Barium Isotope Abundances in Meteorites and Their Implications for Early Solar System Evolution

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ABSTRACT

Several nucleosynthetic processes contributed material to the Solar System; however, the relative contributions of each process, the timing of their input into the solar nebula, and how well these components were homogenized in the solar nebula remain only partially constrained. The Ba isotope system is particularly useful in addressing these issues because Ba contains isotopes are synthesized through three nucleosynthetic processes (*s*-, *r*-, *p*-process). In this study, high precision Ba isotope analyses of 22 different whole rock chondrites and achondrites (carbonaceous chondrites, ordinary chondrites, enstatite chondrites, Martian meteorites, and eucrites) were performed to constrain the distribution of Ba isotopes on the regional scale in the Solar System. A melting method using aerodynamic levitation and CO₂-laser heating was used to oxidize SiC, a primary carrier of Ba among presolar grains in carbonaceous chondrites. Destruction of these grains during the fusion process enabled the complete digestion of these samples. The Ba isotope data presented here are thus the first for which complete dissolution of the bulk meteorite samples was certain. Enstatite chondrites, ordinary chondrites, and all achondrites measured here possess Ba isotope compositions that are not resolved from the terrestrial Ba isotope composition. Barium isotope anomalies are evident in most of the carbonaceous chondrites analyzed, but the ¹³⁵Ba anomalies are generally smaller than previously reported for similarly sized splits of CM2 meteorites. Variation in the size of the ¹³⁵Ba anomaly is also apparent in fused samples from the same parent body (e.g., CM2 meteorites) and in different pieces from the same meteorite (e.g., Orgueil, CI). Here, we investigate the potential causes of variability in ¹³⁵Ba, including the contribution of radiogenic ¹³⁵Ba from the decay of ¹³⁵Cs and incomplete homogenization of the presolar components on the <0.8 g sample scale.

Key words

Barium isotopes, nucleosynthetic isotope anomaly, ¹³⁵Cs-¹³⁵Ba, Solar System evolution.

1. INTRODUCTION

A number of processes can cause isotope ratios in Solar System materials to differ from bulk terrestrial isotope ratios. These processes include (i) radioactive decay (e.g., Reynolds, 1960, 1967; Rowe and Kuroda, 1965), (ii) nuclear reactions following interaction with cosmic rays (e.g., Arnold, 1961), (iii) mass-dependent and -independent isotope fractionation during chemical reactions (e.g., Keesom and van Dijk, 1931; Urey et al., 1932a,b; Thiemens and Heidenreich, 1983; Fujii et al., 2009), and (iv) incomplete mixing in the solar nebula of nucleosynthetic material having exotic isotope abundances from different stellar sources, often referred to as nucleosynthetic isotope anomalies (e.g., presolar grains; Zinner, 1998; Hidaka et al., 2003; Andreasen and Sharma, 2007; Carlson et al., 2007). The non-solar isotope compositions of individual presolar grains (e.g., nucleosynthetic material) result from nucleosynthetic reactions that were active in their stellar source regions (Burbidge et al., 1957). Ion microprobe (SHRIMP-RG) analyses of the Ba isotopic composition of a SiC-enriched bulk sample (KJB) from Murchison (Ávila et al., 2013) show large anomalies in $\delta^{134}\text{Ba}/^{136}\text{Ba}$ (140 ± 13), $\delta^{135}\text{Ba}/^{136}\text{Ba}$ (-423 ± 2), $\delta^{137}\text{Ba}/^{136}\text{Ba}$ (-284 ± 5), and $\delta^{138}\text{Ba}/^{136}\text{Ba}$ (-281 ± 23), where $\delta^i\text{Ba}/^{136}\text{Ba} = [({}^i\text{Ba}/^{136}\text{Ba})_{\text{measured}}/({}^i\text{Ba}/^{136}\text{Ba})_{\text{solar}} - 1] \times 10^3$ (Fig. 1). This isotope signature is generally considered to reflect the *s*-process in Asymptotic Giant Branch (AGB) stars (Savina et al., 2003; Marhas et al., 2007; Ávila et al., 2013). Complementary coupled excesses in ^{135}Ba and ^{137}Ba and have been identified in carbonaceous chondrites and acid leachates (Hidaka et al., 2001; Hidaka et al., 2003; Andreasen and Sharma, 2007; Carlson et al., 2007; Hidaka and Yoneda, 2011; Qin et al., 2011;). The cause of the variations in ^{135}Ba and ^{137}Ba isotope abundances in whole rock meteorites is most frequently attributed to the heterogeneous distribution of presolar grains. This type of isotope anomaly is here termed “nucleosynthetic isotope anomaly” and is the focus of this contribution, which investigates the degree and cause of Ba isotopic heterogeneity in the solar nebula.

Determining the scale of isotope heterogeneity in the solar nebula is an important aspect of cosmochemistry. These data can be used to determine the number and type of supernova contributors to the Solar System, to test the assumption of isotopic homogeneity of the parent isotope that underpins the use of chronometers based on short- and long-lived radioisotopes, and potentially to trace chemical variability in the nebula that might have existed at during

planetesimal and planet formation. Published ages may be inaccurate if the initial parent/reference isotope ratio (e.g., $^{135}\text{Cs}/^{133}\text{Cs}$, $^{26}\text{Al}/^{27}\text{Al}$) had varied throughout the Solar System (e.g., Makide et al., 2011). Such variations in initial abundances could manifest themselves as nucleosynthetic isotope anomalies in bulk meteorite samples. If significant bulk sample nucleosynthetic anomalies were indisputably identified, then the current Solar System chronometers that utilize these isotopes would have to be replaced by regional chronometers that are based on the local abundances of the parent nuclides.

How well presolar grains were homogenized throughout the Solar System on the asteroidal- and planetary scales remains unclear. The primary reason for the uncertainty on this point is the difficulty in obtaining *whole rock* chemical analyses of meteorite samples (e.g., carbonaceous chondrites) that contain varying proportions of acid-insoluble components. Regarding the Ba isotope system, the concern is that the documented coupled excesses in ^{135}Ba and ^{137}Ba in carbonaceous chondrites are the result of incomplete dissolution of presolar SiC grains in the bulk sample (e.g., Andreasen and Sharma, 2007; Carlson et al., 2007). Presolar SiC grains display large deficits in ^{135}Ba and ^{137}Ba isotopes and failure to digest part of this component can result in a complementary enrichment pattern in ^{135}Ba and ^{137}Ba , similar in magnitude to the anomalies that are observed in whole rock carbonaceous chondrites. The fact that differentiated and high metamorphic grade meteorites, which no longer contain intact presolar grains, do not display Ba isotope anomalies lends support to this concern.

To evaluate the distribution of Ba isotopes in the Solar System, 22 bulk meteorites from different meteorite classes were analyzed in this study while taking special care to ensure complete dissolution of insoluble phases by using a high temperature laser fusion method. Barium is a moderately refractory element ($T_c = 1455\text{ K}$, BaTiO_3 ; Lodders, 2003; T_c = temperature at which 50% of the element has condensed from the solar nebula) and is a trace element in chondrites (CI = 2.34 ppm; Anders and Grevesse, 1989) with carrier phases including melilite, feldspar, and SiC (estimates of the Ba concentration in individual SiC grains range from ~ 2-10 ppm; Ávila et al., 2013). Barium has seven stable isotopes that sample three different nucleosynthetic processes (*s*-, *r*-, and *p*-processes, Table 1). Low thermal neutron capture cross sections of all Ba isotopes require long irradiation times by cosmic rays to create significant changes in Ba isotope abundances (Table 1; Mughabghab 2003). There is no reported evidence that indicates secondary

nuclear processes produced shifts in Ba isotope abundances in cosmochemical samples. Modification of Ba isotopic composition could also arise from the decay of radioactive species. For example, positive anomalies in ^{135}Ba could result from the decay of the short-lived radioisotope ^{135}Cs ($t_{1/2} = 2.3 \text{ Ma}$; Hidaka et al., 2001; Hidaka and Yoneda, 2011; Hidaka and Yoneda, 2013; Bermingham et al., 2014). Lanthanum-138 decays to ^{138}Ba with a half-life of $\sim 105 \text{ Ga}$ (Nakai et al., 1986). Because of the very low natural abundance of ^{138}La ($< 0.09 \%$) and its long half-life, radiogenic ^{138}Ba is only a very minor component in bulk meteorites (Ingham et al., 1947; Anders and Grevesse, 1989).

2. ANALYTICAL TECHNIQUES

2.1 Meteorite samples

A total of 25 samples (of 22 meteorites) from different classes (carbonaceous chondrites, low and high metamorphic grade H chondrites, eucrites, and Martian meteorites) were selected for analysis (Table 2). This collection of samples was carefully chosen to determine the Ba isotope composition of material that is as representative of the asteroid belt, Mars, and Earth as possible. To confirm that the methods used for the isolation of Ba and measurement of its isotopic composition did not impart systematic biases, the Ba isotope composition of terrestrial basalt (DU17) and two eucrites (Juvinas and Stannern) were measured. Eucrites are not considered to possess nucleosynthetic anomalies (Andreasen and Sharma, 2007) and, therefore, provide an additional robust test for isotope fractionation effects during sample dissolution, ion-exchange chromatographic separation of Ba, and measurement by thermal ionization mass spectrometry (TIMS).

Nearly all (19) of samples selected for this study are observed falls without evidence for terrestrial weathering. Meteorite finds were avoided because such samples, especially those from hot deserts, can contain large amounts of secondary barite. Barite forms by the interaction of brines with a meteorite during the humid seasons (Lee and Bland, 2004), which may make most of the Ba in such samples terrestrial in origin and unsuitable for this study. Three Martian meteorites that are desert finds (Los Angeles 001, Miller Range 03346, and North West Africa 4864) were included in the sample set. Importantly, however, barite has not been reported in

these samples (Rubin et al., 2000; Day et al., 2006). The analyzed chips of these samples were taken from the center of the respective meteorites to avoid potential terrestrial alteration products.

