



Gallium isotopic evidence for the fate of moderately volatile elements in planetary bodies and refractory inclusions



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ABSTRACT

The abundance of moderately volatile elements, such as Zn and Ga, show variable depletion relative to CI between the Earth and primitive meteorite (chondrites) parent bodies. Furthermore, the first solar system solids, the calcium–aluminum-rich inclusions (CAIs), are surprisingly rich in volatile element considering that they formed under high temperatures. Here, we report the Ga elemental and isotopic composition of a wide variety of chondrites along with five individual CAIs to understand the origin of the volatile elements and to further characterize the enrichment of the volatile elements in high temperature condensates. The $\delta^{71}\text{Ga}$ (permil deviation of the $^{71}\text{Ga}/^{69}\text{Ga}$ ratio from the Ga IUPAC standard) of carbonaceous chondrites decreases in the order of CI > CM > CO > CV and is inversely correlated with the Al/Ga ratio. This implies that the Ga budget of the carbonaceous chondrites parent bodies were inherited from a two component mixing of a volatile rich reservoir enriched in heavy isotope of Ga and a volatile poor reservoir enriched in light isotope of Ga. Calcium–aluminum-rich inclusions are enriched in Ga and Zn compared to the bulk meteorite and are both highly isotopically fractionated with $\delta^{71}\text{Ga}$ down to -3.56% and $\delta^{66}\text{Zn}$ down to -0.74% . The large enrichment in the light isotopes of Ga and Zn in the CAIs implies that the moderately volatile elements were introduced in the CAIs during condensation in the solar nebula as opposed to secondary processing in the meteorite parent body and supports a change in gas composition in which CAIs were formed.

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1. Introduction

Volatile elements control many fundamental characteristics of the terrestrial planets, including their ability to develop and sustain life as well as the geochemical properties that make each planet unique. The asteroid belt is like a cold chamber of relics from the earliest stage of the solar system, and thanks to Jupiter's gravitational perturbation we have free sample delivery in the form of meteorites (Walsh et al., 2011). Among these meteorites, chondrites have not undergone planetary differentiation. A major unresolved issue is the widespread volatile elements depletion observed in terrestrial planets and in meteorites (e.g. Halliday and Porcelli, 2001; Palme and O'Neill, 2003). In particular, carbonaceous chondrites show a volatile depletion trend in the order CI > CM > CO > CV, which has challenged the scientific community for decades.

Calcium–aluminum rich Inclusions (CAIs), together with chondrules and matrix, are the main components of meteorites. Calcium–aluminum-rich inclusions are formed at high-temperature (e.g. Grossman et al., 2002) and represent the first solids of the Solar System with U-corrected Pb–Pb absolute age of 4567.30 ± 0.16 million years (Connelly et al., 2012). However, CAIs show large enrichment in moderately volatile elements (e.g. Zn, Ga, Refs: Chou et al., 1976; Bischoff et al., 1987; Sylvester et al., 1993) whose origin is debated between being introduced by secondary aqueous alteration of the CAIs on the meteorite parent body or by gas–solid exchange in the solar nebula prior to the formation of the parent body.

Volatilization is known to fractionate isotopes; therefore, comparing the isotopic compositions of volatile elements is a very powerful tool to understand the origin of volatile element abundance variations, especially to distinguish between aqueous alteration and volatilization for the origin of the volatile element excesses in CAIs (e.g. Luck et al., 2005). In this work, we will focus on the Ga isotopic composition of bulk meteorites and CAIs.

Gallium is a moderately volatile element with a 50% volatile temperature of 968 K (Lodders, 2003). It has two isotopes, ^{69}Ga and ^{71}Ga , with natural elemental abundances of 60.1% and

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39.9%, respectively (Machlan et al., 1986). Gallium is moderately siderophile as shown by its enrichment in iron meteorites (up to 100 ppm) compared to chondritic meteorites (5–10 ppm; Lodders and Fegley, 1998). In addition, its abundance varies by about two orders of magnitude between iron meteorite groups which depends on many factors but the most important is due to its volatility and Ga concentration has been widely used to classify iron meteorites (e.g. Wai and Wasson, 1979).

The Ga concentration of the bulk silicate Earth (BSE) falls on the terrestrial volatility trend, suggesting that despite its moderately siderophile behavior, large quantities of Ga was not segregated into the Earth's core (McDonough, 2003). Two possible scenarios can explain this apparent lithophilic behavior of Ga during Earth's formation: 1) A fraction of the Ga was partitioned into the Earth's core and later the mantle has been replenished in Ga by a late veneer together with other volatile elements (e.g. Albarède, 2009), and coincidentally Ga falls on the volatile trend defined by lithophile volatile elements, or 2) under the pressure and temperature condition of mantle/core segregation Ga behaved as a lithophile element (e.g. Blanchard et al., 2015). In addition, similar to other moderately volatile elements, Ga is progressively depleted from the CI (~10 ppm) to the CV (~6 ppm) chondrites groups (Lodders and Fegley, 1998) and the Earth's mantle is further depleted (~3.9 ppm; McDonough, 1990).

