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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}O -poor ($\Delta^{17}\text{O} \approx 0\text{‰}$) relative to the ^{16}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, ²⁶Al-free, ¹⁶O-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 ¹⁶O-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., $\sim 4.5\%$, 2σ , 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 H_3PO_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 µm, 50–100 µm, and >100 µm 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 cm²), 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 × 768 analysis points were collected at a pixel-
141 spacing of 1.52 µm. 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 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 ~ 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 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 2.8×10^9 cps for ^{16}O , 1.1×10^6 cps for
175 ^{17}O , and 6.3×10^6 cps for ^{18}O in carbonates, and 2.7×10^9 cps for ^{16}O , 1.0×10^6 cps for ^{17}O ,
176 and 5.5×10^6 cps for ^{18}O in San Carlos olivine. Typical 2σ 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 (Fo_{88-95}) and had low abundances of minor elements
189 such as Ca (≤ 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 (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‰ (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 (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 (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}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 H_3PO_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}Cr excesses induced by the radioactive decay of ^{53}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‰), 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}O -rich nebular CO would have produced ^{16}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., ~ 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}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}Ti , ^{54}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}O -poor (with $\Delta^{17}\text{O}$

320 ≈ 0) relative to the ^{16}O -rich nebular gas (Krot et al., 2010). In this scenario, ^{16}O -rich and ^{16}O -
321 poor reservoirs could have co-existed in the protoplanetary disk, with nebular regions
322 characterized by enhanced dust/gas ratios being ^{16}O -poor relative to solar due to the
323 overabundance of ^{16}O -poor solids. Our data thus suggest that primordial Solar System dust
324 could correspond to CI-like material characterized by a ^{16}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}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}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}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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474 **References**