2.2 Sample preparation

Meteorites were cut using a water-cooled saw with a diamond-coated blade to isolate volumes free of fusion crust or rust patches. Saw marks were removed using corundum paper. After cutting, samples were immersed in water and treated with ultrasound for ~20 minutes. Samples were dried, crushed to sand-sized fragments using a steel piston and a hammer, and then ground by hand into a fine powder using an agate mortar and pestle. In this study, sample sizes between 0.05 and 0.95 g were used. Water used throughout sample preparation, digestion, and column chemistry was deionized with a resistivity of 18.2 MΩcm.

2.3 Laser fusion

Obtaining accurate whole rock chemical analyses of carbonaceous chondrites has been difficult because of the presence of acid insoluble components, in particular presolar SiC (Hidaka et al., 2003; Andreasen and Sharma, 2007; Carlson et al., 2007; Hidaka and Yoneda, 2011). A reliable laboratory method for dissolving the chemically robust moissanite (SiC) is through oxidation ($\text{SiC} + 2\text{O}_2 \rightarrow \text{SiO}_2 + \text{CO}_2$) via melting the sample in an oxidizing environment. An experimental melting method using an aerodynamic levitation combined with CO₂-laser heating at the Geoscience Center at the University of Göttingen, Germany, was used in this study to induce oxidation of SiC (for experimental details, refer to Pack et al., 2010). All carbonaceous chondrites and low metamorphic grade samples were melted by heating ~10 mg aliquots of the powdered sample (total sample mass ranged from 0.05 to 0.95 g; Table 2) under the laser for 5 to 10 s. The laser heating time and desirable sample mass were determined from earlier experiments using synthetic SiC powders (grain sizes from 3 to 17 μm). A random selection of these and sample melt beads were analyzed using a Scanning Electron Microscope (SEM) Leo 1485 (University of Göttingen) to ensure that complete melting of the aliquot had occurred. Complete melting is indicated by a quenched texture as shown in Fig. 2. Subsequently, melt beads were placed in an ultrasonic water bath for 10 minutes to remove adhering material and dissolved using concentrated acids as described below.

2.4 Sample digestion

Meteorite samples processed at the Westfälische Wilhelms-Universität (WWU) were digested in a 5:1 mixture of 29 M HF – 14 M HNO₃ in sealed, non-pressurized Savillex[®] vials at 150 °C on a hotplate for 2 - 5 days. After evaporation, a second attack with concentrated HF – HNO₃ was carried out at 150 °C for two or four days. Solutions were evaporated with 1 - 2 ml of concentrated HClO₄ (Suprapur[®], 70 %). Vials containing the sample and an additional 1 - 2 ml HClO₄ were capped and left overnight at 120 °C. Samples were dried down a second time. At this point, the majority of samples consisted of brown solids with no visible white precipitates. For samples in which white precipitates (presumably fluorides) remained, the HClO₄ treatment was repeated until the white precipitates were gone. Samples were then dissolved in 10 ml of 6 M HCl and left overnight at 120 °C in capped vials. The samples were then dried down, dissolved in ~5 ml of 14 M HNO₃, dried down, and taken up in the loading solution.

A subset of samples was processed at the Department of Terrestrial Magnetism, Carnegie Institution for Science (DTM). These samples were digested as outlined in Carlson et al., (2007), which is essentially the same digestion technique as that used at WWU (with the exception that HClO₄ is not used) and thus is not expected to produce systematic differences between the datasets.

All digested samples produced a clear solution, except for Butsura, Ösede, and Allende_{powder} (Allende_{acid} is an aliquot of Allende that did not undergo laser fusion). Butsura and Ösede had a small amount of fine-grained black residue that was identified by X-ray diffraction as Cr-spinel. The Ba abundance in Cr-spinel is negligible (e.g., Grégoire et al., 2000), thus loss of Ba due to incomplete dissolution of this phase is not expected. There was a substantial amount of very fine-grained black residue observed in Allende_{acid}, which is considered to be a mix of organic compounds and insoluble presolar material.

2.5 Chemical separation of Ba, REE, and major elements

At WWU, a chemical separation method derived from previous studies was used to separate and isolate Ba, REE, and the major elements (Eugster et al., 1969; Andreasen and Sharma, 2007; Carlson et al., 2007). The first step of this two-stage cation-exchange chemistry uses standard procedures (1-cm diameter x 20 cm long quartz glass columns filled with AG50W-X8 cation-

exchange resin, equilibrated in water) with 2 M HNO₃, 2.5 M and 6 M HCl as eluents to isolate REE and Ba and major elements, respectively. The same columns were used for the second step, but were cleaned before use and 2.5 M and 6 M HCl were used as the eluents to separate Ba from the major elements.

The digested meteorite samples were redissolved in 2 M HNO₃ and centrifuged before column chemical separation of the desired elements. Samples were loaded onto the first column in 12 ml of 2 M HNO₃. Columns were rinsed with 26 ml of 2 M HNO₃ to elute a Ba and major element cut for processing through a second column. The REE cut was eluted in 18 ml of 6 M HCl. For the second step chemistry, the Ba and major element cut was dried down, dissolved in 6 ml of 2.5 M HCl, and then loaded onto the column. The major element fraction was eluted in 20 ml of 2.5 M HCl and the Ba fraction was eluted with 24 ml of 6 M HCl. The Ba yield from the combined column chemistries was 70 - 80 %.

The method of chemical separation of Ba, REE, and major elements used at DTM is the same as that reported in Carlson et al. (2007). This isolation process is very similar to that used at WWU and is not expected to produce systematic differences between the datasets.

The total procedural Ba blank for non-fused samples processed at WWU and DTM was 0.2 - 1 ng. The amount of Ba blank stemming from the laser fusion was estimated by processing ca. 0.5 g of powdered gem-quality quartz of known Ba concentration. After correcting for the Ba contribution of the quartz, the total blank arising from the melting of a sample was 5 ng or less, which typically corresponds to 0.3 % or less of the total Ba processed. The total procedural blanks for fused samples (5.2 - 6 ng) and non-fused samples (0.2 - 1 ng) are insignificant, thus blank corrections were not applied.

2.6 Isotope measurements

Barium isotope data were collected during different measurement campaigns at WWU and DTM using a *Thermo Fisher* TIMS. Almost identical loading and measurement methods were used at DTM and WWU, so a systematic difference between the datasets resulting from the loading and measurement techniques is unlikely.

At WWU, a double filament assembly (Ta–evaporation, Re–ionization) was used because it produced more stable signals and thus more reproducible measurements of a Ba standard solution than a single Re filament. The separated Ba was dissolved in 6 M HCl and, just before loading, ~1 μ L 0.04 M H₃PO₄ was mixed into this solution.

The TIMS at WWU is equipped with nine Faraday collectors enabling $^{132,134-138}\text{Ba}$ and interference monitors ^{139}La and ^{140}Ce to be measured using the static mode. Barium-130 was not measured because the tether connecting L4 and L3 was too short to allow simultaneous measurement of ^{130}Ba and ^{132}Ba . Generally, the $^{132}\text{Ba}/^{136}\text{Ba}$ values have large analytical uncertainties (ca. 220 ppm) associated with their measurement because of the low natural abundance of the *p*-process isotope ^{132}Ba (~0.1%). As a result of the poor analytical precision, only upper limits can be given for the *p*-process contribution. The analyses were carried out in static mode rather than dynamic mode because all isotopes (except ^{130}Ba) could be measured in 2 hours (200 - 400 ng Ba load produce a ^{138}Ba intensity of ~20 V for ~2 hours; 18 blocks of 18 ratios, ~320 integrations per filament). The alternative dynamic mode that was considered would have significantly increased the measurement time, which may have led to detrimental sample fractionation effects (e.g., Roth et al., 2014) and filament reservoir mixing effects (e.g., Upadhyay et al., 2008; Andreasen and Sharma 2009). Barium isotope data were collected during four different analytical sessions, which were each separated by ~4 months. Because long-term drift in Faraday cup performance does not cancel out in static analyses as it can in multi-dynamic analyses, the samples were referenced only to the standard data collected within their respective analytical session. For this reason, the reported external precision varies among the analytical sessions (Tables 3 - 6; Figs. 3 and 4). Peak centering and lens focusing were performed every fourth block. The amplifier connections were rotated after each block and baselines were measured at the beginning of each block. Isobaric interferences from ^{138}La and $^{136,138}\text{Ce}$ were monitored using ^{139}La and ^{140}Ce but were found to be at baseline levels during all measurements, thus no interference corrections were applied. For details of TIMS data acquisition at DTM, see Carlson et al. (2007). Notably, ^{130}Ba and ^{132}Ba were both measured during the analytical campaign at DTM because the tether connecting L4 and L3 was long enough to allow simultaneous measurement of ^{130}Ba and ^{132}Ba .

The Ba from each sample digestion was measured up to 4 times, using new Ba-loads on new filaments. Sample data were compared to terrestrial basalt (DU17) data and a Ba standard solution (Specpure[®] Alfa Aesar). Multiple analyses of the Ba standard solution yielded precisions better than ± 12 ppm, ± 29 ppm, and ± 11 ppm for $^{135}\text{Ba}/^{138}\text{Ba}$, $^{136}\text{Ba}/^{138}\text{Ba}$, and $^{137}\text{Ba}/^{138}\text{Ba}$, respectively, as determined from the 2σ of the population of replicate standard measurements (see Tables 3 - 6).

