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## Primordial water and dust of the Solar System: Insights from in situ oxygen measurements of CI chondrites

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1 **Primordial water and dust of the Solar System:**  
2 **Insights from *in-situ* oxygen measurements of CI chondrites**

3  
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14  
15 **Abstract**

16  
17 As the chemical compositions of CI chondrites closely resemble that of the Sun's photosphere,  
18 their oxygen isotopic compositions represent a powerful tool to constrain the origin and  
19 dynamics of dust and water ice grains in the protoplanetary disk. However, parent-body  
20 alteration processes make straightforward estimation of the primordial isotopic compositions  
21 of CI chondritic water and anhydrous minerals difficult. In this contribution, we used *in-situ*  
22 SIMS measurements to determine the oxygen isotope compositions of mechanically isolated  
23 olivine and carbonate grains from the CI chondrite Orgueil and carbonates in a polished  
24 section of the CI chondrite Ivuna. Most CI olivine grains have Earth-like O isotopic  
25 compositions ( $\Delta^{17}\text{O} \approx 0\%$ ) plotting at the intersection of the terrestrial fractionation line and  
26 the primitive chondrule minerals line. Ca-carbonates from Orgueil and Ivuna define a trend  
27 with  $\delta^{17}\text{O} = (0.50 \pm 0.05) \times \delta^{18}\text{O} + (0.9 \pm 1.4)$  that differs from mass-independent variations  
28 observed in secondary phases of other carbonaceous chondrites. These data show that CIs are

29 chemically solar but isotopically terrestrial for oxygen isotopes. This supports models  
30 suggesting that primordial Solar System dust was  $^{16}\text{O}$ -poor ( $\Delta^{17}\text{O} \approx 0\text{‰}$ ) relative to the  $^{16}\text{O}$ -  
31 rich nebular gas. Based on results, mass balance calculations reveal that the pristine O  
32 isotopic compositions of carbonaceous chondrite matrices differ significantly from the CI  
33 composition, except for CR chondrites (calculated  $\Delta^{17}\text{O}$  values of CM, CO, CV and CR  
34 matrices being  $-3.97 \pm 1.19\text{‰}$ ,  $-4.33 \pm 1.45\text{‰}$ ,  $-7.95 \pm 1.95\text{‰}$ , and  $-0.07 \pm 1.16\text{‰}$ ,  
35 respectively). This confirms an open chondrule-matrix system with respect to oxygen isotopes  
36 where chondrule compositions reflect complex processes of chondrule precursor recycling  
37 and gas-melt interactions. As the Mg-Si-Fe chondrule budget is also partially controlled by  
38 gas-melt interactions, the complementary formation of chondrules and matrix from a single  
39 solar-like reservoir –if it exists– require that (i) this reservoir must have been in a closed  
40 system with the gas or (ii) the gas had a CI composition to satisfy the elemental mass balance.

41

42 **Keywords:** CI chondrites, water, primordial dust, oxygen isotopes, chondrule-matrix  
43 complementarity

44

## 45 **1. Introduction**

46

47 CI carbonaceous chondrites contain the highest abundances of hydrogen (~1.5 wt.%;  
48 Alexander et al., 2012) of all chondrite classes and groups, mostly in their hydrated minerals.  
49 These minerals are considered to result from intense episodes of hydrothermal alteration  
50 induced by the melting of accreted water ice grains, which strongly modified the initial CI  
51 petrography (Brearley, 2006). CI chondrites indeed contain high abundances of secondary  
52 minerals such as phyllosilicates (81–84 vol.%), magnetites (6–10 vol.%), sulfides (4–7 vol.%),  
53 and carbonates (<3 vol.%; King et al., 2015). Although CI chondrites are the most altered  
54 chondrites known so far, they are of primary importance as their bulk chemical compositions  
55 are similar to that of the solar photosphere, which is representative of the average Solar  
56 System composition (Lodders, 2003).

57 Chondrites generally comprise four main components in variable proportions:  
58 chondrules (20–80 vol.%), refractory inclusions (0.1–3 vol.%), Fe-Ni metal beads (0.1–70  
59 vol.%), and matrix material (1–75 vol.%). These different components formed during the  
60 evolution of the solar protoplanetary disk and thus represent temporal and spatial records of  
61 the formation of the Solar System. The chondrule-matrix complementarity model proposes  
62 that chondrules and matrices in carbonaceous chondrites are genetically linked based on their  
63 Mg/Si ratios (Hezel and Palme, 2010; Palme et al., 2015; Hezel et al., 2018): although bulk  
64 carbonaceous chondrites exhibit CI chondritic Mg/Si ratios, chondrules and matrices have  
65 super- and sub-CI chondritic ratios, respectively, suggesting that chondrules and matrices  
66 formed from a unique chemical reservoir of bulk CI composition. Although this model is  
67 widely debated (see Hezel et al., 2018, and Zanda et al., 2018, for recent reviews), it has  
68 fundamental implications for the dynamics of the protoplanetary disk and highlights the  
69 importance of deciphering the nature of the primitive CI matrix in terms of understanding

70 (i) the conditions of chondrule formation (Hezel et al., 2018; van Kooten et al., 2019) and  
71 (ii) the transport of dust in the protoplanetary disk (Anders, 1964; Zanda et al., 2018).  
72 Unfortunately, CI chondrites are rare and only a few have been described: Alais (~6 kg),  
73 Ivuna (705 g), Orgueil (~14 kg), Revelstocke (1 g), and Tonk (7.7 g). In addition, the intense  
74 parent-body fluid circulation processes that altered CI chondrites limit our knowledge of their  
75 original characteristics. Thus, it remains unclear if CI chondrites were entirely composed of  
76 fine-grained matrix or if they initially contained significant proportions of refractory  
77 inclusions, chondrules, and Fe-Ni metal beads that were subsequently erased by secondary  
78 alteration processes. To our knowledge, only one partially altered, <sup>26</sup>Al-free, <sup>16</sup>O-rich Ca-Al-  
79 rich refractory inclusion has been described in Ivuna (Frank et al., 2011, 2017), and no  
80 chondrules or pseudomorphosed chondrules have yet been reported in CI chondrites.

81         Although extensively altered, high-temperature minerals such as olivine and pyroxene  
82 are present in significant proportions in CI chondrites (~3 vol.%; King et al., 2015) and can be  
83 isolated by mechanical processing (Reid et al., 1970; Kerridge and MacDougall, 1976; Leshin  
84 et al., 1997) and identified in thin sections. These grains are generally anhedral and have been  
85 reported to contain Fe-Ni metal blebs and glass inclusions with shrinking bubbles (Reid et al.,  
86 1970; Kerridge and MacDougall, 1976; Leshin et al., 1997; Le Gac et al., 2009). Most  
87 olivines extracted from CI chondrites and measured by secondary ion mass spectrometry  
88 (SIMS; Leshin et al., 1997) have O isotopic compositions plotting at the intersection of the  
89 Primitive Chondrule Minerals line (PCM,  $\delta^{17}\text{O} = 0.987 \times \delta^{18}\text{O} - 2.7$ ; Ushikubo et al., 2012)  
90 and the Terrestrial Fractionation Line (TFL,  $\delta^{17}\text{O} = 0.52 \times \delta^{18}\text{O}$ ). Few olivine grains exhibit  
91 more <sup>16</sup>O-rich compositions (Leshin et al., 1997) similar to chondrule olivines commonly  
92 measured in CM, CO and CV carbonaceous chondrite (i.e.,  $\delta^{18}\text{O} \approx \delta^{17}\text{O} \approx -7\text{‰}$ ; Clayton and  
93 Mayeda, 1999; Marrocchi and Chaussidon, 2015; Marrocchi, et al., 2018a, 2019; Tenner et al.,  
94 2018), suggesting that chondrules might represent a significant component accreted by CI

95 chondrites. However, the large uncertainties on these measurements (i.e.,  $\sim 4.5\%$ ,  $2\sigma$ , inherent  
96 in SIMS measurements at that time) preclude definite interpretation regarding the signature of  
97 high-temperature minerals in CI chondrites. In addition, it is not clear if CI chondrite-  
98 accretion region(s) experienced chondrule formation processes or if chondrules were radially  
99 transported from other chondrule-forming regions.

100         Among the secondary phases in CI chondrites, carbonates are of primary importance  
101 as they represent a direct snapshot of the alteration fluid from which they precipitated  
102 (Clayton and Mayeda, 1984). However, only a few measurements have been reported so far  
103 for carbonates in CI chondrites, and only the  $\delta^{18}\text{O}$  values were reported (Grady et al., 1988;  
104 Alexander et al., 2015). In addition, these data correspond to analyses after  $\text{H}_3\text{PO}_4$  acid  
105 treatment (Clayton and Mayeda, 1984; Leshin et al., 2001), and potential contamination from  
106 adjacent matrix cannot be excluded. To date, no SIMS O isotopic analyses have been reported  
107 for CI carbonates, despite the aptitude of this technique for deciphering the origin of water  
108 accreted by chondrites and their alteration conditions (Tyra et al., 2012, 2016; Lee et al.,  
109 2013; Fujiya et al., 2015; Vacher et al., 2016, 2017, 2019; Verdier-Paoletti et al., 2017, 2019;  
110 Jilly-Rehak et al., 2017; Marrocchi et al., 2018b).

111         Here, we report results from a protocol in which we have (i) mechanically isolated  
112 mafic minerals and carbonate grains from the CI chondrite Orgueil, and  
113 (ii) determined their oxygen isotopic composition by SIMS. For comparison, we surveyed a  
114 polished section of Ivuna and analysed the O isotopic compositions of carbonates. We use our  
115 results to investigate the pre-alteration oxygen isotopic composition of CI chondrites and to  
116 constrain the source of water accreted by CI chondrites. In addition, we perform mass-balance  
117 calculations to (i) estimate the pristine oxygen isotopic compositions of carbonaceous  
118 chondrite matrices and (ii) bring constraints on the controversial concept of chondrule-matrix  
119 complementary.

## 120 **2. Material and methods**

121

### 122 **2.1 Grain separation and preparation**

123

124 Mineral grains were separated from ~15 mg of Orgueil following the method  
125 described in Leshin et al. (1997), and briefly summarized here. Chips of Orgueil were gently  
126 crushed in an agate mortar and the powder was ultrasonically cleaned in ethanol for 45 min.  
127 After drying, the powder was sieved into <50  $\mu\text{m}$ , 50–100  $\mu\text{m}$ , and >100  $\mu\text{m}$  size fractions.  
128 Coarse olivine and carbonate grains were then handpicked, mounted in epoxy, and polished  
129 using diamond paste (Fig. 1).

