathering than silicate assemblages, CaS enclosed in metal should be better preserved because metal has no fracture veins like silicates. No clear relationship has been found between REE fractionation and the type of samples (group, fall vs. found, etc). To minimize the amplitude of the correction on $^{142}$Nd/$^{144}$Nd ratios and reduce the errors caused by a potential Sm/Nd fractionation during weathering, we selected a subset of EC samples (11 samples, 16 measurements) with measured $^{147}$Sm/$^{144}$Nd ratios very close to the CHUR value i.e. between 0.1926 and 0.1994 (±1.7%). Within this range, the correction reaches a maximum of ± 5 ppm on the $^{142}$Nd/$^{144}$Nd ratios, i.e. equivalent to the average external reproducibility. Taking only this subset of EC samples yields an average $\mu$ $^{142}$Nd of -10 ± 12 (2SD) for the two methods A and B, hence does not change the mean nor improve the standard deviation obtained for the whole EC group.

4.2. Stable Nd isotope signature of EC relative to other groups

Neodymium has 7 isotopes from mass 142 to 150. Nucleosynthetic anomalies, which are mass-independent variations, can be identified by measuring 3 stable isotope ratios: $^{145}$Nd/$^{144}$Nd, $^{148}$Nd/$^{144}$Nd and $^{150}$Nd/$^{144}$Nd. Both $^{142}$Nd/$^{144}$Nd and $^{143}$Nd/$^{144}$Nd are modified by radiogenic productions from $^{146}$Sm and $^{147}$Sm respectively, and $^{146}$Nd/$^{144}$Nd is used for internal mass bias correction and normalization to a common ratio value of 0.7219. Except $^{142}$Nd which is mainly produced by s-process and $^{150}$Nd which is a pure r-process nuclide, all other isotopes
are formed by a combination of \textit{s-} and \textit{r-processes} in different stellar environments (Arlandini et al., 1999; Bisterzo et al., 2011). A change in the -s/-r proportions thus modifies the Nd isotope patterns. Excess in \textit{s-process} produces excess in $^{142}\text{Nd}$ and deficits in $^{143}\text{Nd}$, $^{145}\text{Nd}$, $^{148}\text{Nd}$ and $^{150}\text{Nd}$ with the deficits increasing from mass 143 to mass 150. Measurements obtained on individual EC are presented in Figure 6. The average values for both CC and OC groups are shown for comparison. For clarity, errors bars (2SD) are not shown in the figure but are instead provided in the figure caption. The large errors calculated for CC reflect the variability of \textit{s-process} nuclides among this group owing to the contribution of CAIs. In a $\mu^{150}\text{Nd}$ vs $\mu^{148}\text{Nd}$ binary plot, bulk CC and separated CAIs plot on the mixing line calculated from isotope production in stellar models (see Fig. 3 in Bouvier and Boyet, 2016). CAIs can be the carrier of large \textit{s-process} excess signature with deficits in $\mu^{148}\text{Nd}$ and $\mu^{150}\text{Nd}$ of several tens of ppm (Brennecka et al., 2013; Marks et al., 2014; Bouvier and Boyet, 2016).

The compositions of EC are scattered in the 3 binary plots without any clear correlation (Figure 6). Results of mixing of the \textit{s-model} prediction with the terrestrial composition are indicated by the dashed lines depending on the model used: 1) stellar model by Arlandini et al. (1999) or 2) using the composition of SiC as representative of a pure \textit{s-process} component (Hopp and Ott, 1997). Deficits in $^{142}\text{Nd}$ measured in enstatite chondrites from the EH group and the EL6 subgroup are small and equal to -11 ppm in average. For such small deficit, the expected deviations in 145, 148 and 150-Nd are equal to +5, +11 and +17 ppm, respectively. These variations remain very small in comparison to the measurement precision (external reproducibility) obtained with the last generation of multi-collector thermal ionization mass spectrometers.
(TIMS). In the recent study of Burkhardt et al. (2016), both $^{142}\text{Nd}/^{144}\text{Nd}$ and $^{148}\text{Nd}/^{144}\text{Nd}$ ratios were measured using a dynamic procedure leading to better external reproducibilities than static measurements. The relative deterioration of the Faraday collectors and their change of efficiency through time are important sources of imprecision and inaccuracy for the determination of isotope ratios using static multicollection (Makishima and Nakamura, 1991; Thirlwall, 1991). However, even with dynamic procedures on TIMS, the external reproducibilities for $^{148}\text{Nd}/^{144}\text{Nd}$ reach 11 and 15 ppm on repeated measurements of the BHVO-2 rock standard and JNd-1 standard, respectively. Better precisions have been recently reported by Saji et al. (2016) using multi-collector inductively coupled plasma mass spectrometry and a sample-standard bracketing technique. Five measurements of the carbonaceous chondrite Allende (CV3) give an external reproducibility of 1 ppm on the $^{148}\text{Nd}/^{144}\text{Nd}$ ratio with intensity of 14 V at mass 142. Because enstatite chondrites are about 2 times more depleted in Nd than CV3 carbonaceous chondrites, this technique would require large quantities of EC samples (i.e. 2 g). A new 4-mass-step acquisition scheme developed by Garçon et al. (2018) by thermal ionization mass spectrometry is very promising because this routine is able to return all Nd isotope ratios dynamically and provide a robust evaluation of the data to assess the quality of the data (e.g., mixing effects on the filament, fractionation between acquisition steps).

