



# Oxygen isotopic ratios of primordial water in carbonaceous chondrites



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## ARTICLE INFO

### Article history:

Received 16 June 2017

Received in revised form 23 September 2017

Accepted 23 October 2017

Available online xxxx

Editor: F. Moynier

### Keywords:

carbonaceous chondrites

primordial water

oxygen isotopes

hydrogen abundances

water/rock ratios

## ABSTRACT

In this work, I estimate the  $\delta^{18}\text{O}$  and  $\delta^{17}\text{O}$  values of primordial water in CM chondrites to be  $55 \pm 13$  and  $35 \pm 9\%$ , respectively, based on whole-rock O and H data. Also, I found that the O and/or H data of Antarctic meteorites are biased, which is attributed to terrestrial weathering. This characteristic O isotopic ratio of water together with corresponding water abundances in CM chondrites are consistent with the origin of water as ice processed by photochemical reactions at the outer regions of the solar nebula, where mass-independent O isotopic fractionation and water destruction may have occurred. Another possible mechanism to produce the inferred O isotopic ratio of water would be O isotopic fractionation between water vapor and ice, which likely occurred near the condensation front of  $\text{H}_2\text{O}$  (snow line) in the solar nebula. The inferred O isotopic ratio of water suggests that carbonate in CM chondrites formed at low temperatures of  $<150^\circ\text{C}$ . The O isotopic ratios of primordial water in chondrites other than CM chondrites are not well constrained.

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## 1. Introduction

Chondrites are aqueously altered on their parent asteroids, which is pronounced in CI, CM, and CR chondrites (e.g., Brearley, 2006; Krot et al., 2015). Oxygen and H isotopic ratios of water are clues for understanding its origins and the processes it has undergone. Variations in the H isotopic ratios (D/H) of planets, meteorites, and comets have been widely studied to explore genetic links (e.g., Robert, 2006; Alexander et al., 2012; Sarafian et al., 2014). Because H has only two isotopes, it is not possible to evaluate mixing of more than three H reservoirs based on the D/H ratios of samples. In contrast, O has three isotopes, which make its isotopic ratios more useful to clarify the origins of O reservoirs in the solar system and the processes these reservoirs underwent.

The  $^{18}\text{O}/^{16}\text{O}$  and  $^{17}\text{O}/^{16}\text{O}$  ratios of extraterrestrial materials vary not only under a mass-dependent law but also under a mass-independent law (e.g., Clayton, 1993). Consequently, observed O isotopic variation cannot be attributed to a single O reservoir affected by physicochemical processes associated with mass-dependent isotopic fractionation, but requires mixing of multiple O reservoirs with different isotopic ratios resulting from mass-independent fractionation.  $\text{H}_2\text{O}$  has been considered an O reservoir with an  $^{17,18}\text{O}$ -rich composition, and thus a key component in shaping the O isotopic ratios of solar system materials (Clayton and Mayeda, 1984; Choi et al., 1998; Yurimoto and Kuramoto, 2004;

Lyons and Young, 2005; Sakamoto et al., 2007). Therefore, understanding the O isotopic ratios of  $\text{H}_2\text{O}$  in chondrites is important, yet difficult, because water–rock interactions in chondrite parent asteroids modify the O isotopic ratios of water via O-isotope exchange with the rock. Thus, initial isotopic ratios of water in chondrites have been poorly understood.

Clayton and Mayeda (1999) explained the bulk O isotopic ratios of CM chondrites by a closed-system alteration model. In their model, however, the initial O isotopic ratio of water and water/rock ratios of CM chondrites cannot be determined individually. Therefore, they estimated the initial O isotopic ratio of water assuming alteration temperatures and complex two-stage interactions of O reservoirs, i.e., O-isotope exchange between solids and gas in the solar nebula, and between anhydrous silicate and water in the CM chondrite parent asteroid (Clayton and Mayeda, 1984). Their estimated values of  $\delta^{18}\text{O}$ ,  $\delta^{17}\text{O}$ , and  $\Delta^{17}\text{O}$  for the CM primordial water are  $\sim 30.3$ ,  $\sim 20.2$ , and  $\sim 4.4\%$ , respectively ( $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$ , i.e., the deviation from mass-dependent isotopic fractionation).