130

### 131 **2.2 Petrographic and mineralogical characterisation**

132

133 We acquired elemental distribution maps of Ivuna at the Natural History Museum in  
134 London using a Zeiss EVO 15LS scanning electron microscope (SEM) equipped with an  
135 Oxford Instruments XMax silicon drift energy dispersive X-ray (EDX) detector and INCA  
136 version 18 software (Fig. 2). Backscattered electron images and EDX maps were acquired at  
137 an accelerating voltage of 20 kV and 3 nA beam current. Due to the size of the samples (each  
138 approximately 3.5  $\text{cm}^2$ ), coverage at high resolution required the overnight collection of over  
139 200 separate maps per section using the Smartmap automated-stage raster procedure. For each  
140 X-ray map area, multiple frames of 1024  $\times$  768 analysis points were collected at a pixel-  
141 spacing of 1.52  $\mu\text{m}$ . X-ray emission spectra (0–10 keV in 2048 channels) were stored for each  
142 pixel location. Elemental X-ray maps were subsequently extracted from the data set  
143 (approximately 1.2 GB), and combined in a conventional Mg = red, Fe = blue, Ca = green  
144 colour scheme to reveal areas of specific interest (Fig. 2).

145 Minerals isolated from Orgueil were coated with a 25-nm-thick carbon layer to avoid  
146 charging effects under the electron beam. SEM imaging was performed on a JEOL JSM-6510  
147 SEM equipped with a Genesis EDX detector at the Centre de Recherches Pétrographiques et  
148 Géochimiques (CRPG-CNRS, Nancy, France) using 3 nA electron beam accelerated at 15 kV.

149 Quantitative chemical compositions of carbonate and olivine grains were obtained  
150 using a Cameca SX Five electron microprobe at the Université Pierre et Marie Curie (UPMC,  
151 Camparis, Paris, France) using a 40 nA focused beam accelerated to 15 kV. We analysed Na,  
152 Mg, Si, Al, K, Ca, Fe, Ti, Cr, and Mn in Orgueil olivines, Mg, Si, Ca, Fe, and Mn in Orgueil  
153 carbonates, and Na, Mg, Si, Al, K, Ca, Fe, Cr, and Mn in Ivuna carbonates. The high beam  
154 current allowed detection limits for silicates to be 150 ppm for Al, Ca, and Ti, 200 ppm for  
155 Mn and Si, and 400 ppm for Na, K, Cr, Fe, and Mg. The PAP software was used for matrix  
156 corrections. We estimate the detection limits for carbonates to be 900 ppm for Ca, Fe, Mn, Na,  
157 and K, and 600 ppm for Mg, Al, Si, and Cr.

158

### 159 **2.3 Oxygen-isotope analysis**

160

161 We measured the oxygen isotopic compositions of CI minerals via SIMS using the  
162 CAMECA IMS 1270 E7 at CRPG-CNRS.  $^{16}\text{O}^-$ ,  $^{17}\text{O}^-$ , and  $^{18}\text{O}^-$  ions produced by a  $\text{Cs}^+$   
163 primary ion beam ( $\sim 15\ \mu\text{m}$ ,  $\sim 4\ \text{nA}$ ) were measured in multi-collection mode using three  
164 Faraday cups. A normal-incidence electron gun was used for charge compensation.

165 To remove interference from  $^{16}\text{OH}^-$  on the  $^{17}\text{O}^-$  peak and achieve maximum flatness  
166 atop the  $^{16}\text{O}^-$  and  $^{18}\text{O}^-$  peaks, the entrance and exit slits of the central Faraday cup were  
167 adjusted to reach a mass resolving power ( $\text{MRP} = M/\Delta M$ ) of  $\sim 7000$  for  $^{17}\text{O}^-$ .  $^{16}\text{O}^-$  and  $^{18}\text{O}^-$   
168 were measured on L'2 and H1 (slit 1,  $\text{MRP} \approx 2500$ ). As an additional safeguard against  $^{16}\text{OH}^-$   
169 interference, a  $\text{N}_2$  trap was used to reduce the pressure in the analysis chamber to  $< 5 \times 10^{-9}$

170 mbar. The total measurement duration was 240 s (180 s of measurement + 60 s of pre-  
171 sputtering). We used five terrestrial standard materials (San Carlos olivine, magnetite,  
172 diopside, calcite, and dolomite) to define the instrumental mass fractionation (IMF) line for  
173 the three oxygen isotopes and correct for IMF due to matrix effects in olivine and carbonate  
174 grains. Typical count rates obtained on standards were  $2.8 \times 10^9$  cps for  $^{16}\text{O}$ ,  $1.1 \times 10^6$  cps for  
175  $^{17}\text{O}$ , and  $6.3 \times 10^6$  cps for  $^{18}\text{O}$  in carbonates, and  $2.7 \times 10^9$  cps for  $^{16}\text{O}$ ,  $1.0 \times 10^6$  cps for  $^{17}\text{O}$ ,  
176 and  $5.5 \times 10^6$  cps for  $^{18}\text{O}$  in San Carlos olivine. Typical  $2\sigma$  measurement errors, accounting  
177 for internal errors on each measurement and the external reproducibility of the standard, were  
178 estimated to be  $\sim 0.2\text{‰}$  for  $\delta^{18}\text{O}$ ,  $\sim 0.3\text{‰}$  for  $\delta^{17}\text{O}$ , and  $\sim 0.9\text{‰}$  for  $\Delta^{17}\text{O}$  (representing deviation  
179 from the TFL:  $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$ ).

180

### 181 3. Results

182

183 We found 5 olivine, 6 calcite, 14 dolomite, and 8 ferromagnesite grains in Orgueil  
184 after crushing, sieving, and handpicking (representative olivine grains are shown in Fig. 1).  
185 Additionally, we selected 16 dolomite grains in the thick section of Ivuna for chemical and  
186 isotopic characterization. Hereafter, grains are referred to using the identifiers “O” for Orgueil  
187 and “I” for Ivuna.

188 Orgueil olivines were Mg-rich ( $\text{Fo}_{88-95}$ ) and had low abundances of minor elements  
189 such as Ca ( $\leq 0.2$  wt.% CaO, Table 1). Four of the five olivine grains had similar O isotopic  
190 compositions plotting at the intersection of the TFL and PCM, with average values of  $\delta^{18}\text{O} =$   
191  $5.69 \pm 0.42\text{‰}$  and  $\delta^{17}\text{O} = 2.96 \pm 0.23\text{‰}$  (Fig. 3A, Table 2). Only olivine grain O16-8 had  
192 markedly different O isotopic values ( $\delta^{18}\text{O} = 1.14 \pm 0.18\text{‰}$  and  $\delta^{17}\text{O} = -1.41 \pm 0.38\text{‰}$ , Fig.  
193 3A, Table 3) and plotted on the PCM below the TFL; this olivine had the highest Fo content  
194 and was enriched in Al, Ca, and Cr compared to the other olivine grains (Table 1).

195 We observed Ca-rich, Ca-Mg-rich, and Mg-Fe-Mn-rich carbonate groups in Orgueil,  
196 which represent calcites, dolomites, and ferromagnesites, respectively (Table 3). Based on  
197 microprobe analyses, we identified 6 calcites, 14 dolomites, and 8 ferromagnesites. The  
198 ferromagnesites include two breunnerites ( $[\text{Mg}_x\text{Fe}_{(1-x)}]\text{CO}_3$  with  $0.7 < x < 0.9$ ; grains O6-2  
199 and O7-13), five mesitites ( $[\text{Mg}_x\text{Fe}_{(1-x)}]\text{CO}_3$  with  $0.5 < x < 0.7$ ; grains O6-7, O6-10, O7-8,  
200 O7-18, and O17-6), and one magnesite ( $\text{MgCO}_3$ , grain O21-6). Calcites in Orgueil had  
201 constant O isotopic compositions averaging  $\delta^{18}\text{O} = 25.55 \pm 0.08\text{‰}$  and  $\delta^{17}\text{O} = 13.29 \pm 0.36\text{‰}$   
202 (Fig. 3A, Table 3). Dolomite grains had varying O isotopic compositions, with  $\delta^{18}\text{O}$  values  
203 ranging from 24.16 to 32.49‰ and  $\delta^{17}\text{O}$  values from 12.81 to 17.22‰, but constant  $\Delta^{17}\text{O}$   
204 values averaging  $0.02 \pm 0.80\text{‰}$  (Fig. 3A, Table 3). In Ivuna, we observed only dolomites (Fig.  
205 2) of similar isotopic compositions to Orgueil dolomites, with  $\delta^{18}\text{O}$  values ranging from 24.57  
206 to 29.66‰,  $\delta^{17}\text{O}$  values from 13.06 to 15.68‰, and constant  $\Delta^{17}\text{O}$  values averaging  $0.40 \pm$   
207  $0.80\text{‰}$  (Fig. 3A, Table 3).

208

## 209 **4. Discussion**

210

### 211 **4.1 Nature and origin of CI olivine grains**

212

213 All but one olivine grain isolated from Orgueil have O isotopic compositions plotting  
214 at the intersection of the PCM and TFL ( $\Delta^{17}\text{O} = 0\text{‰}$ , Fig. 3), which corresponds to the typical  
215 oxygen isotopic composition of olivine grains from the Earth's mantle (Eiler, 2001). In  
216 addition, these olivine grains have lower Fo contents ( $\text{Fo}_{88-95}$ ) than those commonly reported  
217 in (i) CI chondrites (i.e.,  $\text{Fo} > 95$ ; Fig. 4; Reid et al., 1970; Kerridge and MacDougall, 1976;  
218 Steele, 1990; Leshin et al., 1997; Le Gac et al., 2009; Frank et al., 2014) and (ii) type I  
219 chondrules in different types of carbonaceous chondrites (i.e.,  $\text{Fo} > 94$ ; Frank, et al., 2014;

220 Tenner et al., 2018). This raises the question of whether these oxygen isotopic compositions  
221 reflect terrestrial contamination. However, the olivine grains were recovered after crushing a  
222 solid piece of Orgueil, and not from a powder that could have had been contaminated by  
223 terrestrial minerals. In addition, CI olivine grains with Fo < 95 are not uncommon as olivine  
224 grains with Fo as high as 55 have been reported in previous studies (Fig. 4; Reid et al., 1970;  
225 Kerridge and MacDougall, 1976; Steele, 1990; Leshin et al., 1997; Le Gac et al., 2009; Frank  
226 et al., 2014). Furthermore, among the recovered olivine grains, one plots on the PCM but  
227 below the TFL (Fig. 2), suggesting an extraterrestrial origin. Finally, previous studies of  
228 mechanically isolated mafic minerals from Orgueil reported similar olivine and pyroxene O  
229 isotopic compositions (Leshin et al., 1997). These observations suggest that the mafic  
230 minerals isolated from Orgueil indeed correspond to primary extraterrestrial minerals, in good  
231 agreement with their relative abundances in Orgueil as inferred from X-ray diffraction  
232 patterns (~3%; King et al., 2015).