4.3. The isotope similarity between EL3 samples and the Earth

Amongst all the classified meteorites, enstatite chondrites represent only 2% of the chondrites with 189 and 160 samples for the EH and the EL subgroups,
respectively. They have been distinguished on the basis of their Fe/Si bulk ratios as high-Fe (EH) or low-Fe (EL) (Sears et al., 1982; Kallemy and Wasson, 1986). The two EC subgroups also differ in their sulfide mineralogy. In addition to troilite (FeS), daubréelite (FeCr$_2$S$_4$) and oldhamite, which are common to both EH and EL subgroups, niningierite (MgS) is the diagnostic sulfide of EH, whereas alabandite (MnS) is specific to EL. Most of enstatite chondrites are found objects that may have been affected by weathering during their long residence time at the Earth surface. Fall objects are extremely rare with only 7 samples for EH (2 EH3: Parsa and Qingzhen; 3 EH4: Abee, Adhi Kot, Indarch; 2 EH5: St. Mark’s and Saint-Sauveur; Itqiy and Galim (b) are classified as EH but present highly fractionated trace element patterns), whereas 8 fall samples for EL are all from the EL6 group, except small fragments from the newly discovered Almahata Sitta meteorite. This meteorite was classified as a polymict ureilite and contains EC fragments of a few grams amongst a variety of lithologies. Fragment MS-177 was originally assigned an equilibration grade as EL3/4 with claimed impact-melt metal pockets (Bischoff et al., 2010). In contrast, El Goresy et al. (2017) argued for an origin by condensation based on the study of textures and sulfide mineral chemistries. REE patterns reported for EL6 samples are fractionated (Figure 1). Rubin et al. (2009) claims this depletion observed in many EL6 and some more primitive EL as a result of impact processes. EL6 samples would then represent a residuum largely depleted in LREE and Ca. No evidence for claimed impact melting was reported by El Goresy et al. (2017) in any EH or EL investigated. The good agreement observed between $\mu^{142}$Nd values corrected using Methods A and B for EL6 samples supports a fractionation event early in the history of the parent body. Ar-Ar isochron ages suggest closure of the K-Ar system at 4.45-4.49 Ga for EL5 and EL6 chondrites (Hopp et al., 2014).
When the \(^{142}\text{Nd}/^{144}\text{Nd}\) ratios are classified by metamorphic types (Figure 7), there is no difference for samples from the EH group. In contrast, two different populations are distinguished for the EL samples. EL6 samples have a larger deficit in \(^{142}\text{Nd}\) with an average of \(-12.7 \pm 6.0\) (2SD) ppm when corrected with method B. EL3 samples have a \(\mu^{142}\text{Nd}\) value similar to the terrestrial standard within the error bars: \(-0.8 \pm 7.0\) (2SD) ppm. The p-value from two sample t-test yields 0.001, meaning that the EL3 and EL6 populations likely do not share the same \(^{142}\text{Nd}\) means when corrected with method B. Samples form EL3 and EL6 subgroups are also statistically different when considering the two-sided Student’s t-values 95% confidence intervals: \(\mu^{142}\text{Nd \, EL3} = -0.8 \pm 4.4\) ppm and \(\mu^{142}\text{Nd \, EL6} = -12.7 \pm 2.3\) ppm (see Table S1). This difference suggests either that EL3 and EL6 samples come from different parent bodies or that the correction for the \(^{142}\text{Nd}\) radiogenic production is not correct for EL6 samples. The correction assumes an evolution over 4.568 Ga while Ar-Ar isochron on EL6 samples (Hopp et al., 2014) suggests a fractionation event 70 Ma after the beginning of the solar system history. Considering a two-stage evolution (chondritic \(^{147}\text{Sm}/^{144}\text{Nd}\) ratio of 0.196 from \(T_0\) to \(T_1\) with \(T_0-T_1 = 70\) Ma, the EL6 average \(^{147}\text{Sm}/^{144}\text{Nd}\) ratio of 0.2005 from \(T_1\) to present time shift the correction by \(-2\) ppm only. This average value \(\mu^{142}\text{Nd}\) for EL6 remains statistically different from the EL3 subgroup. Another compelling result for the difference amongst EC compositions comes from the measurement of Ru isotopes (Fischer-Gödde and Kleine, 2017). Although the average of all EH and EL chondrites has a small deficit in \(\varepsilon^{100}\text{Ru} (-0.08 \pm 0.04, 95\% \text{ confidence interval of the mean})\), the sample suite includes only one EL3 PCA 91020. This EL3 gave \(\varepsilon^{100}\text{Ru} = +0.02\) which is outside the 95% confidence interval of the average of the combined EL and EH data and similar to the Earth's
