



# Barium stable isotope composition of the Earth, meteorites, and calcium–aluminum-rich inclusions

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## ABSTRACT

High-precision stable Ba isotope ratios are reported in a variety of terrestrial samples, undifferentiated primitive meteorites, and calcium–aluminum-rich inclusions (CAIs) from the Allende chondrite. All whole-rock terrestrial and meteorite samples are isotopically indistinguishable at a 50 parts per million (ppm) level per atomic mass unit (amu). Three CAIs are isotopically light, with  $\delta^{138/137}\text{Ba}$  (permil deviation of the  $^{138}\text{Ba}/^{137}\text{Ba}$  ratio from a terrestrial standard) values down to  $-0.6\text{‰}$  compared to whole-rock meteorites, whereas the matrix is enriched in heavy isotopes ( $\delta^{138/137}\text{Ba}$ :  $+0.2\text{‰}$ ). Similar light isotope enrichments in CAIs have been previously observed for Eu, Sr, and Ca, while for most other elements CAIs are enriched in the heavier isotopes (e.g. Mg, Fe). Kinetic isotopic fractionation is a possible explanation for the enrichment in the lightest isotopes, either by condensation from a vapor phase enriched in light isotopes by kinetic effects or by kinetic fractionation during non-equilibrium condensation of an undercooled gas as suggested for Ca isotopes. However, the common property of Ba, Eu, and Sr is that they all have a low first ionization potential. We suggest that electromagnetic sorting of ionized species in the early Solar System is a possible alternative mechanism to explain the depletion in heavy isotopes observed in refractory inclusions for those elements.

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## 1. Introduction

Isotopic variability between Solar System materials is caused by a few general mechanisms: 1) Stable isotope fractionation, which originates from vibrational energy partitioning between coexisting phases (e.g., Bigeleisen and Mayer, 1947). This process is typically proportional to the mass difference between the isotopes and inversely proportional to the square of the temperature; however, non-mass-dependent effects have been observed in some specific systems (e.g., Fujii et al., 2006a, 2006b; Moynier et al., 2013; Thiemens, 1999). 2) Nuclear reactions such as the decay of long-lived (e.g.  $^{87}\text{Rb}$ – $^{87}\text{Sr}$ ) or short-lived (e.g.  $^{26}\text{Al}$ – $^{26}\text{Mg}$ ) radioactive isotopes or spallation by cosmic rays or solar wind. 3) Inheritance of isotopic heterogeneities originating from an incomplete mixing of nucleosynthetic products. Such inherited isotopic heterogeneities are observed after correction of isotopic ratios for stable isotope fractionation, radioactive decay, and spallation effects. Only for elements with three or more isotopes can one clearly distinguish between mass-dependent and mass-independent processes. Mass-dependent isotopic variations in meteorites and planetary bodies

are attributable to processes that fractionated isotopes during either nebular (e.g. Luck et al., 2003; Moynier et al., 2006) or planetary events (e.g. Georg et al., 2007).

It is known from observation that the Solar System evolved through a solar protoplanetary disk phase after the collapse of the proto-solar cloud (Dullemond and Monnier, 2010). The lifetime of the protoplanetary disk was very short (a few million years) and it requires transport of mass inward and angular momentum outward. All the mechanisms proposed to explain this transport (e.g. magnetorotational instabilities, Balbus and Hawley, 1991; Wardle, 2007) require an efficient coupling between the ionized gas and the cloud's magnetic field. The only observations of the presence of a magnetic field in the early Solar System come from the paleomagnetic study of meteorites (e.g. Cournede et al., 2014; Fu et al., 2014), from astronomical observations of young stellar objects (Stephens et al., 2014), and possibly by studying the isotopic composition of the Solar System's first solids, the calcium–aluminum-rich Inclusions (CAIs) (Moynier et al., 2006).

Refractory lithophile elements (RLE) are the least affected by planetary processes such as core formation and volatility effects (e.g. evaporation/condensation) and are therefore ideal tracers for studying the conditions and evolution of the solar nebula and for identifying the precursor materials that formed the Earth. For example, the alkaline earth elements Ca and Sr are isotopically fractionated between some groups

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of chondrites and terrestrial basalts, suggesting that carbonaceous chondrites cannot be the primary building blocks of Earth (Moynier et al., 2010; Simon and DePaolo, 2010; Valdes et al., 2014).