233 CI chondrites are the only subgroup of chondrites that do not contain chondrules, not  
234 even pseudomorphosed chondrules as observed in heavily altered CM1 chondrites (Zolensky  
235 et al., 1997; Vacher et al., 2018). It has been proposed that mafic minerals mechanically  
236 isolated from CIs could correspond to chondrule fragments (McSween, 1977; Richardson and  
237 McSween, 1978; Jones, 1992; Leshin et al., 1997) or grains formed by condensation  
238 processes (Steele, 1986; Weinbruch et al., 1993). A magmatic origin for the CI olivine grains  
239 is supported by the presence of glass inclusions (Fig. 1) and normal igneous zonings within  
240 isolated olivine grains (Leshin et al., 1997). In addition, the covariation of CaO with olivine  
241 Fo content is similar to that observed within chondrules (Leshin et al., 1997; Villeneuve et al.,  
242 2015; Marrocchi et al., 2019), suggesting that chondrules could represent a potential  
243 constituent of CI chondrites. However, the presence of chondrule-like olivine grains in CI  
244 chondrites does not imply that the mechanism(s) at the origin of chondrules operated in the

245 CI-forming region as they could also have been inherited from other chondrule-forming  
246 regions. Interestingly, CI olivine grains show  $^{17,18}\text{O}$ -rich isotopic compositions compared to  
247 other chondrites (Tenner et al., 2018; Marrocchi et al., 2019), at the exception of CR, ordinary  
248 and enstatite chondrites that commonly show type I chondrules with  $\Delta^{17}\text{O} \approx 0 \text{ ‰}$  (Schrader et  
249 al., 2013; Tenner et al., 2015, 2018). However,  $\mu^{26}\text{Mg}^*-\mu^{54}\text{Cr}$  systematics of CI and CR  
250 chondrites suggest they accreted in spatially isolated regions during the evolution of the  
251 protoplanetary disk (van Kooten et al., 2016). Furthermore, chondrule olivines are extremely  
252 rare in enstatite chondrite chondrules (Piani et al., 2016) and commonly show  $\Delta^{17}\text{O} > 0 \text{ ‰}$  in  
253 ordinary chondrites (Tenner et al., 2018), making their contribution to the CI budget unlikely.

254 CI olivine grains also show similarities with crystalline silicates from comet 81P/Wild  
255 2 samples and interplanetary dust particles (IDPs) that show a large range of Fo contents  
256 ( $\text{Fo}_{60-99}$ ; Zolensky and Barrett, 1994; Zolensky et al., 2006; Nakamura et al., 2008; Starkey  
257 and Franchi, 2013; Frank et al., 2014). In addition, the large majority of olivine grains  
258 recovered from comet 81P/Wild 2 aerogel tracks have  $^{16}\text{O}$ -poor isotopic compositions with  
259  $\Delta^{17}\text{O} \approx 0 \text{ ‰}$  (Fig. 5; McKeegan et al., 2006; Nakamura et al., 2008; Ogliore et al., 2012;  
260 Defouilloy et al., 2017), similar to the  $\Delta^{17}\text{O}$  values of olivine grains isolated from Orgueil and  
261 Ivuna (Leshin et al., 1997; this study). Although showing a large range of O isotopic  
262 compositions (i.e.,  $-40 \text{ ‰} < \delta^{18}\text{O} < +200 \text{ ‰}$ ), IDP silicate minerals commonly cluster around  
263 the intersection of the PCM and TFL (Aléon et al., 2009; Nakashima et al., 2012; Starkey and  
264 Franchi, 2013; Starkey et al., 2014). Thus, the picture that emerges from petrographic and  
265 isotopic characterization of mafic minerals isolated from CI chondrites is that (i) chondrules  
266 could have represent a potential constituent of CIs before the onset of aqueous alteration and  
267 (ii) cometary materials could have sampled mafic minerals similar to those observed in CI  
268 chondrites.

269

## 270 **4.2 Insights into the compositions of primordial dust and water of the Solar System**

271

272 Our data represent the first reported SIMS measurements of carbonates in CI  
273 chondrites. Carbonates in both Orgueil and Ivuna plot on the TFL with mean  $\Delta^{17}\text{O}$  values of  
274  $0.01 \pm 0.18\text{‰}$  and  $0.39 \pm 0.17\text{‰}$ , respectively (Fig. 3, Table 3). These results are similar,  
275 within errors, to those reported based on  $\text{H}_3\text{PO}_4$  acid treatments of the whole rocks (Clayton  
276 and Mayeda, 1984; Leshin et al., 2001). Similar to the olivine grains (see section 4.1), it is  
277 unlikely that calcite and dolomite grains correspond to terrestrial contamination as Mn-Cr  
278 studies revealed systematic  $^{53}\text{Cr}$  excesses induced by the radioactive decay of  $^{53}\text{Mn}$  (Petitat et  
279 al., 2009, 2011; Fujiya et al., 2013). Our results thus suggest that all CI carbonates  
280 precipitated from a fluid with Earth-like O isotopic compositions (i.e.,  $\Delta^{17}\text{O} \approx 0\text{‰}$ ; Fig. 3B).  
281 In addition, CI chondrite matrices, which are largely dominated by phyllosilicates (i.e., 80–  
282 85%; King et al., 2015), show  $\Delta^{17}\text{O} = 0.34\text{‰}$  (Clayton and Mayeda, 1999) that is similar  
283 within error to our results from CI Ca-carbonates (Table 2). This supports the formation of CI  
284 chondrite secondary phases from a fluid with Earth-like O isotope composition (i.e.,  $\Delta^{17}\text{O} \approx$   
285  $0\text{‰}$ ), as also inferred from pyrolysis (Baker et al., 2002). CI chondrites thus does not show  
286 significant  $^{17,18}\text{O}$  enrichments that would have been expected if the CI parent body had  
287 formed in a region of the Solar System or the parent molecular cloud where self-shielding of  
288 UV light by  $^{16}\text{O}$ -rich nebular CO would have produced  $^{16}\text{O}$ -poor water (Thiemens and  
289 Heidenreich, 1983; Clayton, 2002; Yurimoto and Kuramoto, 2004; Desch et al., 2018).  
290 Instead, CI water is characterized by Earth-like O isotopic compositions (Fig. 3). However,  
291 this may not represent the primordial isotopic composition of water ice grains in the CI-  
292 accreting region and does not necessarily exclude the self-shielding process. As CIs formed  
293 relatively late in the disk history (i.e.,  $\sim 3$  My after CAIs, Sugiura and Fujiya, 2014), water ice  
294 could have experienced thermal processing and O-isotope exchange with amorphous silicates

295 and CO in the protoplanetary disk. For instance, recent experimental approach points out that  
296 amorphous silicate grains (i.e., the main constituent of primordial chondritic matrices; Le  
297 Guillou et al., 2015) would experience oxygen isotopic exchange with water vapor within the  
298 lifetime of the disk at temperatures above  $\sim 500\text{-}650$  K (Yamamoto et al., 2019).  
299 Alternatively, potentially  $^{17,18}\text{O}$ -rich CI water could also have experienced isotopic exchange  
300 on the CI,  $^{16}\text{O}$ -rich, parent body. This would shift the water O isotope composition toward  
301 Earth-like compositions as commonly observed for other chondrites (CR, CM, CV, CO; e.g.  
302 Horstmann et al., 2014; Fujiya et al., 2015; Vacher et al., 2017, 2018, 2019; Verdier-Paoletti  
303 et al., 2017, 2019; Jilly-Rehak et al., 2017; Marrocchi et al., 2018b). This is supported by the  
304 heavy O isotope compositions of CI magnetite grains (i.e.,  $\Delta^{17}\text{O} = 1.67\text{‰}$ , Fig. 3B; Clayton  
305 and Mayeda, 1999) that are statistically different from those of other secondary phases. Such  
306 values cannot result from temperature-driven mass-dependent fractionations and thus suggest  
307 that initial CI water could have had a  $\Delta^{17}\text{O}$  value slightly above 0‰. However, the isotopic  
308 exchange during the evolution of the CI parent body should have been limited due to the  
309 small  $\Delta^{17}\text{O}$  variations observed between CI magnetite and phyllosilicates + olivines (whose  
310  $\Delta^{17}\text{O}$  is assumed to represent the O isotope composition of the CI anhydrous protolith). Taken  
311 together, this suggests that CI parent body accreted water ice grains with Earth-like  
312 composition (i.e.,  $\Delta^{17}\text{O} \approx 0$ ).

313 CI anhydrous silicates also show terrestrial values ( $\Delta^{17}\text{O} \approx 0\text{‰}$ ; Fig. 3), implying that  
314 the different constituents of CI chondrites all had Earth-like O isotopic compositions.  
315 Although CI chondrites are slightly depleted in oxygen relative to the Sun's photosphere  
316 (Lodders, 2003), our data show that CIs are chemically solar but isotopically terrestrial for  
317 oxygen isotopes. Interestingly, this is not the case for non-traditional isotopic system (e.g.,  
318  $^{50}\text{Ti}$ ,  $^{54}\text{Cr}$ ; Dauphas 2017). However, our results provide a strong argument in favor of a  
319 scenario where the primordial dust of the Solar System would have been  $^{16}\text{O}$ -poor (with  $\Delta^{17}\text{O}$

320  $\approx 0$ ) relative to the  $^{16}\text{O}$ -rich nebular gas (Krot et al., 2010). In this scenario,  $^{16}\text{O}$ -rich and  $^{16}\text{O}$ -  
321 poor reservoirs could have co-existed in the protoplanetary disk, with nebular regions  
322 characterized by enhanced dust/gas ratios being  $^{16}\text{O}$ -poor relative to solar due to the  
323 overabundance of  $^{16}\text{O}$ -poor solids. Our data thus suggest that primordial Solar System dust  
324 could correspond to CI-like material characterized by a  $^{16}\text{O}$ -poor isotopic composition.