All data presented here have been corrected for mass fractionation using the exponential law referenced to $^{134}\text{Ba}/^{138}\text{Ba} = 0.03371$ (De Bièvre and Taylor, 1993; Andreasen and Sharma, 2007). Previous studies commonly used $^{134}\text{Ba}/^{136}\text{Ba}$ (s-process only isotopes) for nominalization (Hidaka et al., 2003; Andreasen and Sharma, 2007; Carlson et al., 2007). In this study, ratios involving ^{136}Ba showed unexpectedly poor reproducibility, which would be propagated into all ratios if the data were fractionation corrected to $^{134}\text{Ba}/^{136}\text{Ba}$. We found an improvement in ratio precision ranging from a factor of 4 on ^{135}Ba and 2 on ^{137}Ba when fractionation correcting instead to $^{134}\text{Ba}/^{138}\text{Ba}$. The cause of the lowered precision on ^{136}Ba is not clear, but we suspect degradation of the axial faraday cup as this phenomenon was not observed in the Ba data collected on the same instrument, using essentially the same techniques, as reported by Carlson et al. (2007).

An advantage of normalizing to $^{134}\text{Ba}/^{138}\text{Ba}$ is that it covers a larger spread in masses and thus yields a more precise measure of the isotope fractionation during the analysis. As long as there is no significant contribution from ^{139}La to ^{138}Ba , this normalization scheme is appropriate for cosmochemical samples. The La/Ba was monitored and was found to be chondritic or subchondritic in all samples, thus indicating that referencing data to $^{134}\text{Ba}/^{138}\text{Ba}$ is a sound approach. All data are presented using the μ notation (deviations in parts per million from the terrestrial standard) for comparison between present and published data.

3. RESULTS

3.1 Barium isotope distribution in whole rock meteorite samples

The Ba isotope compositions of DU17, Juvinas, and Stannern were found to be analytically indistinguishable from those of the Ba standard measured during this study (Table 3, Fig. 3). The data for DU17 and Stannern, and the replication of reported Ba isotopic composition of Juvinas, indicate that the methods of digestion, chemical separation, and isotope measurement used do not impart biases. The Ba isotope compositions of Martian meteorites, eucrites, enstatite and ordinary chondrites fall within the external precision of the Ba standard solution indicating that these samples have Ba isotope compositions indistinguishable from terrestrial values (Tables 3 - 5; Fig. 4). The absence of Ba isotope anomalies in these meteorites concurs with published data of meteorites from the same groups (Hidaka et al., 2003; Andreasen and Sharma, 2007; Carlson et al., 2007). Excesses in ^{135}Ba and ^{137}Ba are analytically resolvable from the Ba standard in Orgueil, Allende, Vigarano, Cold Bokkeveld, Lancé, and Kainsaz (Tables 5 - 6 and Fig. 4). The Ba isotope data from Renazzo (CR2), Murchison (CM2), and Mighei (CM2) do not have isotope anomalies that are well resolved from the terrestrial standard.

The Ba isotope anomalies in the studied carbonaceous chondrites are generally similar to those reported by Hidaka et al. (2003), Andreasen and Sharma (2007), and Carlson et al. (2007). When comparing our fused meteorite data with published data (unfused) from the same sample, however, there is a smaller ^{135}Ba anomaly in CM2 samples that underwent fusion, despite similar levels of precision reached in the relevant studies (Table 7). For example, data from the present study for Cold Bokkeveld shows excesses in $^{135}\text{Ba}/^{138}\text{Ba}$ of 34 ± 7 ppm whereas Carlson et al., (2007) reported $^{135}\text{Ba}/^{138}\text{Ba} = 53\pm11$ ppm anomalies (the $^{134}\text{Ba}/^{138}\text{Ba}$ -normalized data were obtained from unpublished data collected as part of the Carlson et al. 2007 study). The Ba isotope data for Mighei collected in our study do not possess resolvable ^{135}Ba and ^{137}Ba anomalies. This differs from the results from Carlson et al. (2007), who reported a small $^{135}\text{Ba}/^{138}\text{Ba}$ excess in Mighei (24 ± 11 ppm).

A slight reduction in $^{135}\text{Ba}/^{138}\text{Ba}$ variability, however, is not evident when comparing our Ba isotope compositions obtained on fused pieces of Orgueil (CI) to published data on unfused samples of the same meteorite. Harper et al., (1991) published two compositions for Orgueil ($^{135}\text{Ba}/^{138}\text{Ba}$ of 20 ± 7 and 40 ± 8 ppm). Carlson et al., (2007) reported a $^{135}\text{Ba}/^{138}\text{Ba}$ value of 31 ± 11 ppm, which is between those of two different fused pieces studied here (Orgueil 1: 53 ± 12 ppm, and Orgueil 2: 22 ± 7 ppm; Fig. 4c,d; Table 7).

Closer examination of the Ba isotope compositions of fused samples in this study reveals a notable variation in the magnitude of the $^{135}\text{Ba}/^{138}\text{Ba}$ anomaly between samples from the same group. For example, the Ba isotope composition of samples from the CM2 parent body, Cold Bokkeveld ($^{135}\text{Ba}/^{138}\text{Ba} = 34 \pm 7$ ppm) and Mighei ($^{135}\text{Ba}/^{138}\text{Ba} = 14 \pm 7$ ppm), have resolved differences in ^{135}Ba isotope compositions. Variation in ^{135}Ba is also apparent in different pieces of the same meteorite that have undergone fusion (e.g., Orgueil, CI). When comparing the Ba isotope compositions obtained on 2 pieces of Orgueil (both 0.7 - 0.8 g and fused) obtained from two different museums (NHM UK and Smithsonian USA), a resolvable difference is observed in $^{135}\text{Ba}/^{138}\text{Ba}$ (Orgueil 1: 53 ± 12 ppm; Orgueil 2: 22 ± 7 ppm).

4. DISCUSSION

4.1 Ba isotope distribution in the Solar System

The coupled excesses in ^{135}Ba and ^{137}Ba identified here in carbonaceous chondrites generally concur with previous studies (Hidaka et al., 2003; Andreasen and Sharma, 2007; Carlson et al., 2007). Fused samples from the CM2 group have slightly smaller Ba isotope anomalies than the same samples processed only by acid dissolution. The magnitude of the $^{135}\text{Ba}/^{138}\text{Ba}$ anomaly varies among fused samples from the same parent body (e.g., Cold Bokkeveld and Mighei; CM2 meteorites) and also among different pieces of the same meteorite (e.g., Orgueil 1 and Orgueil 2; CI). This small but resolvable variation in ^{135}Ba is striking because it is generally assumed that meteorites originating from the same parent body possess the same nucleosynthetic isotope composition, as long as the isotope system in question is not susceptible to modification via secondary reactions, such as cosmic ray exposure effects (e.g., Wittig et al., 2013) or radioactive decay. The CRE ages of samples studied here are all ≤ 41 Ma (data from Pepin et al., 1966; Heymann and Anders, 1967; Marti and Graf, 1992; Bischoff et al., 1993; Bischoff et al., 1996; Schultz and Weber 1996; Herzog et al., 1997; Mazar et al., 1998; Miura et al., 1998; Terribilini et al., 2000; Eugster et al., 2002; Huss et al., 2008; Roth et al., 2011; Table 2) and are not old enough to produce the resolved Ba isotope variations via neutron capture reactions.

The variation in $^{135}\text{Ba}/^{138}\text{Ba}$ compositions of samples that were fused from those that were not fused, and among fused samples of the same meteorite groups/meteorite is unlikely to stem from

neutron capture effects because all Ba isotopes have low neutron capture cross sections (Table 1). The development of a positive ^{135}Ba anomaly could potentially occur via a neutron reaction pathway from neighboring nuclides, but such a pathway is currently unknown. The Ba isotope anomaly variations are also unlikely to originate from nuclear field effects because the pattern of the observed ^{135}Ba and ^{137}Ba anomalies does not match the pattern expected for mass-independent fractionation (Fujii et al., 2009). Instead, these effects could be related to: (1) nucleosynthetic or radiogenic variation in the abundance of ^{138}Ba used in the normalization ratio; (2) the digestion techniques employed in the respective studies; (3) contribution of radiogenic ^{135}Ba ; or (4) incomplete homogenization of the presolar or refractory component at the sampling scale of this and published studies.

4.2 Potential effects of nucleosynthetic or radiogenic ^{138}Ba anomalies

In the present study, all data were corrected for mass fractionation using the exponential law with $^{134}\text{Ba}/^{138}\text{Ba} = 0.03371$ as the reference ratio (De Bièvre and Taylor, 1993; Andreasen and Sharma, 2007). This ratio comprises isotopes that were produced via different nucleosynthetic processes (*s*-only and *s+r*-processes, respectively) and potentially radiogenic ^{138}Ba from the decay of the rare ^{138}La . Nucleosynthetic isotope anomalies or radiogenic contributions to these normalizing ratios can thus potentially result in propagation of ^{138}Ba variability into the other Ba isotope ratios through the fractionation correction. Isotope anomalies in ^{138}Ba have been measured in the acid residue of Murchison (Qin et al., 2011) and in pre-solar SiC grains (Pellin et al., 2000). Generally, these isotope variations have been attributed to *s*-process variability. Direct evidence for nucleosynthetic variation in ^{138}Ba on the whole rock sample scale, however, is yet to be found. The coupled excesses in ^{137}Ba and ^{138}Ba reported by Ranen and Jacobsen (2006) were initially explained by incomplete mixing of neutron burst material in the early solar nebula, but were later shown to result from an analytical artifact (Andreasen and Sharma, 2007). The absence of nucleosynthetic Ba isotope variations suggests that if there are anomalous nucleosynthetic contributions to ^{138}Ba , they are smaller than the current analytical uncertainty achievable with state-of-the-art mass spectrometers and, therefore, would not adversely affect the mass fractionation corrected data.