The CAIs are the oldest Solar System condensates, with ages of ~4.568 Ga (e.g. Bouvier et al., 2007; Connelly et al., 2008, 2012; Jacobsen et al., 2008; Amelin et al., 2010; Bouvier and Wadhwa, 2010). Their origin and evolution involved condensation from the solar nebula followed by various thermal modifications, including high-T melting and shock reheating (e.g. Davis and Grossman, 1979; Grossman, 1972; Wark and Boynton, 2001). The complex thermal history of CAIs is likely to have resulted in massive evaporative loss, which is reflected by the heavy isotope enrichments observed for most elements (e.g. Mg, Si; Shahar and Young, 2007; Bullock et al., 2013). However, heavy isotope enrichments in CAIs is not ubiquitous; enrichments in the lighter isotopes of Sr (Moynier et al., 2010; Patchett, 1980b), Eu (Moynier et al., 2006), and Ca (Huang et al., 2011; Lee et al., 1978; Niederer and Papanastassiou, 1984) have also been observed. Two scenarios have been proposed to explain the unusual enrichment in light isotopes seen for Sr, Eu, and Ca: 1) kinetic isotope fractionation recorded by condensation of refractory solids and 2) electromagnetic sorting of isotopes in the nebular gas (Moynier et al., 2006).

Kinetic isotopic fractionation has previously been proposed to explain the light isotope enrichments in CAIs, which could occur either by condensation from a vapor phase pre-enriched in light isotopes by kinetic effects (Patchett, 1980a, 1980b; Huang et al., 2011) or during non-equilibrium condensation of an undercooled gas (e.g. Simon and DePaolo, 2010). However, Moynier et al. (2006) suggested that since elements (e.g. Sm and Eu) show distinct isotopic trends, kinetic isotope fractionation cannot account for the light isotope enrichments in CAIs. Electromagnetic sorting of isotopes in the nebular gas was proposed as an alternative mechanism to explain light isotope enrichments in CAIs based on Eu isotopes (Moynier et al., 2006). In contrast to Eu, which is mostly present in its monoatomic form in the nebular gas and thus maintains a low ionization potential, other REE (with the exception of Tm and Yb) such as Sm are mostly present in their oxide form in the solar nebula (Boynton, 1975), which increases their ionization potential. Electromagnetic isotope fractionation would preferentially affect elements with low ionization potentials ( $I_p$ ). This does in fact seem to be the case for the refractory elements for which CAIs have light isotope enrichments ( $I_p\text{Ca} = 6.1$  eV;  $I_p\text{Sr} = 5.69$  eV,  $I_p\text{Eu} = 5.67$  eV), while elements with higher ionization potentials ( $I_p\text{Fe} = 7.9$  eV,  $I_p\text{Mg} = 7.64$  eV,  $I_p\text{Si} = 8.1$  eV) have heavy isotope enrichments. This model can be further confirmed by measuring the isotopic composition of other refractory elements with low ionization potentials.

Barium is a refractory lithophile alkaline earth element with a 50% condensation temperature ( $T_c$ ) of 1455 K (Lodders, 2003), and it is among the elements with the lowest ionization potential ( $I_p = 5.2$  eV). It has seven stable isotopes:  $^{130}\text{Ba}$  (0.106%),  $^{132}\text{Ba}$  (0.101%),  $^{134}\text{Ba}$  (2.417%),  $^{135}\text{Ba}$  (6.592%),  $^{136}\text{Ba}$  (7.854%),  $^{137}\text{Ba}$  (11.23%), and  $^{138}\text{Ba}$  (71.70%). Because Ba is comprised of isotopes formed by a variety of nucleosynthetic processes, previous studies have focused on Ba isotopic anomalies (non-mass-dependent isotopic variations) wherein the data are internally normalized to correct for mass-dependent fractionation using Thermal-Ionization Mass Spectrometry (TIMS). This preferential interest in the nucleosynthetic anomalies of Ba coupled with the numerous Xe isobaric interferences that are unavoidable in Multi-Collector Inductively-Coupled-Plasma Mass-Spectrometry (MC-ICP-MS) have left the mass-dependent isotopic variations of Ba basically untouched. However, to study the mass-dependent isotopic composition of an element one needs only 3 stable isotopes;  $^{135}\text{Ba}$ ,  $^{137}\text{Ba}$ , and  $^{138}\text{Ba}$  are free of Xe interferences and therefore their ratios are potentially measurable at high precision by MC-ICP-MS.