composition. A difference between Earth and EC has been reported for Mo isotopes but no EL3 samples have been analyzed in this dataset meaning that the reported average Mo isotopic composition of EC is based on EL6 samples only (Burkhardt et al., 2011; Dauphas et al., 2002; Dauphas 2017). The Zn isotopic compositions of the two EL3 chondrites measured by Moynier et al. (2011) are also identical to the terrestrial values, whereas the analyzed EL6 chondrites are all enriched in heavy isotopes and highly depleted in Zn. The origin of the volatile element depletion between EL3 and EL6 chondrites can be explained by volatilization, probably during thermal metamorphism. All these measurements suggest that EL samples do not come from a single parent body. Similar conclusions were reached from petrographic and geochemical observations for EH3 samples. Both normal and reverse MgS–FeS zoning profiles in niningerite grains adjacent to troilite reveal an origin on different parental asteroids with different MnS-contents in the niningerites (Lin and El Goresy, 2002; Gannoun et al., 2011b). At least 8 different parent bodies have been recently proposed for the entire EC group based on chemical variations measured in sulfide and metal phases (Weyrauch et al., 2017). However all these samples share very close isotope signatures suggesting they condensed within the same region of the solar nebula. Although our conclusion supports a genetic relationship between EL3 samples and the Earth, this statement is based on the analysis of cosmochemical materials present in our meteorite collection that represent a very small record of materials present in the Solar System. Recent high-precision Mo and Ru isotope measurements have highlighted a terrestrial signature in IAB iron meteorites indicating that their parent bodies could be the building blocks of the Earth (Worsham et al., 2017). Unfortunately the lithophile behavior of REE makes it impossible to study these meteorites for $^{146}$Sm-$^{142}$Nd systematics.
Among all groups of chondrites, EC have measured $^{142}\text{Nd}/^{144}\text{Nd}$ ratios the closest to the modern terrestrial signature ($\mu^{142}\text{Nd} = -11 \pm 12 \text{(2SD ppm)}$). In order to compare initial $^{142}\text{Nd}/^{144}\text{Nd}$ ratios, data must be corrected from the $^{142}\text{Nd}$ radiogenic production since the formation of the Solar System. Two methods of corrections are compared ($\text{measured}^{147}\text{Sm}/^{144}\text{Nd}$ vs. $\text{integrated}^{147}\text{Sm}/^{144}\text{Nd}$ ratios). The two methods yield results that are identical within the errors bars. Enstatite chondrites show a larger scatter of Sm/Nd ratios in comparison to other chondrite groups that can be attributed to early fractionation processes (e.g. samples from the EL6 subgroup) and/or recent changes caused by weathering. In these chondrites, REE are mainly concentrated in the calcium-sulfide phase (oldhamite), which is very sensitive to weathering. For this latter reason we conclude that corrections based on integrated $^{147}\text{Sm}/^{144}\text{Nd}$ ratios are more accurate for enstatite chondrites. The average of these corrected values $\mu^{142}\text{Nd}$ give $-10 \pm 12 \text{(2SD ppm)}$. We report no significant variation in the 3 stable Nd isotope ratios ($^{145}\text{Nd}/^{144}\text{Nd}$, $^{148}\text{Nd}/^{144}\text{Nd}$ and $^{150}\text{Nd}/^{144}\text{Nd}$) in EC relative to terrestrial Nd standard.