Evidence for the decay of ^{138}La to ^{138}Ba has recently been suggested to be the source of ^{138}Ba variations identified in three Group II CAIs (Brennecka et al., 2013) and could also potentially

affect $^{134}\text{Ba}/^{138}\text{Ba}$ -normalized data. The potential contribution from ^{138}La can be constrained by obtaining La/Ba for those samples with anomalous Ba isotope compositions and contrasting it with chondritic La/Ba ~ 0.1 (Lodders, 2003). An aliquot of the carbonaceous chondrites processed at DTM was removed before column chemistry and analyzed using an *iCAPQ* ICP-MS (ThermoFisher) housed at DTM to obtain whole rock elemental abundances. These carbonaceous chondrites have chondritic to subchondritic La/Ba (0.09 – 0.12) which constrain $^{138}\text{Ba}^*/^{138}\text{Ba}$ to $< 4.6 \times 10^{-6}$, where $^{138}\text{Ba}^*$ refers to radiogenic ^{138}Ba . In order to produce significant radiogenic $^{138}\text{Ba}^*$ ($^{138}\text{Ba}^*/^{138}\text{Ba} > 1 \times 10^{-5}$), La/Ba would have to exceed 0.26. Thus, there is no significant contribution by ^{138}La for these samples and the variation seen in the magnitude of Ba isotope anomalies from samples of the same meteorite cannot be attributed to radiogenic ^{138}Ba . Another aspect of this issue is that variability only in ^{138}Ba would be propagated into all the isotope ratios, with the magnitude of offset increasing proportional to the mass difference from ^{138}Ba . Where anomalous Ba is observed in the data reported here, the data do not show the mass-dependent anomalies that would be expected if the anomaly were present only in ^{138}Ba .

Additionally, variation in ^{135}Ba among samples from the CM2 parent body (CM2: Cold Bokkeveld $^{135}\text{Ba}/^{136}\text{Ba} = 58 \pm 7$ ppm; Mighei $^{135}\text{Ba}/^{136}\text{Ba} = 27 \pm 7$ ppm; Murray $^{135}\text{Ba}/^{136}\text{Ba} = 12 \pm 7$ ppm; CV3: Grosnaja $^{135}\text{Ba}/^{136}\text{Ba} = 40 \pm 7$ ppm and Mokoia $^{135}\text{Ba}/^{136}\text{Ba} = -12 \pm 7$ ppm) also is evident in the dataset published by Carlson et al., (2007) in which normalization was by the *s*-process only ($^{134}\text{Ba}/^{136}\text{Ba}$) ratio. This suggests that the source of the ^{135}Ba variation is not related to the choice of normalizing ratio.

4.3 Digestion techniques

The pre-digestion laser treatment employed in the present study effectively oxidized SiC, a primary carrier of large Ba isotope anomalies (e.g., Marhas et al., 2007, Ávila et al., 2013) enabling complete sample dissolution. The digestion techniques employed in earlier studies on Ba isotopes in meteorites (Hidaka et al., 2003; Andreasen and Sharma, 2007; Carlson et al., 2007) were designed to dissolve SiC and other chemically resistant presolar phases, but complete dissolution was not documented. To determine if the pre-digestion laser treatment improved dissolution, two different samples of the Allende Smithsonian Reference Powder were analyzed: (a) Allende_{fused} (0.52 g) which was melted with the laser before acid digestion, and (b) Allende_{acid} (0.61 g) which underwent acid digestion only. Allende was chosen for this test

because it contains SiC that can alter its bulk Ba isotope composition if not digested (concentration of SiC in Allende ~ 0.01 ppm; Huss, 1995). A fine black residue (assumed to be a mixture of primarily C-bearing compounds and insoluble refractory grains) remained after Allende_{acid} was treated with two rounds of concentrated HF-HNO₃. In contrast, Allende_{fused} produced a clear solution after two rounds of HF-HNO₃ digestion. The Ba isotope composition of both Allende (a) and (b) are identical within analytical uncertainty (Fig. 4d), suggesting that there is no significant dependence of the Ba isotope composition on the digestion method, and that, for Allende, laser fusion is not required to obtain complete dissolution. This conclusion, however, need not apply to other meteorite groups (or isotope systems). The generally smaller magnitude of Ba isotope anomalies identified in the CM2 chondrites (e.g., Cold Bokkeveld and Mighei) here as compared to those found in previous studies may indicate that incomplete dissolution does slightly increase the magnitude (≤ 20 ppm) of ^{135}Ba anomalies for SiC-rich samples. The similarity in the Allende sample compositions may be because there is so little SiC in these samples that the effects of laser fusion are not pronounced. Conducting a comparable test using a SiC-rich sample (e.g., Murchison, CM2, SiC + spinel = 1640 ppm; Huss et al., 2003) may clarify this point, however, obtaining large amounts of such samples was not possible during this study.

4.4 Potential effects of radiogenic ^{135}Ba

Recent studies have discussed the potential contribution to ^{135}Ba compositions in acid leachates and Allende CAIs and from the decay of short lived ^{135}Cs ($^{135}\text{Ba}_{\text{radiogenic}} \sim 16 - 48$ ppm; Hidaka et al., 2001; Hidaka and Yoneda, 2011; Hidaka and Yoneda, 2013; Bermingham et al., 2014). The variation in ^{135}Ba observed in samples from the same parent body (Cold Bokkeveld and Mighei, CM2) and also in aliquots from the same meteorite (Orgueil) could result from variable contribution of ^{135}Cs decay to a pre-existing coupled ^{135}Ba and ^{137}Ba nucleosynthetic anomaly. If this is the cause of the variation in ^{135}Ba , there should be a corresponding variation in the suprachondritic Cs/Ba of these samples. For the carbonaceous chondrites samples analyzed here, however, Cs/Ba was below detection limit because of Cs loss during laser fusion. Carlson et al., (2007) did not report Cs/Ba for their samples. Thus the potential effect of radiogenic ^{135}Ba remains only a possible explanation for the variation in ^{135}Ba observed.

The quantity of radiogenic ^{135}Ba can be estimated from literature Cs/Ba values for whole rock samples of the relevant meteorites. Using the only available published data for the relevant carbonaceous chondrites (Allende, Mighei, Kainsaz, and Vigarano; Ahren et al., 1973; Krähenbühl et al. 1973; Anders et al., 1976; Wolf et al., 1980; Knab, 1981; Becker et al., 1987; Barrat et al., 2012), these samples all have subchondritic (0.01-0.06) Cs/Ba. Prima facie, this may suggest that there is not a radiogenic ^{135}Ba component in these samples. However, using Cs/Ba values from the literature for this application is tenuous because Cs and Ba can be mobilized by aqueous alteration (Hidaka and Yoneda, 2013; Bermingham et al., 2014). Thus, an accurate assessment of the potential radiogenic ^{135}Ba component can only be made from Ba isotope data and Cs/Ba measured on the same digested material. Moreover, because isotopic excesses of radiogenic ^{135}Ba are often masked by s-process nucleosynthetic anomalies and the Cs/Ba of bulk rocks is typically low, identifying isotopic evidence of radiogenic ^{135}Ba on the whole rock meteorite scale is difficult. In future studies, the physical separation of major presolar materials from meteorite samples before digestion might allow more precise estimates of radiogenic ^{135}Ba contributions.

If the variable abundance of ^{135}Ba identified in some whole rock meteorites is a result of a radiogenic ^{135}Ba component, this has important implications. First, it indicates that the CM2 and CI parent bodies formed in the early stages Solar System evolution (≤ 9 Myr after CAI formation), that is, within the lifetime of ^{135}Cs ($t_{1/2} = 2.3$ Myr; Hidaka et al, 2001). Second, it suggests that significant Cs/Ba fractionation, likely during parent body aqueous alteration, also occurred during these early stages. Supporting evidence for the early formation and onset of aqueous alteration (2.4 - 5 Myr after CAI formation) on parent bodies has been identified from Mn-Cr ages for carbonates in CM and CI chondrites (e.g., Fujiya et al., 2012, 2013; Jilly et al., 2014). The range in estimates of the $[^{135}\text{Cs}/^{133}\text{Cs}]_0$ from 1×10^{-5} to 2×10^{-3} (Hidaka et al., 2001; Nichols et al., 2002; Srinivasan et al., 2011; Hidaka and Yoneda, 2013), coupled with highly variable Cs/Ba for a limited number of carbonaceous chondrites (0.0141- 0.082; Rocholl and Jochum, 1993; Hidaka and Yoneda, 2013; Bermingham et al., 2014), makes it very difficult to estimate the potential amount of radiogenic Ba or meaningful ages for the onset of aqueous alteration. The large ranges in proposed $[^{135}\text{Cs}/^{133}\text{Cs}]_0$ and observed Cs/Ba may in fact reflect significant remobilization of Cs and/or Ba in the parent body during aqueous alteration (Hidaka and Yoneda, 2013; Bermingham et al., 2014). Obtaining a more precise initial ratio is critical in

establishing a more precise time for the onset of aqueous alteration on different parent bodies. Development and application of the ^{135}Cs - ^{135}Ba system can address outstanding questions regarding the timing of parent body formation and onset of aqueous alteration. Future work should include refinement of the $[\text{}^{135}\text{Cs}/\text{}^{133}\text{Cs}]_0$ and expansion of the current whole rock meteorite database for which high precision coupled Ba isotope and Cs/Ba data are available.