In this paper, I present the initial O isotopic ratio of water in CM chondrites based on whole-rock O and H data. From the inferred O isotopic ratio of the CM primordial water and water/rock ratios of CM chondrites, the origin of the CM primordial water and where the parent asteroid of CM chondrites accreted are discussed. It is shown that the inferred O isotopic ratio of the CM primordial water is consistent with the O isotopic ratios of carbonate in CM chondrites. The O isotopic ratios of primordial water in chondrites

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other than CM chondrites are also discussed, although they are not well constrained.

## 2. Material and methods

### 2.1. Data sources

Most O and H data used in this study are obtained by Clayton and Mayeda (1999) and Alexander et al. (2013), respectively. Exceptions are data for Paris, Sayama, and Sutter's Mill. The mean  $\delta^{18}\text{O}$  and  $\delta^{17}\text{O}$  values of anhydrous minerals in Murchison are  $-4.2\text{‰}$  and  $-7.4\text{‰}$ , respectively (Clayton and Mayeda, 1984), which are used in this work as the representative values of anhydrous silicate in CM chondrites. Clayton and Mayeda (1999) also reported the  $\delta^{18}\text{O}$  and  $\delta^{17}\text{O}$  values of matrix separates from CM chondrites, which range from 11.20 to 12.70 and 3.52 to 4.72‰ (except for LEW 85311 and Essebi), respectively. The average  $\delta^{18}\text{O}$  and  $\delta^{17}\text{O}$  values of the matrix separates are 11.73 and 4.04‰, respectively, which are used as the representative values of hydrous minerals in CM chondrites. Browning et al. (1996) reported the chemical compositions of serpentine in CM chondrites. Based on these data and a generalized stoichiometric phyllosilicate formula, the H abundances of the serpentine can be calculated.

### 2.2. Specific meteorites with notes

#### 2.2.1. Paris

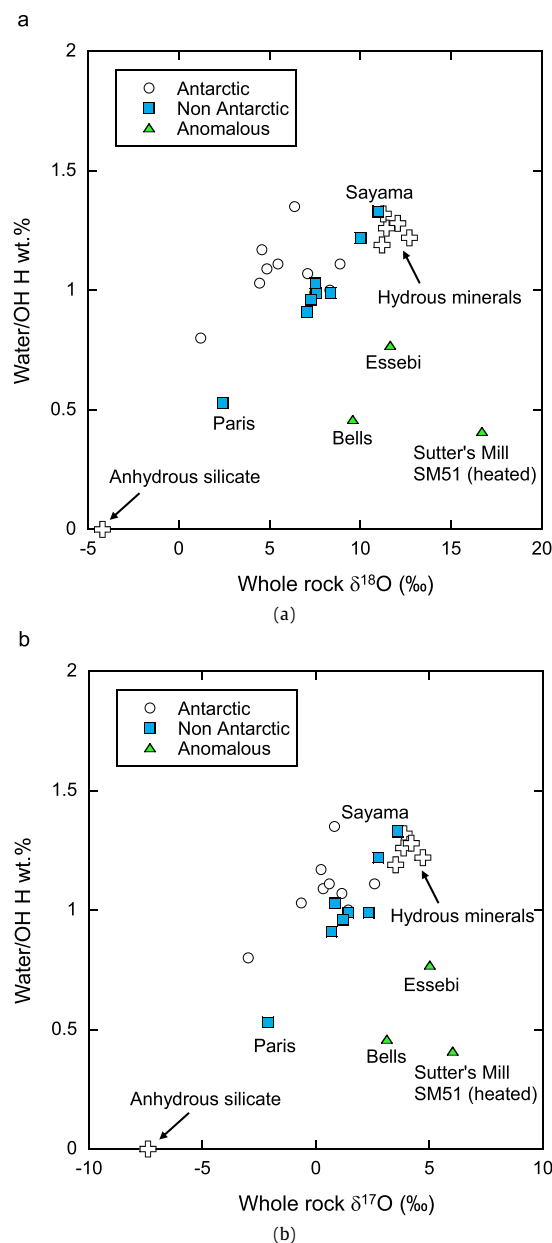
Paris is one of the least aqueously altered CM chondrites. Detailed mineralogical and petrological description of this meteorite is given by Hewins et al. (2014). The water content of Paris is 4.78 wt.%, which corresponds to 0.53 wt.% H (Vacher et al., 2016). Eleven analyses of bulk O isotopes were carried out (Hewins et al., 2014). It should be stressed that Paris shows heterogeneous degrees of alteration among domains. Corresponding to the degree of alteration, O isotopic ratios from individual analyses vary significantly, with lower  $\delta^{18}\text{O}$  values and greater heterogeneity for less altered samples. It is unclear whether samples analyzed by Vacher et al. (2016) and Hewins et al. (2014) were extracted from the same or different aliquots. Here, the lowest  $\delta^{18}\text{O}$  value (2.43‰) obtained by Hewins et al. (2014) is adopted for the sample analyzed for H by Vacher et al. (2016) because of its high  $\delta\text{D}$  value (72‰) and low water content, which indicate a low degree of alteration. These O and H data, together with those of the other CM Falls, yield the best correlation between  $\delta^{17,18}\text{O}$  values and H abundances (Fig. 1).