When the $^{142}\text{Nd}/^{144}\text{Nd}$ ratios are ordered by enstatite chondrite sub-groups, there is no difference for samples from the EH sub-group. In contrast, two different populations are distinguished for the EL sub-group. EL6 samples have a larger deficit in $^{142}\text{Nd}$ with an average of $-12.7 \pm 6.0 \text{(2SD ppm)}$. EL3 samples have a $\mu^{142}\text{Nd}$ value similar to the terrestrial standard within the error bars: $-0.8 \pm 7.0 \text{(2SD ppm)}$. These
two populations are statistically different (p-values and 95% confidence interval uncertainties). These results are in agreement with those obtained for other elements (e.g. Zn and Ru) and suggest that 1) EL3 and EL6 samples do not come from the same parent body, and 2) EL3 samples are isotopically similar to the Earth. Because these enstatite chondrites are relatively small in size and number, they are usually not available for destructive isotopic measurements. Average values based on both the geochemical and isotopic compositions of EL6 samples should not be considered as representative of the whole EL group. The enstatite chondrite model (Javoy, 1995; Javoy et al., 2010, Kaminski and Javoy, 2013) considers instead that the Earth is built from essentially pure EH material for explaining the Mg/Si ratio of the Earth’s upper mantle. A better knowledge on the mineralogy of the lower mantle and the proportion of silicon in the Earth’s core could help in resolving this discrepancy.

Acknowledgments

Many thanks to Ahmed El Goresy for giving us this precious EL3 sample MS-177. We thank Christa Göpel and Maria Schönbachler for they invitation to present this study in the 2017 Goldschmidt session entitled “Presolar Grains and Isotopic Heterogeneity in Planetary Materials” and motivated to write this overview on this subject 12 years after the first high-precision $^{142}$Nd measurements on chondrites were published. This project has received funding from the European Union’s Horizon 2020 research and innovation program under Grant Agreement N° 682778). AB thanks support from the NSERC Discovery Grant and Canada Research Chair programs, and the France-Canada Research Fund. This is Laboratory of Excellence ClerVolc contribution number 286.
References


Li, C. F., Li, X. H., Li, Q. L., Guo, J. H., Li, X. H., Feng, L. J., & Chu, Z. Y. (2012). Simultaneous determination of $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{147}\text{Sm}/^{144}\text{Nd}$ ratios and Sm–Nd contents from the same filament loaded with purified Sm–Nd aliquot from geological samples by isotope dilution thermal ionization mass spectrometry. *Analytical chemistry, 84*(14), 6040-6047.


List of Figures

Figure 1. Rare earth element (REE) patterns measured in enstatite chondrites from both EH and EL subgroups normalized to CI chondrites (Anders and Grevesse, 1989). New data are represented in black and Grey fields/line are data from Barrat et al. (2014) A) EH3 and impact melts (IM). LAP 02225 is an impact melt (Antarctic meteorite newsletter, Izawa et al., 2010). Galim is an unusual LL/EH polymict breccia with two lithologies: Galim (a) is a heavily-shocked LL6 chondrite and Galim (b) an impact melt breccia. Kota Kota (EH3) displays a strong enrichment in LREE. B) EH4 and EH5. Abee (EH4) is enriched in REE compared to other samples from the EH4 and EH5 groups. C) EL3 and one meteorite from the EL5 subgroup (Adrar Bous). MAC 88180 (two dissolutions) has two patterns that are both strongly enriched. Almahata Sitta MS-177 has the lowest REE contents. D) EL6 meteorites. All EL6 samples present very similar REE patterns, except EET 92063 described as highly weathered and classified in an anomalous EL6 by Barrat et al. (2014).