- 475
- 476 Aléon J. (2018) Closed system oxygen isotope redistribution in igneous CAIs upon spinel
477 dissolution. *Earth Planet. Sci. Lett.* 482, 324–333.
- 478 Aléon J., Engrand C., Leshin L. A. and McKeegan K. D. (2009) Oxygen isotopic composition
479 of chondritic interplanetary dust particles: A genetic link between carbonaceous chondrites
480 and comets. *Geochim. Cosmochim. Acta* 73, 4558–4575.
- 481 Alexander C. M. O'D, Bowden R., Fogel M. L., Howard K. T., Herd C. D. K. and Nittler L. R.
482 (2012) The Provenances of Asteroids, and Their Contributions to the Volatile Inventories
483 of the Terrestrial Planets. *Science* 337, 721–723.
- 484 Alexander C. M. O'D, Bowden R., Fogel M. L. and Howard K. T. (2015) Carbonate
485 abundances and isotopic compositions in chondrites. *Meteorit Planet Sci* 50, 810–833.
- 486 Anders E. (1964) Origin, age, and composition of meteorites. *Space Sci Rev* 3, 583–714.
- 487 Baker L., Franchi I. A., Wright I. P. and Pillinger C. T. (2002) The oxygen isotopic
488 composition of water from Tagish Lake: Its relationship to low- temperature phases and to
489 other carbonaceous chondrites. *Meteoritics & Planetary Science* 37, 977–985.
- 490 Birnstiel T., Dullemond C.P. and Brauer F. (2010) Gas- and dust evolution in protoplanetary
491 disk. *Astron. & Astrophys.* 513, A79, 21 pp.
- 492 Bland P. A., Alard O., Benedix G. K., Kearsley A. T., Menzies O. N., Watt L. E. and Rogers
493 N. W. (2005) Volatile fractionation in the early solar system and chondrule/matrix
494 complementarity. *Proceedings of the National Academy of Sciences* 102, 13755–13760.
- 495 Brearley, A.J., 2006. The action of water. *Meteorites and the Early Solar System II*. (eds.
496 Lauretta D. S. and McSween H. Y. Tucson, Arizona: The University of Arizona Press. pp.
497 587–624.
- 498 Budde G., Kleine T., Kruijer T. S., Burkhardt C. and Metzler K. (2016) Tungsten isotopic
499 constraints on the age and origin of chondrules. *Proceedings of the National Academy of*
500 *Sciences* 113, 2886–2891.
- 501 Clayton R. N. (2002) Solar System: Self-shielding in the solar nebula. *Nature* 415, 860–861.
- 502 Clayton R. N. and Mayeda T. K. (1984) The oxygen isotope record in Murchison and other
503 carbonaceous chondrites. *Earth Planet. Sci. Lett.* 67, 151–161.
- 504 Clayton, R.N., Mayeda, T.K., 1999. Oxygen isotope studies of carbonaceous chondrites.
505 *Geochim. Cosmochim. Acta* 63, 2089–2104.
- 506 Cuzzi J. N., Hogan R. C. and Shariff K. (2008) Toward planetesimals: Dense chondrule
507 clumps in the protoplanetary nebula. *The Astrophysical Journal* 687, 1432–1447.
- 508 Dauphas N. (2017) The isotopic nature of the Earth's accreting material through time. *Nature*
509 *541*, 521-524.
- 510 Defouilloy C., Nakashima D., Joswiak D. J., Brownlee D. E., Tenner T. J. and Kita N. T.
511 (2017) Origin of crystalline silicates from Comet 81P/Wild 2: Combined study on their
512 oxygen isotopes and mineral chemistry. *Earth Planet. Sci. Lett.* 465, 145–154.
- 513 Desch S.J., Kalyaan A. and Alexander C. M. O'D (2018) The Effect of Jupiter's Formation on
514 the Distribution of Refractory Elements and Inclusions in Meteorites. *The Astrophysical J.*
515 *238*, 31.
- 516 Eiler J. M. (2001) Oxygen Isotope Variations of Basaltic Lavas and Upper Mantle Rocks.
517 *Reviews in Mineralogy and Geochemistry* 43, 319–364.
- 518 Frank D. R., Zolensky M. E. and Le L. (2014) Olivine in terminal particles of Stardust aerogel
519 tracks and analogous grains in chondrite matrix. *Geochim. Cosmochim. Acta* 142, 240-259.
- 520 Frank D., Zolensky M., Martinez J., Mikouchi T., Ohsumi K., Hagiya K., Satake W., Le L.,
521 Ross D. and Peslier A. (2011) A CAI in the Ivuna C11 chondrite. *Lunar Planet. Sci.* 47.
522 *Lunar Planet. Inst., Houston.* #1608.

523 Friend P., Hezel D. C. and Mucerschi D. (2016) The conditions of chondrule formation, Part
524 II: Open system. *Geochim. Cosmochim. Acta* 173, 198–209.

525 Fujiya W. (2018) Oxygen isotopic ratios of primordial water in carbonaceous chondrites.
526 *Earth Planet. Sci. Lett.* 481, 264–272.

527 Fujiya W., Sugiura N., Sano Y. and Hiyagon H. (2013) Mn–Cr ages of dolomites in CI
528 chondrites and the Tagish Lake ungrouped carbonaceous chondrite. *Earth Planet. Sci. Lett.*
529 362, 130–142.

530 Fujiya W., Sugiura N., Marrocchi Y., Takahata N., Hoppe P., Shirai K., Sano Y. and Hiyagon
531 H. (2015) Comprehensive study of carbon and oxygen isotopic compositions, trace
532 element abundances, and cathodoluminescence intensities of calcite in the Murchison CM
533 chondrite. *Geochim. Cosmochim. Acta* 161, 101–117.

534 Ganino C. and Libourel G. (2017) Reduced and unstratified crust in CV chondrite parent body.
535 *Nat Comms*, 1–10.

536 Gerber S., Burkhardt C., Budde G., Metzler K. and Kleine T. (2017) Mixing and Transport of
537 Dust in the Early Solar Nebula as Inferred from Titanium Isotope Variations among
538 Chondrules. *The Astrophysical Journal Letters* 841, 1–7.