#### 2.2.2. Nogoya

Two different H measurements have been presented by Alexander et al. (2012, 2013), one of which was obtained from a sample crushed under acetone during sample preparation. Although acetone contamination may have affected the H abundance, sample heterogeneity is another possible explanation for the inconsistency between the two values. The H content of water/OH in the dry crushed sample appears unreasonably high (1.31 wt.%), close to the H abundance of serpentine in Nogoya (1.32 wt.%). Furthermore, this water/OH H content is the third highest value measured for CM chondrites by Alexander et al. (2012, 2013). These observations indicate that the dry crushed sample was heavily altered and dominated by hydrous minerals without any anhydrous silicate. Therefore, the dry crushed sample may not represent Nogoya, and the water/OH H content of the sample crushed under acetone is adopted in the present study (0.99 wt.%).

#### 2.2.3. Sayama

Sayama is a heavily altered CM chondrite. Its mineralogy, petrology, and geochemical features were reported at a conference



**Fig. 1.** The  $\delta^{18}\text{O}$  values (a) and  $\delta^{17}\text{O}$  values (b) of bulk CM chondrites against the water/OH H contents in the same samples (Clayton and Mayeda, 1999; Alexander et al., 2013). Meteorites to which their names are attached are described in section 2.2. The values of anhydrous silicate are represented by those of Murchison. The O isotopic ratios of bulk CM chondrites are correlated with the water/OH H contents, which indicates that the O isotopic ratios of the bulk CM chondrites reflect variable amounts of anhydrous silicate and hydrous minerals, and that hydrous minerals in CM chondrites must have a common O isotopic ratio.

(Yoneda et al., 2001). Four measurements of bulk O isotopic compositions yielded  $\delta^{18}\text{O}$  values from 9.9 to 12.0‰. A simple average of these four measurements (i.e.,  $\delta^{18}\text{O} = 11.0\text{‰}$ ) is adopted in this work.

The water content of Sayama is 12 wt.%, which corresponds to 1.33 wt.% H in hydrous minerals (Yoneda et al., 2001). This H abundance is close to the value of 1.26 wt.% estimated as Alexander et al. (2013) did by subtracting H of organic matter from the bulk H content of 1.37 wt.%. The H abundance of organic matter is calculated under the assumption that the bulk C in Sayama (1.99 wt.%) is in organic matter with a H/C ratio 0.055 (by weight) derived from insoluble organic matter (IOM) in typical CM chondrites.

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