4.5 Sample size effect

The variation in the size of Ba isotope anomalies between different laser-fused aliquots of the same meteorite may reflect incomplete homogenization of the presolar or refractory component at the ~0.8 g (mass of Orgueil used in this study) sample scale in these meteorites. The effect of sample heterogeneity on the major and trace element composition of carbonaceous chondrites (e.g., Allende and Orgueil) has been discussed in detail by Morlok et al. (2006) and Stracke et al. (2012). Both studies indicate that there is a mass ‘threshold’ for the observance of strong chemical variation within single carbonaceous chondrites, with CAIs exerting a disproportionate control on the refractory element budget of a bulk meteorite sample when present in the sample. The ‘threshold’ at which homogeneous chemical compositions can be obtained has been reported to be about 1 - 2 g (Morlok et al., 2006).

The mass ‘threshold’ for *isotopic homogeneity*, however, has not been discussed widely in the literature. On the basis of our data, measurable variation in the magnitude of $^{135}\text{Ba}/^{138}\text{Ba}$ anomalies is observed in (fused) carbonaceous chondrite samples (specifically Orgueil) that are smaller than ~0.8 g. This variation is unlikely to only reflect an oversampling of refractory inclusions (e.g., CAIs), because the sample for which the most resolvable variation in ^{135}Ba is present (Orgueil), does not contain preserved CAIs. In contrast, the Ba isotope compositions of samples from the CV3 parent body, which have a high proportion of preserved CAIs, are generally more consistent than those of samples from the CM2 or CI parent bodies (Table 7). The cause for the variation in ^{135}Ba in the CM2 and CI fused samples is thus more likely a result of the incomplete heterogeneous distribution of a presolar component (and, potentially a radiogenic component) on the <0.8 g scale.

Samples that are ~0.8 g or less (Cold Bokkeveld, Mighei, Orgueil 1, Orgueil 2) and display variable ^{135}Ba compositions likely indicate that larger samples are required to obtain a

representative isotope composition. Determining the mass ‘threshold’ for isotopic homogeneity would best be done by analyzing multiple large aliquots (1 - 5 g) of samples rich in presolar material (e.g., Murchison) and comparing the Ba isotope composition of each fraction.

4.6 Significance of Ba isotope composition in bulk meteorites

Barium isotope data obtained from different carbonaceous chondrites for which complete dissolution was achieved indicates that there was a degree of heterogeneous distribution of presolar carriers throughout the Solar System. These new Ba isotope data confirm that the previously reported nucleosynthetic anomalies (Hidaka et al., 2003; Carlson et al., 2007; Andreasen and Sharma, 2007) were not a result of incomplete sample dissolution, although there may be a slight reduction in size of the anomaly with complete digestion. The origin of the coupled excesses in ^{135}Ba and ^{137}Ba likely result from an *s*-process deficit in the feeding zones of these parent bodies, as suggested by Hidaka et al. (2003), Carlson et al. (2007), Andreasen and Sharma (2007). These anomalies may be coupled with a minor radiogenic ^{135}Ba component in some meteorite samples; however, a more complete dataset is required to confirm this proposition.

The absence of resolvable Ba isotope anomalies in H chondrites, enstatite chondrites, eucrites, and Martian meteorites indicates that some parts of the Solar System were well mixed with respect to Ba isotopes. In addition, either ^{135}Cs was not present in these regions or there was insufficient Cs/Ba fractionation within the lifetime of ^{135}Cs to create measurable radiogenic Ba at the bulk sample scale (e.g., H-chondrites; Hidaka et al., 2001). Why some regions of the Solar System were homogenized with respect to presolar carriers for Ba whereas other regions were not remains an open question. One explanation is that those regions with homogeneous Ba isotope compositions represent larger and/or better-mixed feeding zones (e.g., Mars and the HED parent body). This would suggest that the presence of nucleosynthetic isotope anomalies on the whole rock meteorite scale in some samples may, in part, reflect a smaller parent body which formed from a less homogeneous feeding zone. Additional high precision Ba isotope data from large samples (>0.8 g) of carbonaceous chondrites and those samples for which there are currently no Ba isotope data (e.g., primitive achondrites) would aid in mapping out the Ba isotope distribution throughout the Solar System.

The preservation of only small Ba isotope anomalies on the bulk sample scale indicates that the presolar grain carriers of highly isotopically anomalous Ba were fairly well mixed in the solar nebula, and even completely homogenized in some regions of the disk (e.g., feeding zones of the HED, H-chondrite, and enstatite chondrite parent bodies, and Mars). This well mixed composition is not evident in other isotopic systems, for example Ti, Cr, Mo, and Ru, which display much larger ($>100 \mu$) nucleosynthetic anomalies in bulk meteorites (Trinquier et al., 2009; Qin et al., 2010; Burkhardt et al., 2011; Fischer-Gödde et al., 2015). The preservation of larger anomalies in these elements but not in Ba provides important constraints. It indicates that the carrier(s) of Ba isotope anomalies were decoupled from those of Ti, Cr, Mo, and Ru. This is not unexpected, because studies have indicated that the main carriers of these elements are likely not SiC (e.g., Trinquier et al., 2009; Qin et al., 2010; Burkhardt et al., 2012; Fischer-Gödde et al., 2015). The preservation of only small isotope anomalies in the Ba isotope system also indicates that the Ba isotope carriers were better mixed in the solar nebula than those for Ti, Cr, Mo, and Ru. The mechanism(s) responsible for spatially variable homogenization is open for debate. Viable mechanisms include 1) variable mixing of different presolar carriers within the solar nebula, or 2) emplacement of Ba isotope carriers into the nebula-forming region before some isotope carriers (e.g., Ti, Cr) thus allowing more time for them to homogenize. Additionally, the Ba isotope carriers may have been more robust and thus resistant to chemical fractionation thereby escaping thermal processing in the nebula, which imparted isotope variations in some isotope systems (e.g., Ti and Mo; Trinquier et al., 2009; Burkhardt et al., 2012). Clearly more work is required to elucidate the processes responsible for generating the origins of nucleosynthetic anomalies in a number of isotope systems and this can only be done using data for which it is confirmed that there are no adverse effects of incomplete sample dissolution.

5. CONCLUSIONS

This study reports the first Ba isotope compositions for carbonaceous chondrites for which complete dissolution was certain. Resolvable anomalies in ^{135}Ba and ^{137}Ba are identified in most of these samples and are generally similar to the previously reported Ba isotope compositions from carbonaceous chondrites for which complete chemical digestion may not have been achieved. The common slight reduction in magnitude of Ba isotope anomalies in fused samples

from the CM2 group as compared to compositions of non-fused samples reported in Carlson et al. (2007) may indicate that incomplete dissolution of presolar grains slightly increases the magnitude (<20 ppm) of Ba isotope variability in bulk meteorite samples that have high presolar grain concentrations. This conclusion could also apply to other isotope systems (e.g., Ti, Cr, Ni) and similar tests should be performed on comparable samples to test for the presence of nucleosynthetic anomalies and their dependence on dissolution protocol. The slight reduction in Ba isotope variation seen in the present dataset when compared to published data could also be partly explained by sampling bias. Evidence for this comes from the variations in the size of the ^{135}Ba isotope anomalies identified in fused samples from the same parent body and also from different pieces of the same meteorite on the <0.8 g sampling scale. The cause of this variation in the ^{135}Ba anomaly likely results from the heterogeneous distribution of the presolar component (e.g., SiC) on the sample scale of this study (<0.8 g) and potentially from a small contribution of radiogenic ^{135}Ba . When looking closely, this variation in ^{135}Ba is also evident in published datasets where the samples have not undergone fusion before digestion. Future work to further constrain the cause(s) of ^{135}Ba variability in whole rock samples would lie in measurement of Ba isotope compositions in larger fused samples (>0.8 g) coupled with Cs/Ba measurements on the same aliquots.

The origin of the nucleosynthetic isotope anomalies in ^{135}Ba and ^{137}Ba is likely a result of an s-process deficit in the feeding zones of the respective parent bodies. Regions with Ba isotope homogeneity in the early Solar System were present as evidenced by the Ba isotope compositions of H-chondrites, enstatite chondrites, eucrites, and Martian meteorites, which are all indistinguishable from terrestrial values. The origin of these isotopically homogeneous regions may represent a larger and, therefore, better mixed feeding zone. The preservation of only small Ba isotope anomalies on the bulk sample scale suggests that the highly anomalous Ba isotope presolar grain carriers were fairly well mixed in the solar nebula as compared to the Ti, Cr, Mo, and Ru isotope carriers. This indicates that Ba isotope carriers were decoupled from those of Ti, Cr, Mo, and Ru. Perhaps the Ba isotope carriers either escaped destruction during nebular processing or they were emplaced into the solar nebula-forming region before the Ti, Cr, Mo, and Ru isotope carriers thus had a longer time to homogenize.

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Table 1.

Natural abundances, nucleosynthetic origin, and neutron capture cross sections for Ba isotopes and ¹³⁵Cs.

Isotope	Abundance (%) [*]	Nucleosynthetic origin [†]	Neutron cross section [^] (barns)
¹³⁰ Ba	0.106	p (100)	8.7 ± 0.9
¹³² Ba	0.101	p (100)	6.5 ± 0.8
¹³⁴ Ba	2.417	s (100)	1.5 ± 0.3
¹³⁵ Ba	6.592	s (26), r (74)	5.8 ± 0.9
¹³⁶ Ba	7.854	s (100)	0.68 ± 0.2
¹³⁷ Ba	11.23	s (64), r (36)	3.6 ± 0.2
¹³⁸ Ba	71.70	s (87), r (13)	0.40 ± 0.04
¹³⁵ Cs	-	s, r	-

For each isotope, the main nucleosynthetic process and the percentage proportion that it represents are shown in parentheses (except for ¹³⁵Cs).