539 Gounelle M. (2011) The Asteroid-Comet Continuum: In Search of Lost Primitivity. *Elements*
540 7, 29–34.

541 Gounelle M., Spurný P. and Bland P. A. (2006) The orbit and atmospheric trajectory of the
542 Orgueil meteorite from historical records. *Meteoritics & Planetary Science* 41, 135–150.

543 Grady M. M., Wright I. P., Swart P. K. and Pillinger C. T. (1988) The carbon and oxygen
544 isotopic composition of meteoritic carbonates. *Geochim. Cosmochim. Acta* 52, 2855–2866.

545 Guo W. and Eiler J. M. (2007) Temperatures of aqueous alteration and evidence for methane
546 generation on the parent bodies of the CM chondrites. *Geochim. Cosmochim. Acta* 71,
547 5565–5575.

548 Hezel D. C. and Palme H. (2010) The chemical relationship between chondrules and matrix
549 and the chondrule matrix complementarity. *Earth Planet. Sci. Lett.* 294, 85–93.

550 Hezel D. C., Harak M. and Libourel G. (2017) What we know about elemental bulk chondrule
551 and matrix compositions: Presenting the Chondrite DB Database. *Chemie der Erde -*
552 *Geochemistry*, 91-121.

553 Hezel D. C., Bland P. A., Palme H., Jacquet E. and Bigolski J. (2018) Composition of
554 Chondrules and Matrix and Their Complementary Relationship in Chondrites. In:
555 *Chondrules*. Cambridge University Press, pp 196-246, 441 pp.

556 Hezel D. C., Palme H., Nasdala L. and Brenker F. E. (2006) Origin of SiO₂-rich components
557 in ordinary chondrites. *Geochim. Cosmochim. Acta* 70, 1548–1564.

558 Horstmann M., Vollmer C., Birth M. I. F., Chaussidon M., Gurenko A. and Bischoff A.
559 (2014) Tracking aqueous alteration of CM chondrites—Insight from in-situ oxygen isotope
560 measurements of calcite. *Lunar Planet. Sci.* 45. Lunar Planet. Inst., Houston. #1761.

561 Hsieh H. H. and Jewitt D.J. (2006) A population of comets in the main asteroid belt. *Science*
562 312, 561-563.

563 Jacquet E. and Marrocchi Y. (2017) Chondrule heritage and thermal histories from trace
564 element and oxygen isotope analyses of chondrules and amoeboid olivine aggregates.
565 *Meteorit Planet Sci* 58, 3451–23.

566 Jilly-Rehak C. E., Huss G. R., Nagashima K. and Schrader D. L. (2017) Low-temperature
567 aqueous alteration on the CR chondrite parent body: implications from in situ oxygen-
568 isotope analyses. *Geochim. Cosmochim. Acta* 222, 230-252.

569 Jones R. H. (1992) On the relationship between isolated and chondrule olivine grains in the
570 carbonaceous chondrite ALHA 77307. *Geochim. Cosmochim. Acta* 56, 3593.

571 Kawasaki N., Itoh S., Sakamoto N. and Yurimoto H. (2017) Chronological study of oxygen
572 isotope composition for the solar protoplanetary disk recorded in a fluffy Type A CAI from
573 Vigarano. *Geochim. Cosmochim. Acta* 201, 83–102.

574 Kerridge J. F. and MacDougall J. D. (1976) Mafic silicates in the Orgueil carbonaceous
575 meteorite. *Earth Planet. Sci. Lett.* 29, 341–348.

576 King A. J., Schofield P. F., Howard K. T. and Russell S. S. (2015) Modal mineralogy of CI
577 and CI-like chondrites by X-ray diffraction. *Geochim. Cosmochim. Acta* 165, 148–160.

578 Krot A. N., Nagashima K., Ciesla F. J., Meyer B. S., Hutcheon I. D., Davis A. M., Huss G. R.
579 and Scott E. R. D. (2010) Oxygen isotopic composition of the sun and mean oxygen
580 isotopic composition of the protosolar silicate dust: evidence from refractory inclusions.
581 *Astrophys. J.* 713, 1159–1166.