Figure 2. (La/Sm)$_N$ normalized to CI chondrites using the values of Anders and Grevesse, (1989) compared with Yb contents (in ppm) for EC samples. Data are from this study and literature data from Gannoun et al. (2011b), Barrat et al. (2014) and Kalleymen and Wasson (1986). IM: impact melt.
Figure. 3. Compilation of $\mu^{142}\text{Nd}$ values (in ppm) in carbonaceous, ordinary and enstatite chondrites. For each individual data point, errors bars show internal errors (i.e. in-run errors, 2SE) or 2SD when several replicates of the same sample were measured. Vertical pale yellow bands represent the mean of each chondrite groups ±2SD and vertical green bands represent 95% confidence intervals. A) Measured $\mu^{142}\text{Nd}$ expressed in part per million (ppm) deviations relative to the measured standard values. B) $\mu^{142}\text{Nd}$ normalized to the CHUR $^{147}\text{Sm}/^{144}\text{Nd}$ following the correction method A. C) $\mu^{142}\text{Nd}$ normalized to the CHUR $^{147}\text{Sm}/^{144}\text{Nd}$ value following the correction method B. See Figure 5 and text for further details. Literature data are from: Boyet and Carlson (2005); Andreasen and Sharma (2006); Carlson et al. (2007); Gannoun et al. (2011a); Bouvier and Boyet (2016); Burkhardt et al. (2016); Fukai and Yokohama (2017); and this study for Almahata Sitta MS-177. All values are shown but six samples were removed from calculated weighted averages reported in the figure caption for their respective groups because they have either highly fractionated Sm/Nd ratios (Tagish Lake, NWA 2090, Adrian, Kota Kota and Blithfiled) or correspond to a single run with a large internal uncertainty meaning that the measurement is not reliable (Abee). Weighted mean values for CC are: -33 (± 16, ± 4 for 2SD and 95% confidence interval, respectively); -32 (± 13; ± 3), -32 (± 13; ± 3) in graphs A, B and C. Weighted mean values for OC are: -16 (± 9, ± 2); -15 (± 9; ± 2), -16 (± 9; ± 2) in graphs A, B and C. Weighted mean values for EC are: -11 (± 12, ± 2); -9 (± 15; ± 3), -10 (± 12; ± 2) in graphs A, B and C. Number of measurements is 20 (10 samples) for CC, 28 (24 samples) for OC and 29 (17 samples) for EC. Individual values and calculations (averages, errors) are reported in Table S1.
Figure 4. A. Compilation of measured $\mu^{142}\text{Nd}$ vs. $^{147}\text{Sm}/^{144}\text{Nd}$ for the different chondrite samples. The line corresponds to a slope for a 4.568 Ga isochron. Here the line is forced through the mean value of the ordinary chondrites group. B. Histogram showing the distribution of measured $^{147}\text{Sm}/^{144}\text{Nd}$ ratios in the different groups of chondrites. C. Comparison of the $\mu^{142}\text{Nd}$ normalized to the chondritic evolution following the two calculation methods (A vs B) explained in the main text and shown schematically in Figure 5. D. Same diagram as C for EC samples only.

Figure 5. Schematic illustration of the correction method for the normalization of $^{142}\text{Nd}/^{144}\text{Nd}$ ratios to a common chondritic evolution. A. Method A using the measured $^{147}\text{Sm}/^{144}\text{Nd}$ ratio of the sample. B. Method B using the calculated Sm/Nd ratio from the long-lived $^{147}\text{Sm}-^{143}\text{Nd}$ systematics. $T_0$ is the age of the Solar System formation, i.e. 4.568 Ga (Bouvier and Wadhwa, 2010). More details can be found in the text. The CHUR parameters are those defined by Bouvier et al. (2008).

Figure 6. A. Mass independent stable Nd isotope composition of enstatite chondrites $\mu^{142}\text{Nd}$ corr. B vs $\mu^{148}\text{Nd}$; B. $\mu^{145}\text{Nd}$ vs $\mu^{148}\text{Nd}$; C. $\mu^{145}\text{Nd}$ vs $\mu^{150}\text{Nd}$. Average values $\pm$ 2SD (all in ppm) for CC are -32 $\pm$ 13 ($\mu^{142}\text{Nd}$), 4 $\pm$ 19 ($\mu^{145}\text{Nd}$), 16 $\pm$ 27 ($\mu^{148}\text{Nd}$) and 17 $\pm$ 68 ($\mu^{150}\text{Nd}$). For OC: -15 $\pm$ 9 ($\mu^{142}\text{Nd}$), 3 $\pm$ 15 ($\mu^{145}\text{Nd}$), 7 $\pm$ 15 ($\mu^{148}\text{Nd}$) and 14 $\pm$ 54 ($\mu^{150}\text{Nd}$). CC data include 20 measurements on 10 samples. For OC, a total of 29 measurements carried out on 25 different samples are considered. For comparison, the average values obtained from OC data published in Burkhardt et al. (2016) are -18$\pm$6 ($\mu^{142}\text{Nd}$), 6$\pm$9 ($\mu^{145}\text{Nd}$), 7$\pm$8 ($\mu^{148}\text{Nd}$) and 17$\pm$13 ($\mu^{150}\text{Nd}$) (all in ppm). Dashed lines (long and short respectively) represent mixing of the s-model prediction with the
terrestrial composition following: 1) stellar model by Arlandini et al. (1999), and 2) using the composition of SiC as representative of a pure s-process component (Hopp and Ott, 1997). External reproducibilities are not provided because this compilation includes data obtained by different authors and using different techniques.