^{*} Present day natural isotope abundances (De Bièvre and Taylor, 1993)

[†] Nucleosynthetic proportions of Ba isotopes (Arlandini et al., 1999)

[^] Thermal neutron capture cross sections (Mughabghab, 2003)

Table 2. List of studied meteorites.

Sample	Type	Ba (ppm)	Mass (g)	Laser fusion	Laboratory	Cosmic ray exposure age (Ma)
<i>Terrestrial basalt</i>						
DU17	Basalt	33	0.104	N	WWU	N/A
<i>HED</i>						
Juvinas	EUC	30	0.072	N	WWU	11
Stannern [§] _{fine}	EUC	53	0.097	N	WWU	35
Stannern [§] _{coarse}	EUC	53	0.099	N	WWU	35
<i>Ordinary chondrites</i>						
Oesede	H5	-	0.504	N	WWU	41
Tugalin-Bulen	H6	-	0.456	N	WWU	20
Butsura	H6	1.1	0.537	N	WWU	41
Guenie-Gao	H4/5	-	0.475	Y	WWU	10
<i>Enstatite chondrites</i>						
Indarch	EH4	2.3	0.443	Y	WWU	6.5-16
<i>Carbonaceous chondrites</i>						
Orgueil 1	CI	2.3	0.738	Y	WWU	4.5
Orgueil 2	CI	2.3	0.821	Y	DTM	4.5
Renazzo [#]	CR2	3.4	0.844	Y	WWU	9
Cold Bokkeveld	CM2	-	0.725	Y	DTM	0.3
Mighei	CM2	2.5	0.685	Y	DTM	4.0
Murchison	CM2	1.6	0.598	Y	WWU	4.3
Allende _{acid}	CV3(ox)	5.3	0.516	N	DTM	1.6-5.5
Allende _{fused}	CV3(ox)	5.3	0.610	Y	DTM	1.6-5.5
Vigarano	CV3 (red)	5.2	0.482	Y	DTM	-
Lancé	CO3.5	-	0.473	Y	DTM	3.6
Kainsaz	CO3.2	3.9	0.946	Y	DTM	20.9
<i>SNC</i>						
Zagami	Shergottite	26	0.067	N	WWU	2.9
Shergotty (USNM 6676)	Shergottite	34	0.053	N	WWU	3.0
Los Angeles 001 [†] (USNM 7058)	Shergottite	47	0.076	N	WWU	3.1
NWA 4864 [†]	Shergottite	26	0.070	N	WWU	-
Nakhla	Nakhlite	32	0.055	N	WWU	11
MIL 03346 [†] (NASA, subsample 193)	Nakhlite	26	0.072	N	WWU	9.5

Type of meteorite, bulk sample Ba estimated from previous publications, mass of sample analyzed, indication of analytical treatment, laboratory where samples were processed, and sample repository location are listed. All samples are falls except for those marked ([†]).

[§] Fine or coarse-grained texture evident within whole rock chip

[#] Mineral separates (non-metal fraction)

Table 3. The Ba isotope data for terrestrial basalt (DU17), eucrites, and ordinary chondrites analyzed in the present study.

	$^{132}\text{Ba}/^{138}\text{Ba}$	$^{135}\text{Ba}/^{138}\text{Ba}$	$^{136}\text{Ba}/^{138}\text{Ba}$	$^{137}\text{Ba}/^{138}\text{Ba}$
Ba standard				
Ba std 01	0.00141295 (4)	0.0919379 (3)	0.1095349 (3)	0.1565298 (2)
Ba std 02	0.00141325 (4)	0.0919386 (3)	0.1095352 (2)	0.1565306 (2)
Ba std 03	0.00141319 (4)	0.0919377 (3)	0.1095348 (3)	0.1565297 (2)
Ba std 04	0.00141338 (4)	0.0919376 (2)	0.1095325 (2)	0.1565287 (2)
Ba std 05	0.00141343 (4)	0.0919377 (2)	0.1095335 (2)	0.1565296 (2)
Ba std 06	0.00141334 (4)	0.0919375 (3)	0.1095356 (2)	0.1565289 (2)
Ba std 07	0.00141345 (4)	0.0919386 (3)	0.1095342 (2)	0.1565303 (2)
Ba std 08	0.00141333 (4)	0.0919378 (2)	0.1095326 (2)	0.1565304 (2)
Ba std 09	0.00141347 (3)	0.0919368 (2)	0.1095332 (2)	0.1565313 (2)
Average	0.00141331	0.0919378	0.1095340	0.1565299
<i>External precision (2σ, ppm)</i>	229	12	21	11
DU17A 01	0.00141342 (4)	0.0919369 (3)	0.1095351 (2)	0.1565291 (2)
DU17A 02	0.00141328 (5)	0.0919374 (3)	0.1095311 (3)	0.1565302 (2)
DU17A 03	0.00141342 (5)	0.0919379 (3)	0.1095346 (2)	0.1565298 (3)
DU17B 01	0.00141330 (4)	0.0919383 (3)	0.1095342 (3)	0.1565304 (3)
<i>Average deviation from Ba std (ppm)</i>	31 (± 107)	-2 (± 13)	-1 (± 25)	0 (± 7)
Juvinas 01	0.00141328 (4)	0.0919382 (3)	0.1095356 (2)	0.1565306 (3)
Juvinas 02	0.00141333 (4)	0.0919380 (3)	0.1095351 (2)	0.1565306 (2)
Juvinas 03	0.00141350 (3)	0.0919381 (2)	0.1095360 (2)	0.1565305 (2)
Juvinas 04	0.00141320 (4)	0.0919384 (3)	0.1095350 (2)	0.1565308 (2)
<i>Average deviation from Ba std (ppm)</i>	12 (± 181)	4 (± 4)	13 (± 9)	4 (± 1)
Stannern (C) 01	0.00141335 (4)	0.0919378 (2)	0.1095347 (2)	0.1565296 (2)
Stannern (C) 02	0.00141346 (4)	0.0919386 (2)	0.1095363 (2)	0.1565306 (2)
Stannern (C) 03	0.00141333 (4)	0.0919385 (2)	0.1095342 (2)	0.1565306 (3)
Stannern (C) 04	0.00141319 (4)	0.0919382 (3)	0.1095349 (2)	0.1565308 (2)
<i>Average deviation from Ba std (ppm)</i>	47 (± 155)	5 (± 8)	9 (± 17)	3 (± 7)
Stannern (F) 01	0.00141334 (4)	0.0919378 (3)	0.1095343 (2)	0.1565299 (2)
Stannern (F) 03	0.00141333 (4)	0.0919378 (2)	0.1095348 (2)	0.1565302 (2)
Stannern (F) 04	0.00141345 (4)	0.0919381 (2)	0.1095359 (2)	0.1565299 (2)
Stannern (F) 05	0.00141338 (3)	0.0919381 (2)	0.1095341 (2)	0.1565306 (2)
<i>Average deviation from Ba std (ppm)</i>	47 (± 77)	4 (± 3)	11 (± 15)	4 (± 5)
Butsura 01	0.00141360 (4)	0.0919373 (2)	0.1095360 (3)	0.1565307 (2)
Butsura 02	0.00141359 (4)	0.0919365 (3)	0.1095316 (2)	0.1565297 (2)
<i>Average deviation from Ba std (ppm)</i>	200 (± 7)	-10 (± 13)	-2 (± 56)	2 (± 8)
Oesede 01	0.00141341 (4)	0.0919356 (3)	0.1095379 (3)	0.1565281 (3)
Oesede 02	0.00141339 (5)	0.0919365 (3)	0.1095369 (3)	0.1565283 (3)
<i>Average deviation from Ba std (ppm)</i>	-37 (± 18)	-3 (± 13)	16 (± 13)	-3 (± 2)
Tugalin-Bulen 01	0.0014135 (5)	0.0919383 (3)	0.1095330 (3)	0.1565314 (3)
Tugalin-Bulen 02	0.0014134 (4)	0.0919367 (3)	0.1095361 (3)	0.1565287 (3)
<i>Average deviation from Ba std (ppm)</i>	117 (± 126)	-3 (± 24)	4 (± 41)	1 (± 25)

Barium data are normalized to $^{134}\text{Ba}/^{138}\text{Ba} = 0.03371$ (De Bièvre and Taylor, 1993; Andreassen and Sharma, 2007). Uncertainties in the last digits in parentheses are 2 s.e. of integrations within an analysis. The external precision is based on 2 σ of 9 replicate standard measurements. Number of integrations per analysis (n) is 320. Values (in parentheses) after the average deviation from the Ba standard values are 2 σ of the replicate samples analyses in ppm where more than one analysis was possible. When only one analysis was performed, the values in parentheses after the average deviation from the Ba standard values represent 2 σ of the replicate standards analyzed (in ppm).

Table 4.

The Ba isotope data from Martian meteorites analyzed in the present study.