582 Kruijjer T. S., Burkhardt C., Budde G. and Kleine T. (2017) Age of Jupiter inferred from the
583 distinct genetics and formation times of meteorites. *Proceedings of the National Academy
584 of Sciences* 114, 6712–6716.

585 Le Gac Y., Benedix G. K., Science P. B. A. P. (2009) Are CI Chondrites Cometary Samples?
586 Olivine as a Diagnostic Tool. 72th Meteoritics and Planetary Science Conference, 5242.

587 Le Guillou C., Changela H. G. and Brearley A. J. (2015) Widespread oxidized and hydrated
588 amorphous silicates in CR chondrites matrices: Implications for alteration conditions and
589 H₂ degassing of asteroids. *Earth Planet. Sci. Lett.* 420, 162–173.

590 Lee M. R., Sofo M. R., Lindgren P., Starkey N. A. and Franchi I. A. (2013) The oxygen
591 isotope evolution of parent body aqueous solutions as recorded by multiple carbonate
592 generations in the Lonewolf Nunataks 94101 CM2 carbonaceous chondrite. *Geochim.
593 Cosmochim. Acta* 121, 452–466.

594 Le Guillou C., Changela H. G. and Brearley A. J. (2015) Widespread oxidized and hydrated
595 amorphous silicates in CR chondrites matrices: Implications for alteration conditions and
596 H₂ degassing of asteroids. *Earth Planet. Sci. Lett.* 420, 162–173.

597 Leshin L. A., Rubin A. E. and McKeegan K. D. (1997) The oxygen isotopic composition of
598 olivine and pyroxene from CI chondrites. *Geochim. Cosmochim. Acta* 61, 835–845.

599 Leshin L. A., Farquhar J. and Guan Y. (2001) Oxygen isotopic anatomy of Tagish Lake:
600 Relationship to primary and secondary minerals in CI and CM chondrites. *Lunar Planet.
601 Sci.* 32. Lunar Planet. Inst., Houston. #1843.

602 Levison H. F., Bottke W. F., Gounelle M., Morbidelli A., Nesvorný D. and Tsiganis K.
603 (2009) Contamination of the asteroid belt by primordial trans-Neptunian objects. 460, 364–
604 366.

605 Libourel G. and Portail M. (2018) Chondrules as direct thermochemical sensors of solar
606 protoplanetary disk gas. *Science Advances* 4, eaar3321.

607 Libourel G., Krot A. and Tissandier L. (2006) Role of gas-melt interaction during chondrule
608 formation. *Earth and Planetary Science Letters* 251, 232–240.

609 Lodders K. (2003) Solar system abundances and condensation temperatures of the elements.
610 *The Astrophysical Journal* 591, 1220–1247.

611 Marrocchi Y. and Chaussidon M. (2015) A systematic for oxygen isotopic variation in
612 meteoritic chondrules. *Earth Planet. Sci. Lett.* 430, 308–315.

613 Marrocchi Y. and Libourel G. (2013) Sulfur and sulfides in chondrules. *Geochim.
614 Cosmochim. Acta* 119, 117–136.

615 Marrocchi Y., Villeneuve J., Batanova V., Piani L. and Jacquet E. (2018a) Oxygen isotopic
616 diversity of chondrule precursors and the nebular origin of chondrules. *Earth Planet. Sci.
617 Lett.* 496, 132–141.

618 Marrocchi Y., Bekaert D. V. and Piani L. (2018b) Origin and abundance of water in
619 carbonaceous asteroids. *Earth and Planetary Science Letters* 482, 23–32.

620 Marrocchi Y., Chaussidon M., Piani L. and Libourel G. (2016) Early scattering of the solar
621 protoplanetary disk recorded in meteoritic chondrules. *Science Advances* 2, e1601001–
622 e1601001.