325

### 326 **4.3 $\delta^{17}\text{O}$ - $\delta^{18}\text{O}$ of primordial CI matrix - Implications on chondrule-matrix** 327 **complementary**

328

329         Understanding the potential genetic link between chondrules and chondritic matrices is  
330 fundamental, as it would bring invaluable constraints on the origin of dust in the  
331 protoplanetary disk. Based on their elemental compositions (Hezel and Palme, 2010; Palme et  
332 al., 2015; Hezel et al., 2017, 2018), it has been proposed that the chondrules and matrices of  
333 carbonaceous chondrites formed simultaneously from the same solar-like reservoir. These  
334 reservoirs would have had major element compositions (i.e., Mg, Si, and Fe) similar to CI  
335 chondrites, but with variable degrees of depletion in volatile elements depending on the  
336 chondrite group (Hezel and Palme, 2010; Palme et al., 2015; Hezel et al., 2018). Chondrule-  
337 matrix complementarity has recently been supported by tungsten isotopic measurements of  
338 the matrix and chondrules in Allende (Budde et al., 2016). However, the intrinsic nature, and  
339 indeed the existence, of the chondrule-matrix complementarity are hotly debated. Alternative  
340 models propose that bulk chondrite compositions are better reproduced by considering  
341 (i) a non-CI complementarity (Bland et al., 2005) or (ii) a unique CI-composition matrix  
342 added to various amounts of chondrules (Anders, 1964; Zanda et al., 2018). The latter model  
343 implies that chondrules and chondritic matrices are not genetically related, but formed in  
344 different reservoirs (Zanda et al., 2018).

345 This complementarity is not straightforward for oxygen isotopes as the chondrule  
346 oxygen isotopic compositions show large intra- and inter-chondrule variations (Clayton and  
347 Mayeda, 1984; Marrocchi and Chaussidon, 2015; Tenner et al., 2018) requiring at least three  
348 reservoirs (Palme et al., 2015). However, the matrices accreted by the different groups of  
349 carbonaceous chondrites experienced secondary alteration processes under varying conditions  
350 (Brearley, 2006; Le Guillou et al., 2015; Vacher et al., 2017; Ganino and Libourel, 2017;  
351 Marrocchi et al., 2018b) that affected their O isotopic signatures and now preclude  
352 straightforward determination of their initial compositions. Thus, based on our data, we  
353 calculated the isotopic compositions of the different carbonaceous chondrite (hereafter, CC)  
354 matrices before the onset of alteration by considering the following. It has been shown that  
355 CM, CO, CR, and CV bulk O isotopic compositions have well-defined trends in a  $\delta^{17}\text{O}$ - $\delta^{18}\text{O}$   
356 diagram that correspond to variable extents of isotopic exchange between a  $^{16}\text{O}$ -rich  
357 anhydrous protolith and  $^{17,18}\text{O}$ -rich water (Clayton and Mayeda, 1984; Marrocchi et al.,  
358 2018b; Fujiya, 2018). We calculated the coordinates of the intersections of these respective  
359 trends with the PCM, which correspond to the bulk anhydrous O isotopic compositions of the  
360 different CCs before overprinting by parent-body processes (see Fig. 6 for a schematic  
361 example, Table 4). Importantly, none of the CC groups investigated here has the same bulk  
362 anhydrous O isotopic composition as CIs (Fig. 7A, Table 4). By considering the modal  
363 abundances of Ca-Al-rich inclusions (CAIs), chondrules, and matrix in the different CCs  
364 (Scott and Krot, 2014), we calculated the  $\Delta^{17}\text{O}$  values of the respective pre-alteration matrices  
365 accreted by the different CCs, assuming that (i) CAIs have an average  $\Delta^{17}\text{O}$  value of  $-25\%$   
366 (Kawasaki et al., 2017; Aléon, 2018), (ii) chondrules have variable  $\Delta^{17}\text{O}$  values in the range  
367 of  $-6$  to  $-2\%$  (Tenner et al., 2018), and (iii) CAIs, chondrules, and matrices have similar  
368 oxygen contents.

369 This calculation reveals that the pristine O isotopic compositions of CC matrices  
 370 differed significantly from that of CIs ( $\Delta^{17}\text{O}$  values of CM, CO, and CV matrices being  $-3.97$   
 371  $\pm 1.19\text{‰}$ ,  $-4.33 \pm 1.45\text{‰}$ , and  $-7.95 \pm 1.95\text{‰}$ , respectively), except for CR matrices, which  
 372 had a  $\Delta^{17}\text{O}$  value of  $-0.07 \pm 1.16\text{‰}$  (Fig. 7B, Table 4). This result confirms that the  
 373 complementarity concept cannot be applied to oxygen, mainly because chondrules were  
 374 formed through complex processes of precursor recycling and gas-melt interactions  
 375 (Tissandier et al., 2002; Hezel et al., 2006; Libourel et al., 2006; Schrader et al., 2013, 2018;  
 376 Marrocchi and Libourel, 2013; Marrocchi and Chaussidon, 2015; Friend et al., 2016;  
 377 Marrocchi et al., 2016; Piani et al., 2016). Hence, in the framework of the complementarity  
 378 model, the oxygen budget of carbonaceous chondrites can be written as:

379

$$380 \quad \text{O}_{\text{Bulk CC}} = \text{O}_{\text{Chondrules}} + \text{O}_{\text{Matrix}} \quad (1)$$

381 with

$$382 \quad \text{O}_{\text{Chondrules}} = \text{O}_{\text{Precursors}} + \text{O}_{\text{Gas}} \quad (2)$$

383 and

$$384 \quad \Delta^{17}\text{O}_{\text{Matrix}} (\text{CC}) \neq \Delta^{17}\text{O}_{\text{Matrix}} (\text{CI}) \quad (3)$$

385 However, recent studies have demonstrated that partially molten chondrule precursors  
 386 experienced protracted gas-melt interactions with gaseous SiO and Mg (Libourel and Portail,  
 387 2018; Marrocchi et al., 2018a; Marrocchi et al., 2019). In addition, mass balance calculations  
 388 based on the abundance of sulfides in CV chondrules suggest that about half of their total Fe  
 389 content originated from the gas phase (Marrocchi and Libourel, 2013). These constraints  
 390 suggest that the Mg-Si-Fe budget was strongly affected by gas-melt interactions in the same  
 391 way as oxygen:

392

$$393 \quad (\text{Mg, Si, Fe})_{\text{Bulk CC}} = (\text{Mg, Si, Fe})_{\text{Chondrules}} + (\text{Mg, Si, Fe})_{\text{Matrix}} \quad (4)$$

394 with

$$395 \quad (\text{Mg, Si, Fe})_{\text{Chondrules}} = (\text{Mg, Si, Fe})_{\text{Precursors}} + (\text{Mg, Si, Fe})_{\text{Gas}} \quad (5)$$

396

397 If chondrules and matrices formed complementarily from the same reservoir, this implies that  
398 (i) this reservoir must have been in a closed system with the gas itself, or (ii) the gas was of  
399 CI composition to satisfy the Mg-Si-Fe elemental mass balance. In the protoplanetary disk,  
400 the canonical dust to gas ratio is commonly taken as  $\sim 10^{-2}$  (Birnstiel et al., 2010), meaning  
401 that the condensation of solids and their subsequent interactions with the gas should not have  
402 significantly affected the composition of the gas, which should have maintained, by definition,  
403 a solar composition. However, given that disk evolution and differential dynamical coupling  
404 with the gas motion depends on grain size, the dust/gas ratio was probably heterogeneous,  
405 with chondrules having potentially formed under a large range of dust enhancements (Cuzzi  
406 et al., 2008; Alexander et al., 2012; Marrocchi and Chaussidon, 2015). In such dusty regions,  
407 rapid heating and evaporation of the dust could have lead to environments where the gas had a  
408 predominantly CI-like composition.

409 It has recently been proposed that chondrules result from the recycling of previously  
410 condensed amoeboid olivine aggregates (AOAs; Jacquet and Marrocchi, 2017; Marrocchi et  
411 al., 2018a; Marrocchi et al., 2019). This implies that AOAs experienced rapid and efficient  
412 transport through regions of the protoplanetary disk where the gaseous reservoir was  $^{17,18}\text{O}$ -  
413 enriched relative to their condensation zones (Marrocchi et al., 2019). This outward transport  
414 of AOAs could have been achieved through viscous spreading, as recently proposed for CAIs  
415 (Nanne et al., 2019). However, maintaining a closed (chondrule + gas + matrix) system at the  
416 scale of the protoplanetary disk to satisfy the (O, Mg, Si, Fe) mass balance appears unrealistic  
417 and imposes important constraints on the complementarity model. Interestingly, large-scale  
418 isotopic heterogeneities within the disk, such as the non-carbonaceous–carbonaceous

419 chondrite nucleosynthetic dichotomy (Kruijer et al., 2017; Gerber et al., 2017; Nanne et al.,  
420 2019), could reflect internal heterogeneities within the Solar System's parental molecular  
421 cloud itself, with different regions of the nascent disk receiving variable compositions of  
422 infalling material (Nanne et al., 2019). This supports models where the O isotopic  
423 heterogeneity of the Solar System was partially established by self-shielding in the molecular  
424 cloud (Yurimoto and Kuramoto, 2004). Such complex gas-melt interactions during chondrule  
425 formation should be better accounted for to improve our understanding of the potential  
426 chondrule-matrix complementarity.

427

## 428 **Conclusions**

429

430 We have performed *in-situ* SIMS oxygen isotopic analyses of olivines and carbonates  
431 in the CI chondrites Orgueil and Ivuna. Our main results are:

432 1. All olivine grains but one show O isotopic compositions plotting at the intersection  
433 of the TFL and PCM.

434 2. Ca-carbonates describe a trend with  $\delta^{17}\text{O} = (0.50 \pm 0.05) \times \delta^{18}\text{O} + (0.9 \pm 1.4)$  that  
435 differs from mass-independent variation observed in secondary phases of other carbonaceous  
436 chondrites.

437

438 From these data, we drew the following interpretations:

439

440 1. Olivine grains isolated from Orgueil could correspond to chondrule fragments.