	$^{132}\text{Ba}/^{138}\text{Ba}$	$^{135}\text{Ba}/^{138}\text{Ba}$	$^{136}\text{Ba}/^{138}\text{Ba}$	$^{137}\text{Ba}/^{138}\text{Ba}$
Ba standard				
Ba std 01	0.00141347 (4)	0.0919372 (2)	0.1095353 (2)	0.1565289 (2)
Ba std 02	0.00141354 (4)	0.0919377 (2)	0.1095354 (2)	0.1565282 (2)
Ba std 03	0.00141353 (4)	0.0919370 (2)	0.1095348 (2)	0.1565267 (2)
Ba std 04	0.00141357 (4)	0.0919383 (2)	0.1095350 (2)	0.1565287 (2)
Ba std 05	0.00141346 (4)	0.0919367 (2)	0.1095333 (2)	0.1565264 (2)
Ba std 06	0.00141328 (4)	0.0919379 (3)	0.1095355 (2)	0.1565269 (2)
Average	0.00141348	0.0919375	0.1095349	0.1565277
<i>External precision (2σ, ppm)</i>	149	13	15	14
Los Angeles 001	0.00141347 (4)	0.0919369 (3)	0.1095363 (2)	0.1565279 (2)
<i>Average deviation from Ba std (ppm)</i>	-1 ($\pm$ 149)	-7 ($\pm$ 13)	13 ($\pm$ 15)	1 ($\pm$ 14)
NWA4864	0.00141360 (4)	0.0919381 (3)	0.1095338 (2)	0.1565285 (3)
<i>Average deviation from Ba std (ppm)</i>	89 ($\pm$ 149)	7 ($\pm$ 13)	-10 ($\pm$ 15)	6 ($\pm$ 14)
MIL03346	0.00141343 (6)	0.0919385 (3)	0.1095347 (3)	0.1565297 (3)
<i>Average deviation from Ba std (ppm)</i>	-29 ($\pm$ 149)	12 ($\pm$ 13)	-1 ($\pm$ 15)	13 ($\pm$ 14)
Nakhla 01	0.00141361 (4)	0.0919367 (2)	0.1095375 (2)	0.1565276 (2)
Nakhla 02	0.00141336 (4)	0.0919365 (2)	0.1095364 (2)	0.1565264 (2)
<i>Average deviation from Ba std (ppm)</i>	5 ($\pm$ 253)	-10 ($\pm$ 4)	19 ($\pm$ 14)	-4 ($\pm$ 12)
Zagami 01	0.00141370 (4)	0.0919376 (3)	0.1095368 (2)	0.1565288 (2)
Zagami 02	0.00141347 (4)	0.0919376 (2)	0.1095362 (2)	0.1565282 (2)
<i>Average deviation from Ba std (ppm)</i>	80 ($\pm$ 232)	2 ($\pm$ 1)	15 ($\pm$ 8)	5 ($\pm$ 6)
Shergotty	0.00141355 (4)	0.0919385 (2)	0.10953537 (2)	0.1565278 (2)
<i>Average deviation from Ba std (ppm)</i>	55 ($\pm$ 149)	11 ($\pm$ 13)	4 ($\pm$ 15)	1 ($\pm$ 14)

Barium data are normalized to $^{134}\text{Ba}/^{138}\text{Ba} = 0.03371$ (De Bièvre and Taylor, 1993; Andreasen and Sharma, 2007). Uncertainties in the last digits in parentheses are 2 s.e. of integrations during an analysis. The external precision is based on 2 σ of 6 replicate standard measurements. Number of integrations per analysis (n) is 320. Values (in parentheses) after the average deviation from the Ba standard values are 2 σ of the replicate samples analyses in ppm or 2 σ of 6 replicate standard measurements (in ppm) when no replicate analyses were available.

Table 5. The Ba isotope data from carbonaceous chondrites, enstatite chondrites, thermally unequilibrated meteorites analyzed in the present study.

	$^{132}\text{Ba}/^{138}\text{Ba}$	$^{135}\text{Ba}/^{138}\text{Ba}$	$^{136}\text{Ba}/^{138}\text{Ba}$	$^{137}\text{Ba}/^{138}\text{Ba}$
Ba standards				
Ba std 01	0.00141353 (4)	0.0919370 (2)	0.1095348 (2)	0.1565267 (2)
Ba std 02	0.00141357 (4)	0.0919383 (2)	0.1095350 (2)	0.1565287 (2)
Ba std 03	0.00141337 (4)	0.0919382 (2)	0.1095346 (2)	0.1565278 (2)
Ba std 04	0.00141346 (4)	0.0919367 (2)	0.1095333 (2)	0.1565264 (2)
Ba std 05	0.00141338 (4)	0.0919372 (2)	0.1095373 (2)	0.1565272 (2)
Ba std 06	0.00141338 (4)	0.0919376 (2)	0.1095320 (2)	0.1565273 (2)
Ba std 07	0.00141335 (3)	0.0919379 (2)	0.1095357 (2)	0.1565259 (2)
Ba std 08	0.00141328 (4)	0.0919379 (3)	0.1095355 (2)	0.1565269 (2)
Average	0.00141341	0.0919376	0.1095348	0.1565271
<i>External precision (2σ, ppm)</i>	138	12	29	11
Renazzo 01	0.00141326 (4)	0.0919390 (3)	0.1095334 (3)	0.1565281 (3)
Renazzo 02 (n=200)	0.00141323 (5)	0.0919398 (4)	0.1095367 (4)	0.1565286 (3)
<i>Av. dev. from Ba std (ppm)</i>	-120 ($\pm$ 22)	20 ($\pm$ 12)	2 ($\pm$ 43)	8 ($\pm$ 4)
Orgueil 1	0.00141357 (4)	0.0337039 (2)	0.0919425 (2)	0.1095327 (2)
<i>Av. dev. from Ba std (ppm)</i>	109 ($\pm$ 138)	53 ($\pm$ 12)	-19 ($\pm$ 29)	23 ($\pm$ 11)
Gao-Guenie	0.00141366 (4)	0.0919381 (3)	0.1095356 (2)	0.1565273 (2)
<i>Av. dev. from Ba std (ppm)</i>	176 ($\pm$ 138)	5 ($\pm$ 12)	8 ($\pm$ 29)	1 ($\pm$ 11)
Murchison	0.00141336 (4)	0.0919389 (2)	0.1095344 (3)	0.1565273 (3)
<i>Av. dev. from Ba std (ppm)</i>	-39 ($\pm$ 138)	14 ($\pm$ 12)	-4 ($\pm$ 29)	1 ($\pm$ 11)
Indarch (n=170)	0.00141328 (9)	0.0919385 (4)	0.1095309 (5)	0.1565292 (4)
<i>Av. dev. from Ba std (ppm)</i>	-93 ($\pm$ 138)	10 ($\pm$ 12)	-35 ($\pm$ 29)	13 ($\pm$ 11)

Barium data are normalized to $^{134}\text{Ba}/^{138}\text{Ba} = 0.03371$ (De Bièvre and Taylor, 1993; Andreasen and Sharma, 2007). Uncertainties in the last digits (in parentheses) are 2 s.e. of integrations during an analysis. The external precision is based on 2 σ of 8 replicate standard measurements. Number of integrations per analysis (n) is 320. Values (in parentheses) after the average deviation from the Ba standard values are 2 σ of the replicate samples analyses in ppm.

Table 6. The Ba isotope data from carbonaceous chondrites analyzed in the present study.