623 Marrocchi Y., Euverte R., Villeneuve J., Batanova V., Welsch B., Ferrière L. and Jacquet E.
624 (2019) Formation of CV chondrules by recycling of amoeboid olivine aggregate-like
625 precursors. *Geochim. Cosmochim. Acta* 247, 121–141.

626 [dataset] Marrocchi Y. & Piralla M. (2019), “CI chondrites - Piralla”, Mendeley Data, V2
627 <http://dx.doi.org/10.17632/9xws4jx4fz.2>

628 McKeegan K. D., Aléon J., Bradley J., Brownlee D., Busemann H., Butterworth A.,
629 Chaussidon M., Fallon S., Floss C., Gilmour J., Gounelle M., Graham G., Guan Y., Heck P.
630 R., Hoppe P., Hutcheon I. D., Huth J., Ishii H., Ito M., Jacobsen S. B., Kearsley A., Leshin
631 L. A., Liu M. C., Lyon I., Marhas K., Marty B., Matrajt G., Meibom A., Messenger S.,
632 Mostefaoui S., Mukhopadhyay S., Nakamura-Messenger K., Nittler L., Palma R., Pepin R.
633 O., Papanastassiou D. A., Robert F., Schlutter D., Snead C. J., Stadermann F. J., Stroud R.,
634 Tsou P., Westphal A., Young E. D., Ziegler K., Zimmermann L. and Zinner E. (2006)
635 Isotopic Compositions of Cometary Matter Returned by Stardust. *Science* 314, 1724–1728.

636 McSween H. Y. (1977) On the nature and origin of isolated olivine grains in carbonaceous
637 chondrites. *Geochim. Cosmochim. Acta* 41, 411–418.

638 Nakamura T., Noguchi T., Tsuchiyama A., Ushikubo T., Kita N. T., Valley J. W., Zolensky
639 M. E., Kakazu Y., Sakamoto K., Mashio E., Uesugi K. and Nakano T. (2008) Chondrule-
640 like Objects in Short-Period Comet 81P/Wild 2. *Science* 321, 1664–1667.

641 Nakashima D., Ushikubo T., Zolensky M. E. and Kita N. T. (2012) High precision oxygen
642 three-isotope analyses of anhydrous chondritic interplanetary dust particles. *Meteoritics &*
643 *Planetary Science* 47, 197–208.

644 Nanne J. A. M., Nimmo F., Cuzzi J. N. and Kleine T. (2019) Origin of the non-carbonaceous–
645 carbonaceous meteorite dichotomy. *Earth Planet. Sci. Lett.* 511, 44–54.

646 Nittler L. R., Davidson J., Liu N., Alexander C. M. O. D. and Stroud R. M. (2015) A Search
647 for Cosmic Symplectite in the Acfer 094 and Miller Range 07687 Meteorites. *Lunar*
648 *Planet. Sci.* 46. Lunar Planet. Inst., Houston. #1832.

649 Ogliore R. C., Huss G. R., Nagashima K., Butterworth A. L., Gainsforth Z., Stodolna J.,
650 Westphal A. J., Joswiak D. and Tyliszczak T. (2012) Incorporation of a late-forming
651 chondrule into comet wild 2. *Astrophys. J. Lett.* 745, L19–5.

652 Palme H., Hezel D. C. and Ebel D. S. (2015) The origin of chondrules: Constraints from
653 matrix composition and matrix-chondrule complementarity. *Earth Planet. Sci. Lett.* 411,
654 11–19.

655 Petitat M., McKeegan K., Sci M. G. L. P.2009 Duration and sequence of carbonate
656 crystallization on the Orgueil protolith: ^{53}Mn – ^{53}Cr systematics of their evolution in O and
657 C isotopic composition. *Lunar Planet. Sci.* 40. Lunar Planet. Inst., Houston. #1657.