Figure 7. Corrected µ$^{142}$Nd (normalized to the chondritic evolution using method B) in enstatite chondrites. Samples are ordered by groups. Fall meteorite samples are shown in bold.

List of Tables

Table 1. Rare Earth element contents of enstatite chondrites measured by ICP-MS. Two methods of acid digestion were used: (A) in PFA Savillex® beakers on hot plate at atmospheric pressure, (b) into individual pressurized Parr® bombs.

Table 2. Measured Nd isotope composition of the JNd1-1 standard and MS-177 (EL3). Errors correspond to the external reproducibility for the JNd1-1 standard runs (2SD) and internal error for MS-177. $^{147}$Sm/$^{144}$Nd ratio has been measured by isotope dilution. The initial $\varepsilon^{143}$Nd is calculated at 4.568 Ga using the CHUR parameters defined by Bouvier et al. (2008). The average values measured for the terrestrial standard (n=7) are $1.141850 \pm 6$ for $^{142}$Nd/$^{144}$Nd, $0.512111 \pm 4$ for $^{143}$Nd/$^{144}$Nd, $0.348403 \pm 2$ for $^{145}$Nd/$^{144}$Nd, $0.241578 \pm 2$ $^{148}$Nd/$^{144}$Nd, and $0.236446 \pm 3$ for $^{150}$Nd/$^{144}$Nd (see details in Table S2).
**Table 1.**
Rare Earth Elements measured in enstatite chondrites by ICP-MS

* Results already published in Gannoun et al. (2011b)
* Dissolution A) in PFA Savillex® beakers on hot plate at atmospheric pressure, B) into individual pressurized Parr® bombs.
* mass of the aliquot uptaken for measuring REE that corresponds to 5% of the total dissolution

<table>
<thead>
<tr>
<th>Sample Name</th>
<th>Fall Type</th>
<th>Fall Weight (g)</th>
<th>Dissolution Type</th>
<th>REE Concentration (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>A</td>
<td>A</td>
<td>La 0.247</td>
</tr>
<tr>
<td></td>
<td></td>
<td>B</td>
<td>A</td>
<td>Ce 0.556</td>
</tr>
<tr>
<td></td>
<td></td>
<td>A</td>
<td>A</td>
<td>Pr 0.887</td>
</tr>
<tr>
<td></td>
<td></td>
<td>B</td>
<td>A</td>
<td>Nd 0.423</td>
</tr>
<tr>
<td></td>
<td></td>
<td>A</td>
<td>A</td>
<td>Sm 0.144</td>
</tr>
<tr>
<td></td>
<td></td>
<td>B</td>
<td>A</td>
<td>Eu 0.053</td>
</tr>
<tr>
<td></td>
<td></td>
<td>A</td>
<td>A</td>
<td>Gd 0.205</td>
</tr>
<tr>
<td></td>
<td></td>
<td>B</td>
<td>A</td>
<td>Tb 0.035</td>
</tr>
<tr>
<td></td>
<td></td>
<td>A</td>
<td>A</td>
<td>Dy 0.244</td>
</tr>
<tr>
<td></td>
<td></td>
<td>B</td>
<td>A</td>
<td>Ho 0.054</td>
</tr>
<tr>
<td></td>
<td></td>
<td>A</td>
<td>A</td>
<td>Er 0.161</td>
</tr>
<tr>
<td></td>
<td></td>
<td>B</td>
<td>A</td>
<td>Tm 0.023</td>
</tr>
<tr>
<td></td>
<td></td>
<td>A</td>
<td>A</td>
<td>Yb 0.018</td>
</tr>
<tr>
<td></td>
<td></td>
<td>B</td>
<td>A</td>
<td>Lu 0.024</td>
</tr>
<tr>
<td></td>
<td>JNdi-1</td>
<td>MS-177</td>
<td></td>
<td></td>
</tr>
<tr>
<td>------------------</td>
<td>--------------</td>
<td>--------------</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{145}\text{Nd (V)}$</td>
<td>0.7 - 1.5</td>
<td>0.64</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{142}\text{Nd}$</td>
<td>0.0 ± 5.6</td>
<td>-10.9 ± 5.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{143}\text{Nd}$</td>
<td>0.0 ± 7.4</td>
<td>640 ± 4.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{145}\text{Nd}$</td>
<td>0.0 ± 4.4</td>
<td>5.7 ± 4.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{148}\text{Nd}$</td>
<td>0.0 ± 8.2</td>
<td>9.9 ± 7.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{150}\text{Nd}$</td>
<td>0.0 ± 10.9</td>
<td>11.5 ± 11.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{142}\text{Ce ppm}$</td>
<td>0.4</td>
<td>1.2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{144}\text{Sm ppm}$</td>
<td>0.4</td>
<td>0.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>n</td>
<td>6</td>
<td>1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{147}\text{Sm} / ^{144}\text{Nd}$</td>
<td>0.1896</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{142}\text{Nd A}$</td>
<td>-1.4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{142}\text{Nd B}$</td>
<td>-1.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{143}\text{Nd, 4.568 Ga}$</td>
<td>0.06</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Figure 1
Figure 2.
Figure 3.
Figure 4
<table>
<thead>
<tr>
<th>METHOD A</th>
<th>METHOD B</th>
</tr>
</thead>
<tbody>
<tr>
<td><img src="image1.png" alt="Graph A" /></td>
<td><img src="image2.png" alt="Graph B" /></td>
</tr>
</tbody>
</table>