Early studies measured the isotopic composition in metallic Ga, as a representative of terrestrial Ga, as well as in some iron meteorites (De Laeter, 1972; Inghram and Hess, 1948), however at the analytical precision of these studies, no isotopic variations were found. With the advent of Multi-Collector Inductively-Coupled-Plasma Mass-Spectrometer (MC-ICP-MS), Ga isotopic compositions of terrestrial samples were re-investigated (Zhang et al., 2016; Yuan et al., 2016; Kato et al., 2017; Kato and Moynier, 2017). In particular, Kato et al. (2017) measured the Ga isotopic composition of a large variety of terrestrial igneous samples with a precision of 0.05‰ and showed that igneous processes does not fractionate Ga isotopes by more than the analytical precision and estimated the $\delta^{71}\text{Ga}$ (permil deviation of the $^{71}\text{Ga}/^{69}\text{Ga}$ ratio compared to the IPGP Ga standard) of the mantle to be $0.00 \pm 0.05\text{‰}$. In addition, Kato and Moynier (2017) have shown that the Moon is enriched in the heavier isotopes of Ga compare to Earth, suggesting volatile element loss by evaporation following the giant impact at the origin of the Moon.

Here, we present the Ga isotopic composition of a series of carbonaceous chondrites (CI, CM CO, CV and CBa), along with enstatite (EH and EL) and ordinary chondrites (L, LL, H) in order to understand 1) the volatile element history of the chondrites parent body, and 2) the origin of the apparent excess of Ga in the Earth's mantle. We also measured the isotopes of the moderately volatile elements Ga and Zn in CAIs from Allende CV3 meteorite to test the possible origin of moderately volatile element excesses. By investigating the magnitude of isotopic fractionation it is possible to determine its source, as aqueous alteration processes produce limited isotopic fractionation of Zn (on the order of 0.2‰ for CM chondrites that are highly aqueous altered chondrites; see Luck et al., 2005), Ga isotopes are unlikely to be affected by aqueous alteration as well; however, if Ga was incorporated into the CAIs within the solar nebula, evaporation/condensation processes should produce large isotopic fractionation (e.g. Davis et al., 1990).

2. Samples and methods

2.1. Samples

The analyzed chondrite groups were, in order of volatile element enrichment, CI (Y980115), CM (Cold Bokkeveld, Murchison,

LON 94101, Jbilet Winselwan), CO (Lancé, Ornans, Felix), CV (Allende, Vigarano) and CBa (Gujba). The ordinary chondrites studied were QUE 97030 (H3.4), LAR 06392 (H5), ALHA81030 (L3.4), ALHA76001 (L6), EET 83213 (LL3.7), LAP 02266 and LAP 02266 (LL4) are all meteorites collected in Antarctica (find). Eight enstatite chondrites were studied: Kota-Kota, Qingzhen, Sahara 97072 (all EH3) Indarch and Abee (EH4), St Mark's (EH5), LON 94100 and Hvittis (EL6). Calcium–aluminum-rich inclusions were all from Allende, C1, C2 and C3 are fine-grained CAIs, whereas C4, and C18 are coarse-grained CAIs and are all described in details in Sossi et al. (2017). Reproducibility of the entire methodology (sample dissolution, chemical purification, mass-spectrometry) was investigated through total procedural replicates, and to reaffirm that no isotopic fractionation is induced by the chemical purification, samples were passed through the chemistry multiple extra times following the same approach as in Kato et al. (2017).

2.2. Sample preparation and chemical purification

All bulk meteorites were powdered with an agate mortar and pestle, and the weight of each sample processed through chemistry was estimated in order to obtain approximately 1 µg of Ga (5 mg to 100 mg of bulk sample depending on the Ga concentration). The largest CAIs were selected and sampled from their host meteorite by crushing and collected by tweezers under a microscope and powdered for chemical purification. The glassy matrix was sampled by tweezers upon crushing from a section free of visible inclusions. All samples were dissolved in Teflon beakers on hot plates in a mixed solution of HNO₃ and HF (3:1) at 120 °C for 72 h. After dissolution, samples were passed through a three-step chemical purification: 1) first step to separate Ga and Fe from major elements, 2) second step to separate Ga from Fe and 3) third step to remove the isobaric interference element Ba from Ga. The doubly ionized ^{138}Ba has the same mass over charge ratio as $^{69}\text{Ga}^+$, which would result as an isobaric interference. The achieved yield was above 95% and the final analytical blank (<0.02 ng) was negligible, as shown in details along with the Ga purification method described in Kato et al. (2017). The Zn isotopic composition of the CAIs were measured on the Zn obtained from the Ga separation on which we applied the method detailed in Kato et al. (2015).

2.3. Isotopic measurement

Gallium and Zn concentrations and isotope ratio were analyzed on a Thermo Fisher Scientific Neptune Plus multi-collector inductively-coupled-plasma mass-spectrometer (MC-ICP-MS) at either Washington University in St. Louis, USA or at the Institut de Physique du Globe de Paris (IPGP), France (see Kato et al., 2017 for details on the Ga isotopic measurements and Moynier and Le Borgne, 2015 for Zn isotopic measurements). Samples were dissolved and diluted in a 0.1 N HNO₃ solution to match the concentration of the standard, and were introduced into the plasma source through an ESI APEX HF sample introduction system for Ga, a double-pass glass cyclonic spray chamber introduction system was used for Zn. A blank solution of 0.1 N HNO₃ was measured in the beginning of each sequence and a blank correction was applied. A jet sampler cone and a H skimmer cone was used. Peak centers were taken after tuning and baselines were measured before each measurement. Each analysis was composed of 25 cycles of 8.3 seconds, and was done in low-resolution mode. Gallium and Zn isotopic variations are reported as per mil deviations from the Ga IPGP standard (Aldrich, catalog number 35623-9, Lot no. 00226LX) for Ga and JMC-Lyon standard for Zn. The Ga IPGP standard is used instead of the international standards NIST SRM 3119a which is ~1.4‰ fractionated compared to the bulk silicate

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