658 Petitat M., Marrocchi Y., McKeegan K. D., Mostefaoui S., Meibom A., Zolensky M. E. and
659 Gounelle M. (2011) ^{53}Mn – ^{53}Cr ages of Kaidun carbonates. *Meteoritics & Planetary*
660 *Science* 46, 275–283.

661 Piani L., Marrocchi Y., Libourel G. and Tissandier L. (2016) Magmatic sulfides in the
662 porphyritic chondrules of EH enstatite chondrites. *Geochim. Cosmochim. Acta* 195, 84–99.

663 Reid A. M., Bass M. N., Fujita H., Kerridge J. F. and Fredriksson K. (1970) Olivine and
664 pyroxene in the Orgueil meteorite. *Geochim. Cosmochim. Acta* 34, 1253–1255.

665 Richardson S. M. and McSween H. Y. (1978) Textural evidence bearing on the origin of
666 isolated olivine crystals in C2 carbonaceous chondrites. *Earth Planet. Sci. Lett.* 37, 485–
667 491.

668 Rowe M. W., Clayton R. N. and Mayeda T. K. (1994) Oxygen isotopes in separated
669 components of CI and CM meteorites. *Geochim. Cosmochim. Acta* 58, 5341–5347.

670 Sakamoto N., Seto Y., Itoh S., Kuramoto K., Fujino K., Nagashima K., Krot A. N. and
671 Yurimoto H. (2007) Remnants of the Early Solar System Water Enriched in Heavy
672 Oxygen Isotopes. *Science* 317, 231–233.

673 Schrader D. L., Connolly H. C. Jr, Lauretta D. S., Nagashima K., Huss G. R., Davidson J. and
674 Domanik K. J. (2013) The formation and alteration of the Renazzo-like carbonaceous
675 chondrites II: Linking O-isotope composition and oxidation state of chondrule olivine.
676 *Geochim. Cosmochim. Acta* 101, 302–327.

677 Schrader D. L., Nagashima K., Waitukaitis S. R., Davidson J., McCoy T. J., Connolly H. C.
678 and Lauretta D. S. (2018) The retention of dust in protoplanetary disks: Evidence from
679 agglomeratic olivine chondrules from the outer Solar System. *Geochim. Cosmochim. Acta*
680 223, 405–421.

681 Scott E. R. D. and Krot A. N. (2014) Chondrites and Their Components. In *Meteorites and*
682 *Cosmochemical Processes* pp. 65–137.

683 Seto Y., Sakamoto N., Fujino K., Kaito T., Oikawa T. and Yurimoto H. (2008) Mineralogical
684 characterization of a unique material having heavy oxygen isotope anomaly in matrix of
685 the primitive carbonaceous chondrite Acfer 094. *Geochim. Cosmochim. Acta* 72, 2723–
686 2734.

687 Starkey N. A. and Franchi I. A. (2013) Insight into the silicate and organic reservoirs of the
688 comet forming region. *Geochim. Cosmochim. Acta* 105, 73–91.

689 Starkey N. A., Franchi I. A. and Lee M. R. (2014) Isotopic diversity in interplanetary dust
690 particles and preservation of extreme ¹⁶O-depletion. *Geochim. Cosmochim. Acta* 142,
691 115–131.

692 Steele I. M. (1986) Compositions and textures of relic forsterite in carbonaceous and
693 unequilibrated ordinary chondrites. *Geochim. Cosmochim. Acta* 50, 1379–1395.

694 Steele I.M. 1990. Minor elements in forsterites of Orgueil (C1), Alais (C1) and two
695 interplanetary dust particles compared to C2-C3-UOC forsterites. *Meteoritics and*
696 *Planetary Science* 25, 301-307.

697 Tenner T. J., Nakashima D., Ushikubo T., Kita N. T. and Weisberg M. K. (2015) Oxygen
698 isotope ratios of FeO-poor chondrules in CR3 chondrites: Influence of dust enrichment and
699 H₂O during chondrule formation. *Geochim. Cosmochim. Acta* 148, 228–250.