441 2. The O isotopic compositions of CI anhydrous solids and water show Earth-like  
442 values, thus defining a fundamental characteristic of CIs that appear to be chemically solar  
443 but isotopically terrestrial for oxygen isotopes.

444 3. The  $^{16}\text{O}$ -poor composition of CI anhydrous dust particles supports models where  
445 primordial Solar System dust is characterized by terrestrial values ( $\Delta^{17}\text{O} \approx 0\text{‰}$ ).

446 4. Anhydrous bulk O isotopic compositions of CCs are distinct from the anhydrous CI  
447 bulk composition. Likewise, the pre-alteration O isotopic composition of the CI matrix is  
448 different from those of CCs, except for CR chondrites. This highlights that matrix-chondrule  
449 complementarity does not apply to oxygen isotopes, possibly as the result of complex gas-  
450 melt interactions during chondrule formation within the protoplanetary disk.

451 5. Recent chondrule formation models have proposed that complex interactions  
452 between chondrule precursors and Mg-Si-Fe-rich gas are fundamental for establishing  
453 chondrule characteristics. If chondrules and matrix formed complementarily from the same  
454 reservoir, either the reservoir must have been in a closed system with the gas or the gas must  
455 have been of CI composition to satisfy Mg-Si-Fe elemental mass balance.

456 6. Chondrules experienced gas-melt interactions with a  $^{17,18}\text{O}$ -rich gaseous reservoir  
457 that differs significantly from the  $^{16}\text{O}$ -rich isotopic composition of chondrule precursors.  
458 Assuming a spatial origin of these isotopic variations, rapid and efficient transport toward  
459  $^{17,18}\text{O}$ -enriched regions could have been achieved through viscous spreading, as recently  
460 proposed for CAIs. However, maintaining a closed (chondrule + gas + matrix) system at the  
461 scale of the protoplanetary disk to satisfy the (O, Mg, Si, Fe) mass balance puts important  
462 constraints on the complementarity model.

463

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470

471 **Research data**

472 Original data of this study are available at Mendeley Data (Marrocchi and Piralla,  
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789 **Figure captions**

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791 **Fig. 1.** (A) Optical image of a mechanically isolated olivine grain from Orgueil. (B, C)  
792 Defocused optical images of olivine grains showing a potential fluid/melt inclusion near a  
793 grain boundary (B) and silicate melt along grain boundaries (C).

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795 **Fig. 2.** Composite EDX elemental map of Mg (red), Fe (blue), and Ca (green) in a  
796 representative section of Ivuna, revealing the preferential distribution of carbonates (green) in  
797 the bottom left of the section. Anhydrous silicates were not observed in this section.

798

799 **Fig. 3.** (A) Oxygen three-isotope plot of olivine and carbonate grains from Ivuna and Orgueil  
800 CI chondrites. Olivine grains show terrestrial isotopic values except for olivine grain O16-8,  
801 which is enriched in  $^{16}\text{O}$  and plots on the PCM below the TFL. Carbonate data define a linear  
802 trend that is indistinguishable from the TFL within errors [ $\delta^{17}\text{O} = (0.50 \pm 0.05) \times \delta^{18}\text{O} + (0.9$   
803  $\pm 1.4)$ ]. Uncertainties shown are  $2\sigma$ . (B)  $\Delta^{17}\text{O}$  vs.  $\delta^{18}\text{O}$  values of components of Ivuna and  
804 Orgueil: except for magnetite grains and olivine grain O16-8, all data plot, within errors, on  
805 the TFL. Uncertainties shown are  $2\sigma$ . Data from Clayton and Mayeda, 1984; Rowe et al.,  
806 1994; Leshin et al., 1997; Clayton and Mayeda, 1999.

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808 **Fig. 4.** Distributions of fayalite content (mol. %) for olivine in CI chondrites (data from this  
809 study and Reid et al., 1970; Kerridge and MacDougall, 1976; Steele, 1990; Leshin et al.,  
810 1997; Le Gac et al., 2009; Frank et al., 2014).

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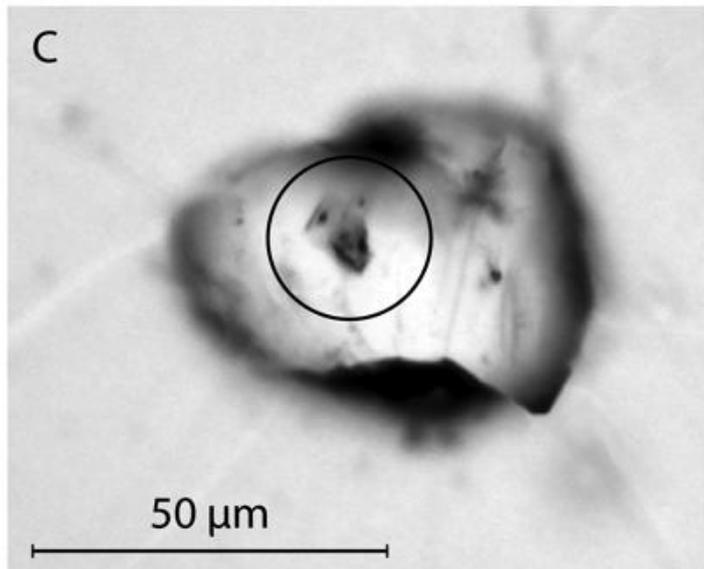
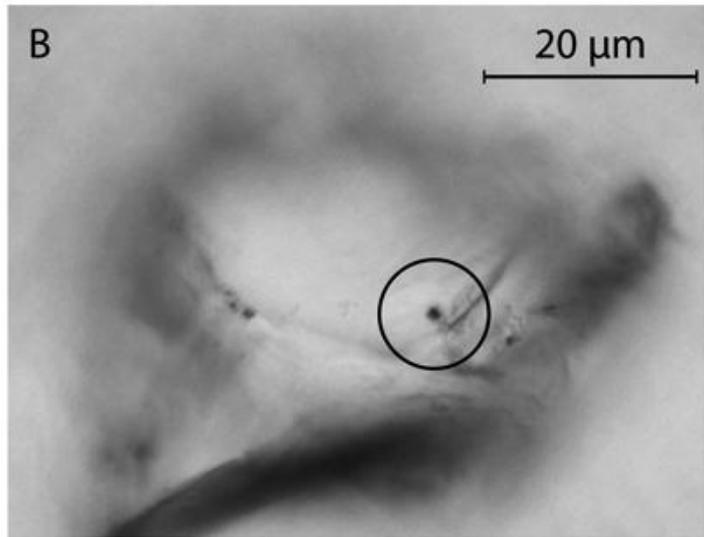
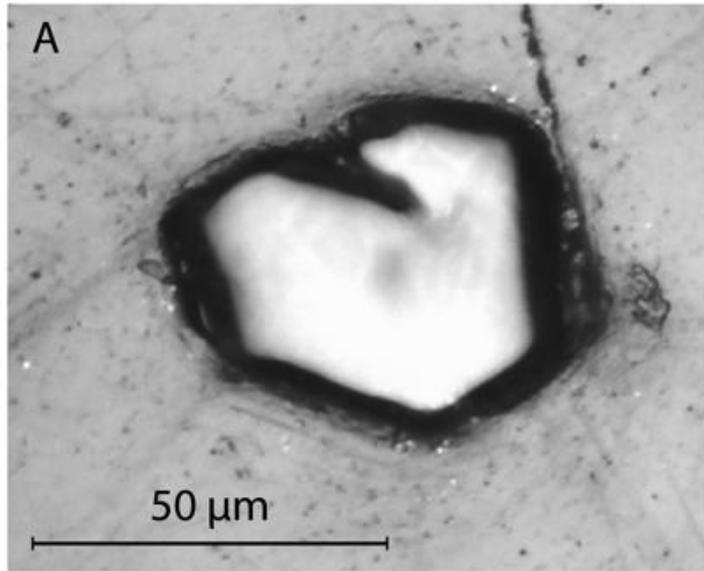
812 **Fig. 5.** Probability density functions of the  $\Delta^{17}\text{O}$  values of silicates in comets and IDPs (n =  
813 146, green) and CI chondrites (olivine and pyroxene, n = 41, orange; see text for references).

814 **Fig. 6.** Schematic oxygen three-isotope plot of bulk CM chondrites. The intersection between  
815 this trend and the PCM corresponds to the bulk anhydrous O isotope composition of CM

816 chondrites before overprinting by parent-body processes. It should be noted that using the  
817 CCAM instead of the PCM does not affect our conclusions within errors.

818

819 **Fig. 7.** (A) Calculated bulk  $\Delta^{17}\text{O}$  values of primordial carbonaceous chondrites before the  
820 onset of aqueous alteration (CM = green, CO = red, CV = blue, and CR = purple). (B)  
821 Calculated  $\Delta^{17}\text{O}$  of primordial, pre-alteration matrices in CM, CO, CV, and CR chondrites.  
822 The dashed orange line in both plots corresponds to the CI value ( $\Delta^{17}\text{O} = 0\%$ ). Uncertainties  
823 shown in both plots are  $2\sigma$ . See text for details.