**Figure 5**
Figure 6.
Figure 7
<table>
<thead>
<tr>
<th></th>
<th>Type</th>
<th>$^{26}$Al</th>
<th>$^{27}$Al</th>
<th>$^{28}$Si</th>
<th>$^{29}$Si</th>
<th>$^{30}$Si</th>
<th>$^{31}$Si</th>
<th>$^{32}$Si</th>
<th>$^{33}$Si</th>
<th>$^{34}$Si</th>
</tr>
</thead>
<tbody>
<tr>
<td>EH3</td>
<td>Found</td>
<td>-16.4</td>
<td>-10.9</td>
<td>-13.2</td>
<td>-14.6</td>
<td>-24.4</td>
<td>-16.1</td>
<td>-33.0</td>
<td>-33.9</td>
<td>-28.0</td>
</tr>
<tr>
<td>L5</td>
<td>Found</td>
<td>11.8</td>
<td>1034.1</td>
<td>1020.9</td>
<td>1013.0</td>
<td>960.5</td>
<td>872.6</td>
<td>969.7</td>
<td>982.6</td>
<td>11.1</td>
</tr>
<tr>
<td>CO3</td>
<td>Fall</td>
<td>1.7</td>
<td>4.3</td>
<td>22</td>
<td>148</td>
<td>2.2</td>
<td>4.1</td>
<td>0.0</td>
<td>2.0</td>
<td>3.9</td>
</tr>
<tr>
<td>CV3</td>
<td>Fall</td>
<td>-7</td>
<td>2.1</td>
<td>6.8</td>
<td>29</td>
<td>28.6</td>
<td>23.0</td>
<td>21.1</td>
<td>148</td>
<td>2.2</td>
</tr>
<tr>
<td>EL6</td>
<td>Fall</td>
<td>-11.2</td>
<td>-7</td>
<td>-14.2</td>
<td>-13.7</td>
<td>-26.0</td>
<td>-19.2</td>
<td>-38.5</td>
<td>-44.4</td>
<td>-42.4</td>
</tr>
<tr>
<td>H4</td>
<td>Fall</td>
<td>11.5</td>
<td>14.0</td>
<td>13.2</td>
<td>10.3</td>
<td>28.0</td>
<td>20.0</td>
<td>13.2</td>
<td>20.0</td>
<td>11.5</td>
</tr>
<tr>
<td>H5</td>
<td>Fall</td>
<td>22.1</td>
<td>13.2</td>
<td>10.3</td>
<td>28.0</td>
<td>20.0</td>
<td>13.2</td>
<td>20.0</td>
<td>11.5</td>
<td>22.1</td>
</tr>
<tr>
<td>L6</td>
<td>Fall</td>
<td>11.1</td>
<td>10.2</td>
<td>10.4</td>
<td>10.6</td>
<td>10.7</td>
<td>10.8</td>
<td>10.9</td>
<td>10.8</td>
<td>11.1</td>
</tr>
<tr>
<td>H4</td>
<td>Fall</td>
<td>-4.2</td>
<td>-6.1</td>
<td>-6.7</td>
<td>-4.9</td>
<td>3.8</td>
<td>5.0</td>
<td>4.1</td>
<td>3.9</td>
<td>3.1</td>
</tr>
<tr>
<td>H5</td>
<td>Fall</td>
<td>-4.2</td>
<td>-6.1</td>
<td>-6.7</td>
<td>-4.9</td>
<td>3.8</td>
<td>5.0</td>
<td>4.1</td>
<td>3.9</td>
<td>3.1</td>
</tr>
<tr>
<td>L6</td>
<td>Fall</td>
<td>-4.2</td>
<td>-6.1</td>
<td>-6.7</td>
<td>-4.9</td>
<td>3.8</td>
<td>5.0</td>
<td>4.1</td>
<td>3.9</td>
<td>3.1</td>
</tr>
<tr>
<td>H4</td>
<td>Fall</td>
<td>-4.2</td>
<td>-6.1</td>
<td>-6.7</td>
<td>-4.9</td>
<td>3.8</td>
<td>5.0</td>
<td>4.1</td>
<td>3.9</td>
<td>3.1</td>
</tr>
<tr>
<td>H5</td>
<td>Fall</td>
<td>-4.2</td>
<td>-6.1</td>
<td>-6.7</td>
<td>-4.9</td>
<td>3.8</td>
<td>5.0</td>
<td>4.1</td>
<td>3.9</td>
<td>3.1</td>
</tr>
<tr>
<td>L6</td>
<td>Fall</td>
<td>-4.2</td>
<td>-6.1</td>
<td>-6.7</td>
<td>-4.9</td>
<td>3.8</td>
<td>5.0</td>
<td>4.1</td>
<td>3.9</td>
<td>3.1</td>
</tr>
</tbody>
</table>