700 Tenner T. J., Ushikubo T., Nakashima D., Schrader D. L., Weisberg M. K., Kimura M. and
701 Kita N. T. (2018) Oxygen Isotope Characteristics of Chondrules from Recent Studies by
702 Secondary Ion Mass Spectrometry. In: *Chondrules*. Cambridge University Press, pp 196-
703 246, 441 pp.

704 Thiemens M. H. and Heidenreich J. E. (1983) The Mass-Independent Fractionation of
705 Oxygen: A Novel Isotope Effect and Its Possible Cosmochemical Implications. *Science*
706 219, 1073–1075.

707 Tissandier L., Libourel G. and Robert F. (2002) Gas-melt interactions and their bearing on
708 chondrule formation. *Meteoritics & Planetary Science* 37, 1377–1389.

709 Tyra M. A., Farquhar J., Guan Y. and Leshin L. A. (2012) An oxygen isotope dichotomy in
710 CM2 chondritic carbonates: A SIMS approach. *Geochim. Cosmochim. Acta* 77, 383–395.

711 Tyra M., Brearley A. and Guan Y. (2016) Episodic carbonate precipitation in the CM
712 chondrite ALH 84049: An ion microprobe analysis of O and C isotopes. *Geochim.*
713 *Cosmochim. Acta* 175, 195–207.

714 Ushikubo T., Kimura M., Kita N. T. and Valley J. W. (2012) Primordial oxygen isotope
715 reservoirs of the solar nebula recorded in chondrules in Acfer 094 carbonaceous chondrite.
716 *Geochim. Cosmochim. Acta* 90, 242–264.

717 Vacher L. G., Marrocchi Y., Verdier-Paoletti M. J., Villeneuve J. and Gounelle M. (2016)
718 Inward radial mixing of interstellar water ices in the solar protoplanetary disk. *The*
719 *Astrophysical Journal Letters* 827, 1–6.

720 Vacher L. G., Marrocchi Y., Villeneuve J., Verdier-Paoletti M. J. and Gounelle M. (2017)
721 Petrographic and C & O isotopic characteristics of the earliest stages of aqueous alteration of
722 CM chondrites. *Geochim. Cosmochim. Acta* 213, 271–290.

723 Vacher L. G., Marrocchi Y., Villeneuve J., Verdier-Paoletti M. J. and Gounelle M. (2018)
724 Collisional and alteration history of the CM parent body. *Geochim. Cosmochim. Acta* 239,
725 213–234.

726 Vacher L.G., Piralla M., Gounelle M., Bizzarro M. and Marrocchi Y., Thermal evolution of
727 hydrated asteroids inferred from oxygen isotopes. *The Astrophysical Journal Letters* 882,
728 L20.

729 van Kooten E.M.M.E., Moynier F. and Agranier A. (2019) A unifying model for the accretion
730 of chondrules and matrix. *PNAS*, pnas.1907592116.

731 Verdier-Paoletti M. J., Marrocchi Y., Avice G., Roskosz M., Gurenko A. and Gounelle M.
732 (2017) Oxygen isotope constraints on the alteration temperatures of CM chondrites. *Earth*
733 *Planet. Sci. Lett.* 458, 273–281.

734 Verdier-Paoletti M.J., Marrocchi Y., Vacher L.G., Gattacceca J., Gurenko A., Sonzogni C. &
735 Gounelle M. (2019). Testing the genetic relationship between fluid alteration and
736 brecciation in CM chondrites. *Meteoritics & Planetary Science* 54, 1692-1709.

737 Villeneuve J., Libourel G. and Soulié C. (2015) Relationships between type I and type II
738 chondrules: Implications on chondrule formation processes. *Geochim. Cosmochim. Acta*
739 160, 277–305.

740 Weinbruch S., Zinner E. K. and El Goresy A. (1993) Oxygen isotopic composition of
741 individual olivine grains from the Allende meteorite. *Geochim. Cosmochim. Acta* 57,
742 2649–2661.