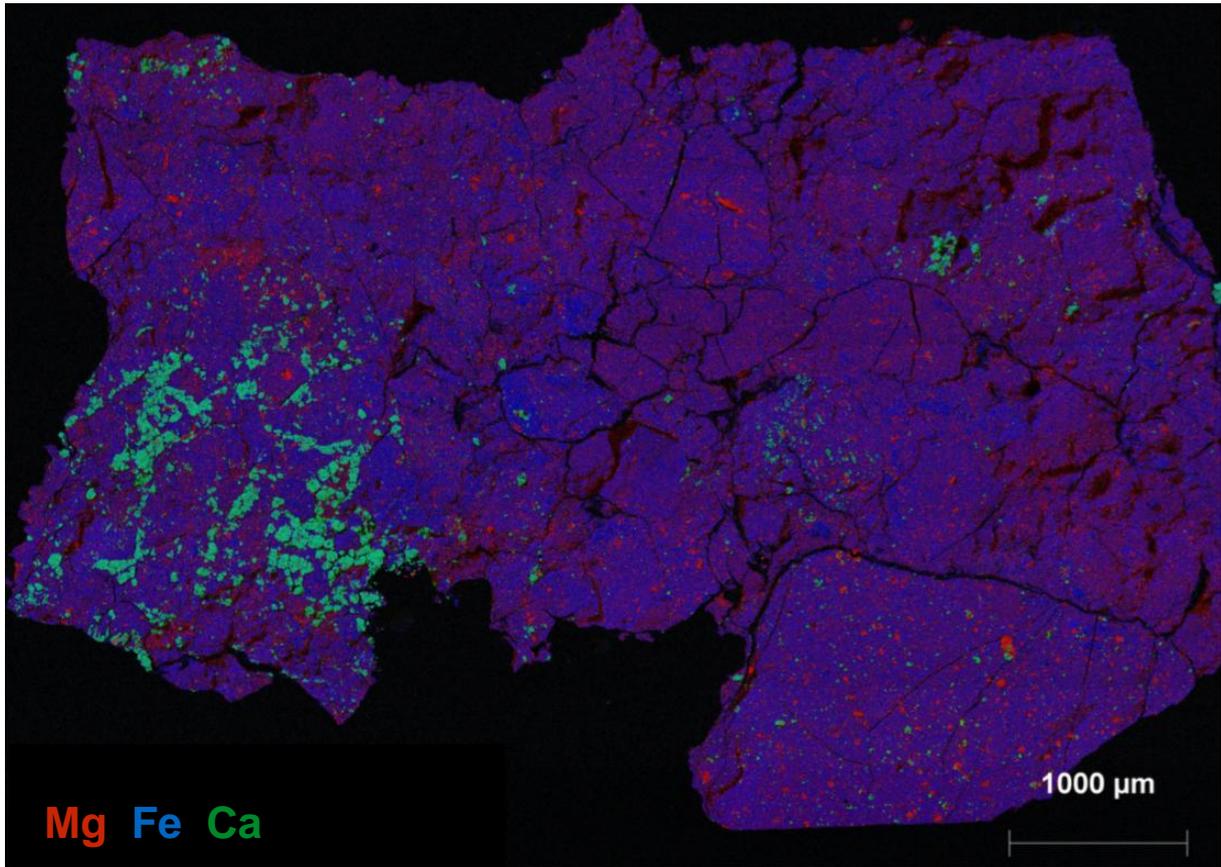
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**Fig. 1**

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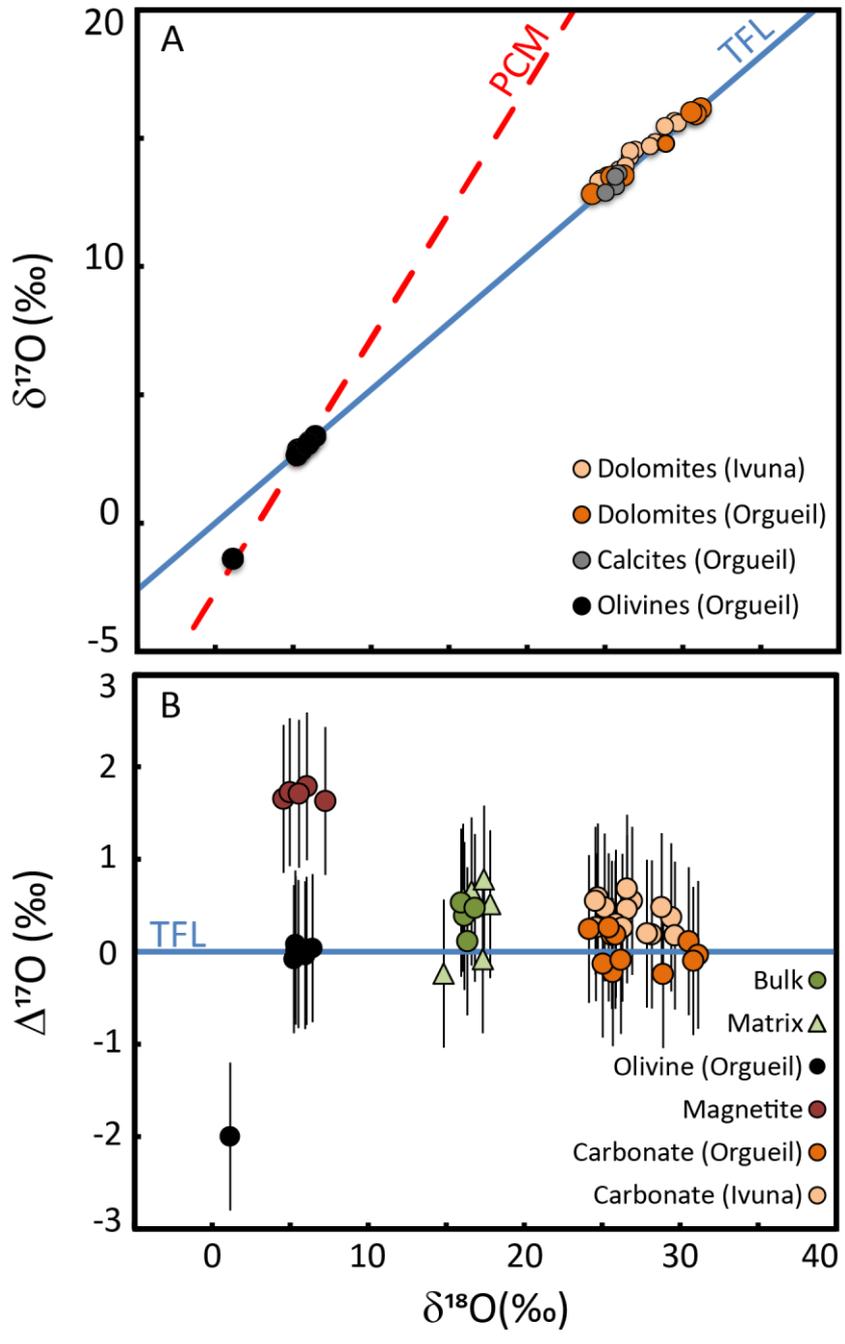
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**Fig. 2**

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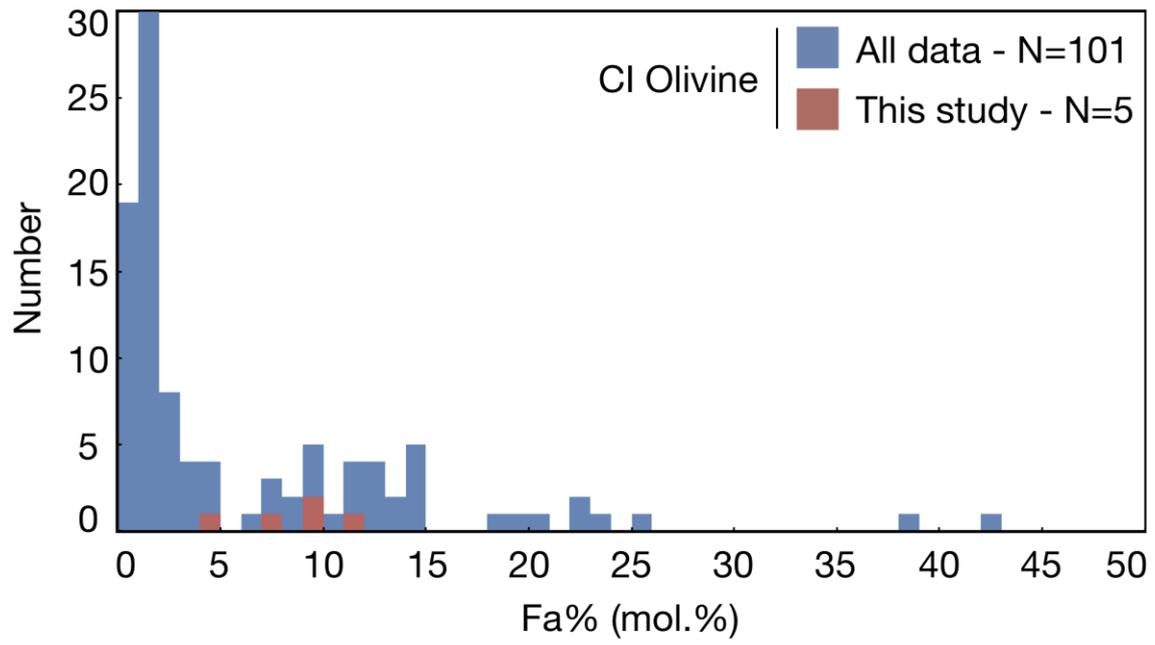


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Fig. 3



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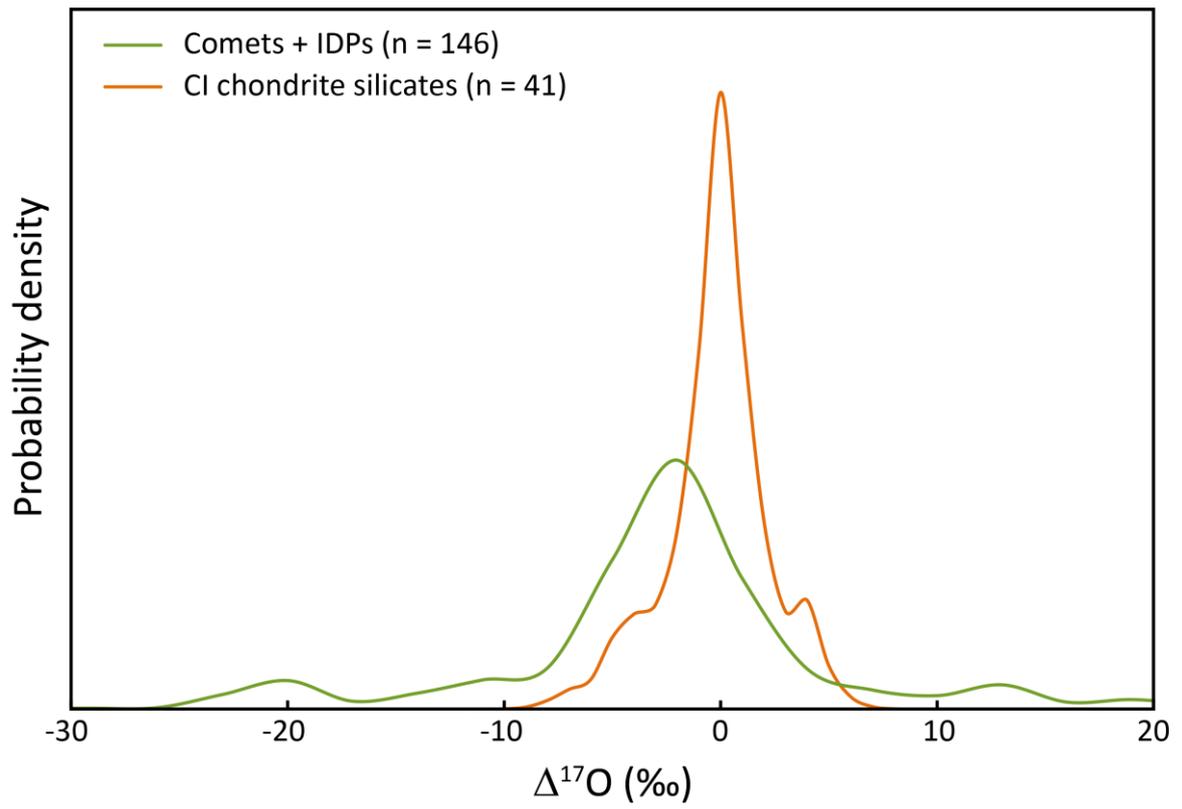
**Fig. 4**