Here we present the first high precision Ba stable isotopic compositions of a variety of meteoritic and terrestrial materials as well as separated individual CAIs in order to 1) study the building blocks of Earth

and 2) test the origin of the stable isotope variations in CAIs (kinetic condensation versus electromagnetic separation).

## 2. Samples and methods

To characterize the stable Ba isotopic composition of the bulk Earth, a selection of geological reference materials were selected for analysis (basalts BHVO-2 and BCR-2; andesites AGV-1 and AGV-2). The meteorite samples include two carbonaceous chondrites, Murchison (CM) and Allende (CV); three ordinary chondrites, Krymka (LL3.2), Kernouve (H6), and Grady (H3.7); one enstatite chondrite, Indarch (EH4); and one eucrite, Cachari.

We also analyzed three CAIs and one matrix-rich aliquot from Allende. Two fine-grained CAIs were extracted from a slab of the Allende meteorite using stainless steel tools at the Center for Meteorite Studies at ASU (CAI AB1 and CAI AB2), and the one other CAI was extracted by FM at the Institut de Physique du Globe de Paris (CAI FM1, 20 mg). CAI AB1 has a trace element pattern typical of the chemical group III and CAI AB2 gives a pattern of chemical group II (see Fig. 1 and Table 1). The REE pattern of group III CAIs (CAI AB1) is usually interpreted as evidence for direct condensation from the solar nebula and therefore of a primitive chemical composition, whereas group II CAIs (CAI AB2) have fractionated REE patterns relative to solar composition and have been explained by fractional condensation processes (e.g. MacPherson, 2003). We did not measure the REE pattern of the CAI FM1. The matrix fraction was separated from the chondrules under a microscope; great care was taken, but it may however contain a small fraction of chondrules that would have been crushed with the matrix. Therefore, our matrix sample represents a matrix-rich sample of Allende.

For bulk rock samples, the Allende matrix-rich sample and the CAI FM1, homogenous powder was fully dissolved in 4:1 HF/HNO<sub>3</sub>, evaporated to dryness, and then dissolved in 6 N HCl to completely remove fluorides. Meteorite samples and the CAI FM1 were dissolved utilizing Parr acid digestion vessels in an oven at 130 °C, while closed Teflon vials were used to dissolve terrestrial samples. Two ~50 mg fragments of fine-grained CAI (CAI AB1 and CAI AB2) were leached prior to dissolution to remove surface contamination: first in cold 1 M HF for 10 min, then in 2.5 M HCl for 10 min in an ultrasonic bath, and finally rinsed with Milli-Q H<sub>2</sub>O. After leaching, the residues (~40 mg) were dissolved using the Parr bomb procedure described in Bouvier et al. (2008). The fraction containing Ba and most of the sample matrix was collected in 2.5 M HCl during the first AG50W-X8 cation resin step following the protocol described in Bouvier et al. (2008) prior to further Ba purification as described below.

All the samples were loaded in 3 N HNO<sub>3</sub> on Teflon columns packed with 300 µL of Eichrom Sr specific resin. The logic of the chemical purification follows the procedure described in Horwitz et al. (1992). The column was first rinsed with 6 mL of 3 N HNO<sub>3</sub> and then the Ba was collected in 8 mL of 3 N HNO<sub>3</sub>. Samples were passed through the column procedure twice to ensure clean chemical separation of Ba. Full procedural blanks were below 0.1 ng Ba, which is insignificant relative to the quantity of Ba measured during analysis (>1 µg).

The third CAI (CAI FM1) was handpicked and ~20 mg was dissolved in a Parr bomb following the same procedure described for the bulk meteorite samples.

The samples were analyzed for their Ba isotopic compositions by MC-ICP-MS, using a Thermo Fisher Neptune Plus coupled with a desolvating nebulizer ESI Apex at the Institut de Physique du Globe de Paris, France. The Faraday cups were positioned to collect masses 134 (L3), 135 (L2), 136 (L1), 137 (C), 138 (H1), 139 (H2), and 140 (H3). One block of 30 ratios was measured for each sample with an integration time of 8.389 sec per scan. Due to large Xe interferences on the minor Ba isotopes, the inability to simultaneously measure both the intensity of an isobaric interference-free Xe isotope ( $^{131}\text{Xe}$ ) and the  $^{140}\text{Ce}$  intensity for Ce correction, and since three major Ba isotopes are free of

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