Reference

- Burkhardt et al., 2016
- Gannoun et al., PNAS 2011
- Fukai and Yokoyama, 2017
| Nd (ng) | I4S (V) | $^{143}$Nd/$^{144}$Nd | 2SE | $^{143}$Nd/$^{144}$Nd | 2SE | $^{143}$Nd/$^{144}$Nd | 2SE | $^{143}$Nd/$^{144}$Nd | 2SE | $^{143}$Nd/$^{144}$Nd | 2SE | $^{143}$Sm/$^{144}$Nd | 2SE | $^{143}$Ce/$^{142}$Nd | 2SE | $^{145}$Nd/$^{144}$Nd | 2SE | $^{145}$Nd/$^{144}$Nd | 2SE | $^{146}$Nd/$^{144}$Nd | 2SE | $^{147}$Nd/$^{144}$Nd | 2SE | $^{148}$Nd/$^{144}$Nd | 2SE | $^{150}$Nd/$^{144}$Nd | 2SE | $^{144}$Nd | 2SE | N |
|-------|--------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|----------------|------|
| JNd1-1_F1R1 | 500 | 0.97 | 1.141852 | 0.512110 | 0.348403 | 0.241576 | 0.236445 | 0.4 | JNd1-1_F1R2 | 500 | 0.97 | 1.141850 | 0.348403 | 0.241577 | 0.236445 | 0.4 | JNd1-1_F2R1 | 500 | 1.28 | 1.141847 | 0.348403 | 0.241576 | 0.236445 | 0.4 | JNd1-1_F3R1 | 500 | 1.68 | 1.141853 | 0.348403 | 0.241576 | 0.236445 | 0.4 | JNd1-1_F4R1 | 250 | 0.70 | 1.141844 | 0.348404 | 0.241576 | 0.236445 | 0.4 | JNd1-1_F5R1 | 250 | 1.48 | 1.141851 | 0.348404 | 0.241576 | 0.236445 | 0.4 | JNd1-1_F6R1 | 250 | 0.97 | 1.141852 | 0.348404 | 0.241576 | 0.236445 | 0.4 |
| Mean JNd1 | 1.141850 | 0.512111 | 0.348403 | 0.241577 | 0.236446 | 0.4 | 2s | 0.000006 | 0.000004 | 0.000002 | 0.000002 | 0.000003 | 0.4 | 2s (ppm) | 5.6 | 7.4 | 4.4 | 8.2 | 10.9 | n | 7 | 7 | 7 | 7 | 7 |
| MS177_F9R1 | 300 | 0.64 | 1.141838 | 0.512439 | 0.348405 | 0.241581 | 0.236448 | 0.4 | 1.2 | 640 | 0.3 |