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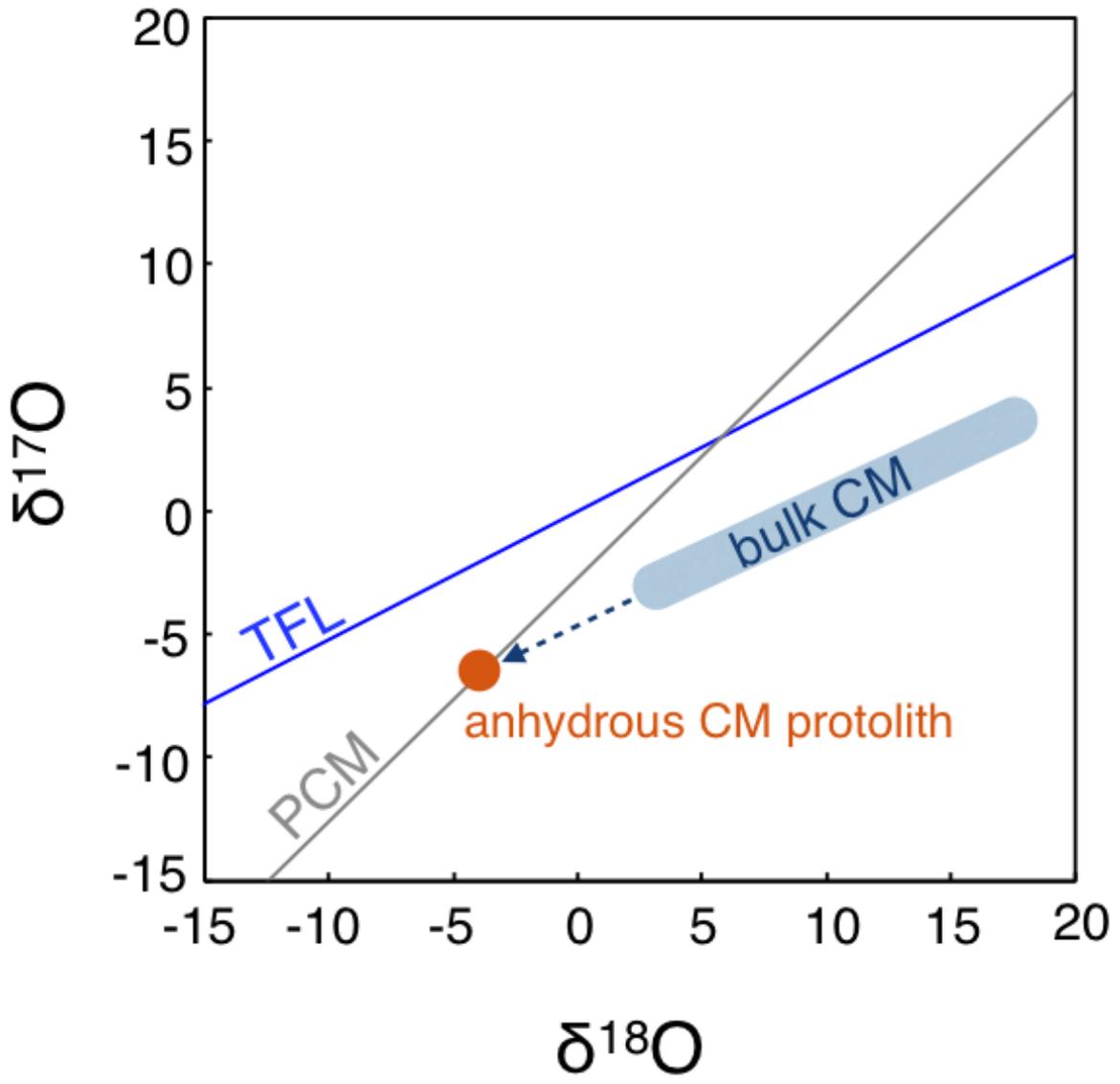
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**Fig. 5**



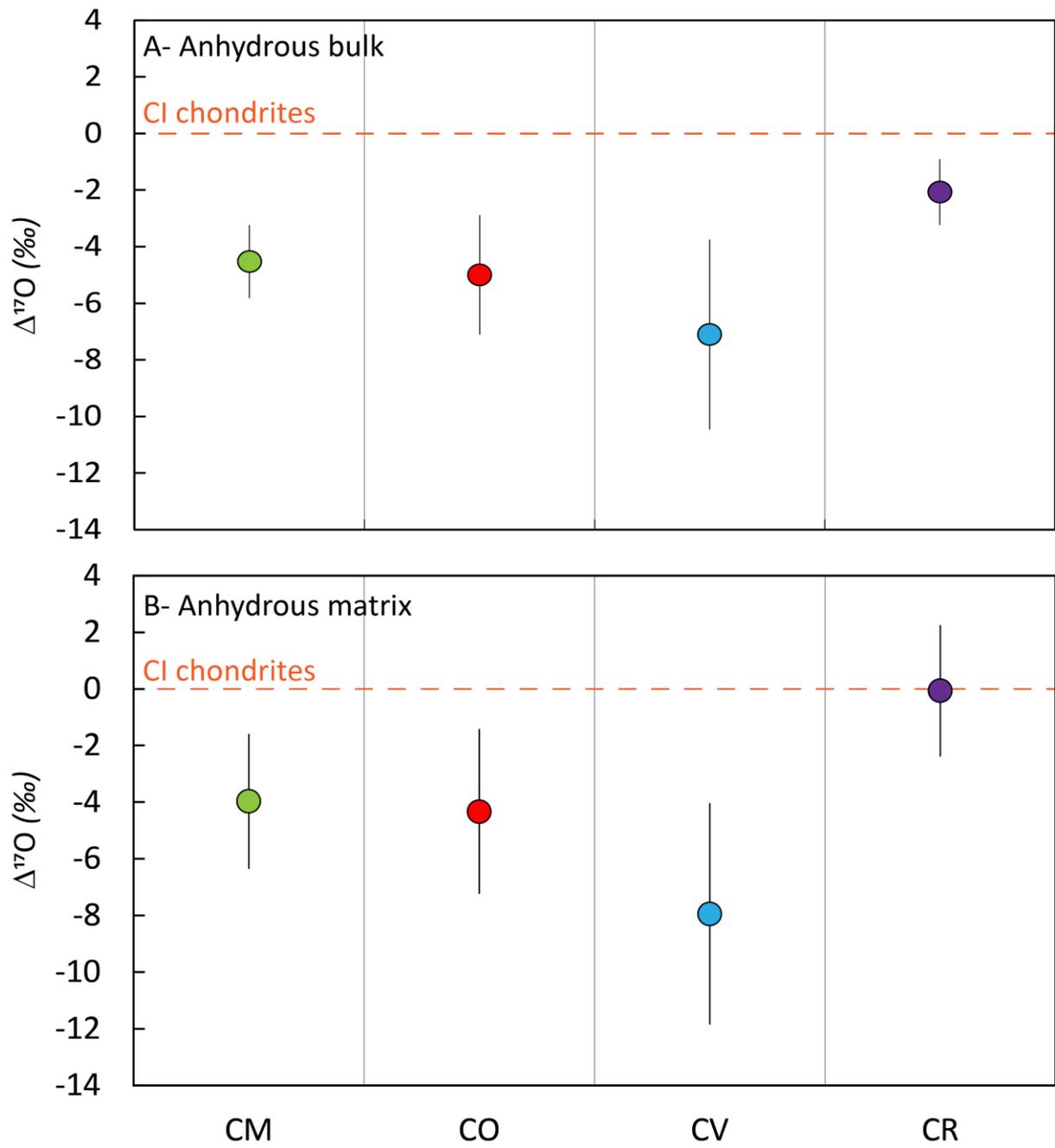
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Fig. 6

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**Fig. 7**

Meteorite	Mineralogy	Sample	SiO <sub>2</sub>	MgO	FeO	Al <sub>2</sub> O <sub>3</sub>	CaO	Cr <sub>2</sub> O <sub>3</sub>	MnO	Total	Mg#
Orgueil	Olivine	O6-18	42.9	49.7	9.0	<i>bdl</i>	0.1	<i>bdl</i>	0.1	101.7	0.91
		O6-23	42.9	49.4	8.9	<i>bdl</i>	0.1	<i>bdl</i>	0.1	101.5	0.91
		O6-24	42.2	47.4	11.5	<i>bdl</i>	0.1	<i>bdl</i>	0.2	101.3	0.88
		O16-8	43.8	53.4	4.7	0.2	0.2	0.6	0.1	102.9	0.95
		O21-5	43.8	50.0	7.7	<i>bdl</i>	0.1	0.5	0.3	102.4	0.92

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Table 1. Chemical compositions and Mg# of olivine mechanically isolated from Orgueil. *Bdl* = below detection limit.