	$^{130}\text{Ba}/^{138}\text{Ba}$	$^{132}\text{Ba}/^{138}\text{Ba}$	$^{135}\text{Ba}/^{138}\text{Ba}$	$^{136}\text{Ba}/^{138}\text{Ba}$	$^{137}\text{Ba}/^{138}\text{Ba}$
Ba standard					
Ba std 01	0.00147580 (5)	0.00141226 (5)	0.0919090 (3)	0.1095073 (2)	0.1565203 (3)
Ba std 02	0.00147556 (3)	0.00141228 (3)	0.0919091 (2)	0.1095072 (2)	0.1565193 (2)
Ba std 03	0.00147570 (3)	0.00141225 (3)	0.0919092 (2)	0.1095082 (2)	0.1565203 (2)
Ba std 04	0.00147578 (3)	0.00141227 (3)	0.0919097 (2)	0.1095048 (2)	0.1565203 (2)
Ba std 05	0.00147575 (3)	0.00141228 (3)	0.0919097 (2)	0.1095084 (2)	0.1565201 (2)
Ba std 06	0.00147569 (3)	0.00141231 (3)	0.0919094 (2)	0.1095069 (2)	0.1565211 (2)
Ba std 07	0.00147572 (3)	0.00141226 (3)	0.0919093 (2)	0.1095077 (2)	0.1565210 (2)
Ba std 08	0.00147575 (3)	0.00141218 (2)	0.0919096 (2)	0.1095055 (2)	0.1565215 (2)
Ba std 09	0.00147576 (3)	0.00141227 (3)	0.0919093 (2)	0.1095060 (2)	0.1565211 (2)
Ba std 10	0.00147581 (3)	0.00141223 (2)	0.0919096 (2)	0.1095059 (2)	0.1565206 (2)
Ba std 11	0.00147573 (3)	0.00141218 (2)	0.0919090 (2)	0.1095059 (2)	0.1565203 (2)
Ba std 12	0.00147570 (4)	0.00141236 (3)	0.0919098 (3)	0.1095059 (2)	0.1565209 (2)
Ba std 13	0.00147585 (2)	0.00141229 (2)	0.0919088 (2)	0.1095042 (1)	0.1565200 (1)
Ba std 14	0.00147572 (3)	0.00141245 (3)	0.0919088 (2)	0.1095075 (2)	0.1565197 (2)
Ba std 15	0.00147564 (3)	0.00141219 (3)	0.0919089 (2)	0.1095077 (2)	0.1565202 (2)
Ba std 16	0.00147583 (3)	0.00141230 (3)	0.0919094 (2)	0.1095083 (2)	0.1565205 (2)
Ba std 17	0.00147576 (3)	0.00141240 (3)	0.0919087 (2)	0.1095071 (2)	0.1565203 (2)
Ba std 18	0.00147572 (2)	0.00141212 (2)	0.0919095 (2)	0.1095059 (2)	0.1565211 (2)
Ba std 19	0.00147576 (2)	0.00141230 (2)	0.0919094 (2)	0.1095061 (1)	0.1565215 (1)
Ba std 20	0.00147571 (2)	0.00141224 (2)	0.0919092 (2)	0.1095057 (2)	0.1565206 (2)
Ba std 21	0.00147583 (4)	0.00141210 (4)	0.0919096 (3)	0.1095069 (3)	0.1565213 (3)
Average	0.00147574	0.00141226	0.0919093	0.1095066	0.1565206
External precision (2 σ , ppm)	92	117	7	21	7
Allende_acid 01	0.00147578 (3)	0.00141228 (3)	0.0919126 (2)	0.1095089 (2)	0.1565235 (2)
Allende_acid 02	0.00147561 (4)	0.00141237 (4)	0.0919119 (2)	0.1095076 (2)	0.1565232 (2)
Av. dev. from Ba std (ppm)	-33 (± 160)	41 (± 89)	32 (± 11)	15 (± 17)	18 (± 2)
Allende_fused 01	0.00147574 (2)	0.00141218 (2)	0.0919130 (2)	0.1095080 (2)	0.1565242 (2)
Allende_fused 02 (n=280)	0.00147554 (3)	0.00141219 (3)	0.0919123 (2)	0.1095074 (2)	0.1565233 (2)
Av. dev. from Ba std (ppm)	-67 (± 188)	-54 (± 13)	37 (± 11)	10 (± 7)	20 (± 8)
Cold Bokkeveld 01	0.00147568 (3)	0.00141222 (3)	0.0919125 (2)	0.1095066 (2)	0.1565229 (2)
Cold Bokkeveld 02	0.00147580 (3)	0.00141224 (2)	0.0919122 (2)	0.1095067 (2)	0.1565228 (2)
Cold Bokkeveld 03	0.00147561 (2)	0.00141223 (3)	0.0919124 (2)	0.1095070 (2)	0.1565231 (2)
Av. dev. from Ba std (ppm)	-29 (± 125)	-23 (± 18)	34 (± 3)	1 (± 3)	15 (± 3)
Mighei (n=480)	0.00147578 (3)	0.00141217 (3)	0.0919106 (2)	0.1095074 (2)	0.1565220 (2)
Av. dev. from Ba std (ppm)	25 (± 92)	-65 (± 117)	14 (± 7)	7 (± 21)	9 (± 7)
Kainsaz 01	0.00147572 (3)	0.00141218 (3)	0.0919125 (2)	0.1095061 (2)	0.1565233 (2)
Kainsaz 02 (n=330)	0.00147568 (5)	0.00141230 (4)	0.0919124 (4)	0.1095052 (4)	0.1565227 (4)
Av. dev. from Ba std (ppm)	-26 (± 39)	-16 (± 125)	34 (± 1)	-9 (± 12)	16 (± 5)
Lancé 01	0.00147574 (3)	0.00141225 (3)	0.0919125 (2)	0.1095054 (2)	0.1565231 (2)
Lancé 02	0.00147562 (2)	0.00141228 (2)	0.0919129 (2)	0.1095069 (2)	0.1565240 (2)
Av. dev. from Ba std (ppm)	-44 (± 118)	3 (± 29)	37 (± 7)	-4 (± 19)	19 (± 8)
Vigarano 01	0.00147575 (3)	0.00141223 (3)	0.0919122 (2)	0.1095047 (2)	0.1565231 (2)
Vigarano 02	0.00147589 (2)	0.00141220 (2)	0.0919126 (2)	0.1095047 (2)	0.1565232 (2)
Vigarano 03	0.00147563 (3)	0.00141211 (3)	0.0919134 (2)	0.1095068 (2)	0.1565243 (2)
Av. dev. from Ba std (ppm)	10 (± 178)	-58 (± 89)	38 (± 13)	-11 (± 23)	19 (± 9)
Orgueil 01	0.00147566 (3)	0.00141223 (3)	0.0919113 (2)	0.1095068 (2)	0.1565228 (2)
Orgueil 02	0.00147535 (3)	0.00141217 (3)	0.0919113 (2)	0.1095077 (2)	0.1565234 (2)
Av. dev. from Ba std (ppm)	-161 (± 295)	-44 (± 67)	22 (± 1)	6 (± 12)	16 (± 5)

Barium data are normalized to $^{134}\text{Ba}/^{138}\text{Ba} = 0.03371$ (De Bièvre and Taylor, 1993; Andreasen and Sharma, 2007). Uncertainties in the last digits (in parentheses) are 2 s.e. of integrations during an analysis. The external precision is based on 2 σ of 21 replicate

standard measurements. Number of integrations per analysis (n) is 540. Values (in parentheses) after the average deviation from the Ba standard values are 2σ of the replicate samples analyses in ppm.

Table 7. Comparison of Ba isotope compositions of different aliquots from the same meteorites from present study and Carlson et al. (2007). Values (in parentheses) are 2σ of the replicate standard analyses in ppm for that analytical session. All data normalized using $^{134}\text{Ba}/^{138}\text{Ba} = 0.03371$ (De Bièvre and Taylor, 1993; Andreassen and Sharma, 2007).

	$^{130}\text{Ba}/^{138}\text{Ba}$	$^{132}\text{Ba}/^{138}\text{Ba}$	$^{135}\text{Ba}/^{138}\text{Ba}$	$^{136}\text{Ba}/^{138}\text{Ba}$	$^{137}\text{Ba}/^{138}\text{Ba}$	Source
CM2						
Cold Bokkeveld	-29 (± 92)	-23 (± 117)	34 (± 7)	1 (± 21)	15 (± 7)	WWU
Cold Bokkeveld	174 (± 108)	102 (± 204)	53 (± 11)	-6 (± 12)	23 (± 12)	Carlson et al. (2007)
Mighei	25 (± 92)	-65 (± 117)	14 (± 7)	7 (± 21)	9 (± 7)	DTM
Mighei	-12 (± 108)	107 (± 204)	24 (± 11)	-4 (± 12)	15 (± 12)	Carlson et al. (2007)
Murray	129 (± 108)	109 (± 204)	11 (± 11)	-1 (± 12)	9 (± 12)	Carlson et al. (2007)
CV3_{ox}						
Allende _{fused}	-67 (± 92)	-54 (± 117)	37 (± 7)	10 (± 21)	20 (± 7)	DTM
Allende _{acid}	-33 (± 92)	41 (± 117)	32 (± 7)	15 (± 21)	18 (± 7)	DTM
Grosnaja	64 (± 108)	189 (± 204)	38 (± 11)	-3 (± 12)	15 (± 12)	Carlson et al. (2007)
Mokoia	58 (± 108)	164 (± 204)	-14 (± 11)	-2 (± 12)	-4 (± 12)	Carlson et al. (2007)
Vigarano _{red}	10 (± 92)	-58 (± 117)	38 (± 7)	-11 (± 21)	19 (± 7)	DTM
CI						
Orgueil 1	-	109 (± 138)	53 (± 12)	-19 (± 29)	23 (± 11)	WWU
Orgueil 2	-161 (± 92)	-44 (± 117)	22 (± 7)	6 (± 21)	16 (± 7)	DTM
Orgueil	-39 (± 108)	168 (± 204)	31 (± 11)	8 (± 12)	15 (± 12)	Carlson et al. (2007)

Fig. 1
The Ba isotopic composition of SiC grains from the KJB fraction (Murchison SiC-enriched sample), where $\delta^i\text{Ba}/^{136}\text{Ba} = [(^i\text{Ba}/^{136}\text{Ba})_{\text{measured}}/(^i\text{Ba}/^{136}\text{Ba})_{\text{solar}} - 1] \times 10^3$. Errors are 1σ and are smaller than the symbols. Data is from Ávila et al. (2013).

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Fig. 2

Back scatter electron images of (a) a ~10 mg sample of pure, synthetic SiC that was exposed to the laser for ~3 s. Partial melting of the SiC is evident; (b) a ~10 mg aliquot of Lancé which was exposed to laser fusion for ~15 s and displays a quenched texture, indicating complete melting of the sample.

Fig. 3

The average Ba isotope ratios (in ppm) of individual filament measurements of (a) terrestrial basalt (DU17) and (b) a selection of eucrites. Barium data are normalized to $^{134}\text{Ba}/^{138}\text{Ba} = 0.03371$ (De Bièvre and Taylor, 1993; Andreassen and Sharma, 2007). The shaded area indicates the external precision as determined from the 2σ of the population of replicate standard measurements. The error bars represent the 2σ of the replicate sample analyses in ppm where multiple measurements of the same sample were possible. Where only one measurement of the sample was possible, the error bars represent 2σ of the replicate standard analyses in ppm. The values and errors for ^{132}Ba data have been divided by 10 to plot on this scale.

Fig. 4

The average Ba isotope ratios (in ppm) of individual filament measurements of (a) ordinary chondrites, (b) Martin meteorites, and (c-e) carbonaceous and enstatite chondrites. Barium data are normalized to $^{134}\text{Ba}/^{138}\text{Ba} = 0.03371$ (De Bièvre and Taylor, 1993; Andreasen and Sharma, 2007). The shaded area indicates the external precision as determined from the 2σ of the population of replicate standard measurements. The error bars represent the 2σ of the replicate sample analyses in ppm where multiple measurements of the same sample were possible. Where only one measurement of the sample was possible, the error bars represent 2σ of the replicate standard analyses in ppm. The values and errors for ^{130}Ba and ^{132}Ba data have been divided by 10 to plot on this scale. For (d) Allende_fused is the sample that underwent laser fusion prior to acid digestion. Allende_acid is the sample which did not undergo laser fusion prior to acid digestion.

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