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Chondrite	Mineralogy	Sample	wt.%						mol.%						Molar Mg/Ca	
			CaO	MgO	FeO	MnO	Na <sub>2</sub> O	SiO <sub>2</sub>	Ca	Mg	Fe	Mn	Na	Si		
Orgueil	Calcite	O21-7	54.56	0.41	<i>bdl</i>	<i>bdl</i>	<i>bdl</i>	<i>bdl</i>	98.97	1.03	-	-	-	-	0.0	
		O21-8	57.88	0.16	<i>bdl</i>	<i>bdl</i>	<i>bdl</i>	<i>bdl</i>	99.63	0.37	-	-	-	-	0.0	
		O21-9	56.45	0.27	<i>bdl</i>	<i>bdl</i>	<i>bdl</i>	<i>bdl</i>	99.34	0.66	-	-	-	-	0.0	
	Dolomite	O6-1	27.78	16.37	1.64	3.47	<i>bdl</i>	0.11	50.81	41.66	2.33	5.02	-	0.18	0.8	
		O6-12	29.05	18.96	1.69	2.10	<i>bdl</i>	<i>bdl</i>	49.73	45.17	2.25	2.84	-	-	0.9	
		O6-17	26.87	18.96	1.75	4.83	<i>bdl</i>	<i>bdl</i>	46.00	45.14	2.33	6.53	-	-	1.0	
		O15-1	27.34	19.27	1.57	3.02	<i>bdl</i>	0.37	47.05	46.14	2.11	4.11	-	0.59	1.0	
		O15-3	27.76	18.74	1.81	2.66	<i>bdl</i>	<i>bdl</i>	48.41	45.46	2.46	3.67	-	-	0.9	
		O15-5	28.79	19.12	1.50	2.52	<i>bdl</i>	<i>bdl</i>	49.17	45.43	2.00	3.40	-	-	0.9	
		O16-4	27.04	14.78	1.88	5.13	<i>bdl</i>	<i>bdl</i>	50.91	38.71	2.76	7.63	-	-	0.8	
		O16-6	26.79	18.15	1.89	3.71	<i>bdl</i>	<i>bdl</i>	47.46	44.73	2.61	5.20	-	-	0.9	
		O16-7	27.57	19.34	1.76	3.30	<i>bdl</i>	0.06	47.13	45.99	2.34	4.45	-	0.09	1.0	
		O16-9	25.03	18.69	2.35	5.68	<i>bdl</i>	<i>bdl</i>	43.64	45.34	3.19	7.83	-	-	1.0	
		O17-2	27.51	17.30	1.34	5.18	<i>bdl</i>	0.12	48.41	42.35	1.83	7.21	-	0.20	0.9	
		O17-7	28.19	17.36	1.80	4.09	<i>bdl</i>	0.25	49.27	42.22	2.46	5.65	-	0.41	0.9	
		O21-1	30.72	20.00	1.36	3.45	<i>bdl</i>	<i>bdl</i>	49.28	44.64	1.70	4.37	-	-	0.9	
		Ferromagnesite	O6-2	0.18	34.94	11.88	7.01	<i>bdl</i>	0.06	0.27	76.37	14.56	8.71	-	0.08	277.8
			O6-7	0.72	28.79	15.14	10.43	<i>bdl</i>	0.06	1.18	65.79	19.41	13.54	-	0.08	55.6
			O6-10	0.31	33.94	16.32	3.73	<i>bdl</i>	<i>bdl</i>	0.48	74.71	20.15	4.66	-	-	154.8
			O7-8	0.19	31.41	16.71	6.32	<i>bdl</i>	0.12	0.31	70.44	21.03	8.05	-	0.17	230.0
O7-13	0.57		31.09	12.49	10.13	<i>bdl</i>	0.22	0.91	70.02	15.78	12.96	-	0.32	76.6		
O7-18	0.10		34.38	16.56	2.80	<i>bdl</i>	<i>bdl</i>	0.15	75.85	20.49	3.51	-	-	503.4		
O17-6	0.30		29.75	21.62	2.72	<i>bdl</i>	0.18	0.49	67.98	27.72	3.53	-	0.28	138.0		
O21-6	<i>bdl</i>		47.06	0.51	<i>bdl</i>	<i>bdl</i>	<i>bdl</i>	-	99.40	0.60	-	-	-	-		

Ivuna	Dolomite															
		I-1	31.76	19.51	1.47	0.95	<i>bdl</i>	<i>bdl</i>	52.24	44.64	1.89	1.23	-	-	0.9	
		I-2	25.86	18.29	1.93	4.55	0.16	1.05	44.84	44.12	2.61	6.24	0.49	1.70	1.0	
		I-3	31.92	19.83	1.28	0.39	0.06	<i>bdl</i>	52.40	45.29	1.65	0.50	0.17	-	0.9	
		I-4	30.23	20.45	2.17	1.63	0.07	<i>bdl</i>	48.92	46.05	2.75	2.09	0.19	-	0.9	
		I-5	29.33	19.15	1.25	4.16	<i>bdl</i>	<i>bdl</i>	48.69	44.23	1.62	5.45	-	-	0.9	
		I-6	28.54	20.32	2.06	1.16	0.07	0.11	47.92	47.47	2.70	1.54	0.21	0.17	1.0	
		I-7	28.38	20.86	2.19	1.23	0.10	0.40	46.82	47.86	2.81	1.60	0.28	0.62	1.0	
		I-8	25.02	21.83	1.49	3.68	0.08	0.26	41.80	50.75	1.95	4.86	0.24	0.41	1.2	
		I-9	30.00	18.77	1.98	2.85	<i>bdl</i>	<i>bdl</i>	50.07	43.59	2.58	3.76	-	-	0.9	
		I-10	26.42	17.67	2.69	2.42	0.11	1.05	47.02	43.75	3.74	3.41	0.34	1.75	0.9	
		I-11	28.31	19.18	2.09	2.11	<i>bdl</i>	0.29	48.33	45.57	2.79	2.85	-	0.46	0.9	
		I-12	27.71	18.46	1.51	4.62	<i>bdl</i>	0.05	47.55	44.07	2.02	6.27	-	0.09	0.9	
		I-13	28.29	19.98	2.11	1.08	0.06	0.16	48.07	47.25	2.80	1.46	0.17	0.26	1.0	
		I-14	29.75	20.75	2.20	1.11	<i>bdl</i>	0.06	48.56	47.12	2.80	1.43	-	0.09	1.0	
		I-15	13.84	23.05	2.56	3.44	0.12	1.28	26.60	61.64	3.83	5.23	0.40	2.29	2.3	
		I-16	27.74	18.78	1.54	4.30	<i>bdl</i>	<i>bdl</i>	47.44	44.69	2.05	5.81	-	-	0.9	

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866 Table 2. Chemical compositions of carbonate grains (calcite, dolomite, and ferromagnesite) mechanically isolated from Orgueil. The values of  
867 dolomite grains measured in the thick section of Ivuna are also reported (see text for details). *Bdl* = below detection limit.  
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Meteorite	Mineralogy	Sample	$\delta^{18}\text{O}$	$2\sigma$	$\delta^{17}\text{O}$	$2\sigma$	$\Delta^{17}\text{O}$	$2\sigma$
Orgueil	Olivine	O21-5	6.1	0.2	3.2	0.3	0.0	0.9
			5.5	0.2	2.8	0.3	0.0	0.8
		O6-18	5.3	0.2	2.9	0.3	0.1	0.9
		O6-23	6.4	0.2	3.4	0.3	0.0	0.9
			5.3	0.2	2.8	0.3	0.0	0.8
		O6-24	5.2	0.2	2.6	0.3	-0.1	0.8
			6.0	0.2	3.1	0.3	0.0	0.8
		O16-8	1.1	0.2	-1.4	0.4	-2.0	0.8
	Calcite	O21-7	25.6	0.1	13.5	0.4	0.2	0.8
		O21-8	25.9	0.1	13.6	0.4	0.2	0.9
		O21-9	25.7	0.1	13.1	0.4	-0.2	0.8
		O21-10	25.0	0.1	12.9	0.4	-0.1	0.9
	Dolomite	O21-1	30.6	0.0	16.0	0.2	0.1	0.8
		O17-7	24.2	0.0	12.8	0.2	0.2	0.9
		O16-6	26.2	0.0	13.5	0.2	-0.1	0.9
		O16-9	25.4	0.0	13.5	0.2	0.3	0.8
		O16-7	31.2	0.0	16.2	0.2	0.0	0.8
		O6-12	28.9	0.0	14.8	0.2	-0.2	0.8
		O6-17	30.9	0.0	15.9	0.2	-0.1	0.8
Ivuna	Dolomite	I-1	26.9	0.1	14.5	0.1	0.5	0.8
		I-2	29.4	0.1	15.7	0.1	0.4	0.7
		I-3	26.6	0.1	14.3	0.1	0.5	0.8
		I-4	29.7	0.1	15.6	0.1	0.2	0.7
		I-5	26.6	0.1	14.5	0.1	0.7	0.7
		I-6	28.8	0.1	15.5	0.1	0.5	0.7

I-7	24.7	0.1	13.4	0.1	0.6	0.6
I-8	28.2	0.1	14.8	0.1	0.2	0.6
I-9	25.2	0.1	13.6	0.1	0.5	0.8
I-10	24.6	0.1	13.1	0.1	0.3	0.6
I-11	25.9	0.1	13.8	0.1	0.3	0.6
I-12	27.9	0.1	14.7	0.1	0.2	0.7
I-13	24.6	0.1	13.3	0.1	0.5	0.7
I-14	26.3	0.1	13.9	0.1	0.3	0.8

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Table 3. Oxygen isotopic compositions of olivine, calcites, and dolomites mechanically isolated from Orgueil. The oxygen isotopic compositions of dolomite grains measured in the thick section of Ivuna are also reported (see text for details).

		$\delta^{18}\text{O}$	$2\sigma$	$\delta^{17}\text{O}$	$2\sigma$	$\Delta^{17}\text{O}$	$2\sigma$
bulk	CM	-3.9	0.5	-6.6	0.4	-4.5	0.6
	CO	-4.9	0.9	-7.6	0.6	-5.0	1.1
	CV	-9.4	1.3	-12.0	1.1	-7.1	1.7
	CR	1.3	0.5	-1.4	0.3	-2.1	0.6
matrix	CM	-3.0	0.9	-5.5	0.8	-4.0	1.2
	CO	-4.0	1.1	-6.4	0.9	-4.3	1.5
	CV	-11.9	1.5	-14.1	1.3	-7.9	2.0
	CR	4.7	0.9	2.4	0.8	-0.1	1.2

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890 Table 4. Calculated primordial (pre-alteration) bulk and matrix oxygen isotopic compositions for different carbonaceous chondrites. Primordial  
891 compositions were calculated from the bulk O isotopic trends (Marrocchi et al., 2018b), the modal abundances of CAIs, chondrules, and matrix  
892 (Scott and Krot, 2014), and the average O isotopic compositions of CAIs and chondrules for each type of carbonaceous chondrite (Tenner et al.,  
893 2018).

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