743 Yurimoto H. and Kuramoto K. (2004) Molecular cloud origin for the oxygen isotope
744 heterogeneity in the solar system. *Science* 305, 1763–1766.

745 Zanda B., Lewin E. and Humayun M. (2018) The Chondritic Assemblage. In *Chondrules:*
746 *Records of Protoplanetary Disk Processes Complementarity Is Not a Required Hypothesis.*
747 *In: Chondrules.* Cambridge University Press pp. 122–150.

748 Zolensky M. E. and Barrett R. A. (1994) Compositional variations of olivines and pyroxenes
749 in chondritic interplanetary dust particles. *Meteoritics & Planetary Science* 29, 616–620.

750 Zolensky M. E., Mittlefehldt D. W. and Lipschutz M. E. (1997) CM chondrites exhibit the
751 complete petrologic range from type 2 to 1. *Geochim. Cosmochim. Acta* 61, 5099–5115.

752 Zolensky M. E., Zega T. J., Yano H., Wirick S., Westphal A. J., Weisberg M. K., Weber I.,
753 Warren J. L., Velbel M. A., Tsuchiyama A., Tsou P., Toppani A., Tomioka N., Tomeoka
754 K., Teslich N., Taheri M., Susini J., Stroud R., Stephan T., Stadermann F. J., Snead C. J.,
755 Simon S. B., Simionovici A., See T. H., Robert F., Rietmeijer F. J. M., Rao W., Perronnet
756 M. C., Papanastassiou D. A., Okudaira K., Ohsumi K., Ohnishi I., Nakamura-Messenger
757 K., Nakamura T., Mostefaoui S., Mikouchi T., Meibom A., Matrajt G., Marcus M. A.,
758 Leroux H., Lemelle L., Le L., Lanzirotti A., Langenhorst F., Krot A. N., Keller, L. P.,
759 Kearsley A. T., Joswiak D., Jacob D., Ishii H., Harvey R., Hagiya K., Grossman L.,
760 Grossman J. N., Graham G. A., Gounelle M., Gillet P., Genge M. J., Flynn G., Ferroir T.,
761 Fallon S., Ebel D. S., Dai Z. R., Cordier P., Clark B., Chi M., Butterworth A. L., Brownlee
762 D. E., Bridges J. C., Brennan S., Brearley A., Bradley J. P., Bleuet P., Bland P. A. and
763 Bastien R. (2006) Mineralogy and Petrology of Comet 81P/Wild 2 Nucleus Samples.
764 *Science* 314, 1735–1739.

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789 **Figure captions**

790

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}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 2σ . (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 2σ . 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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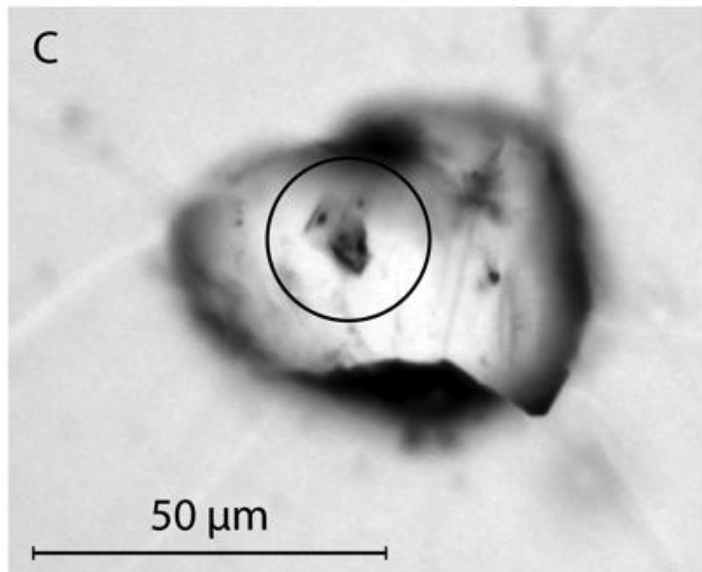
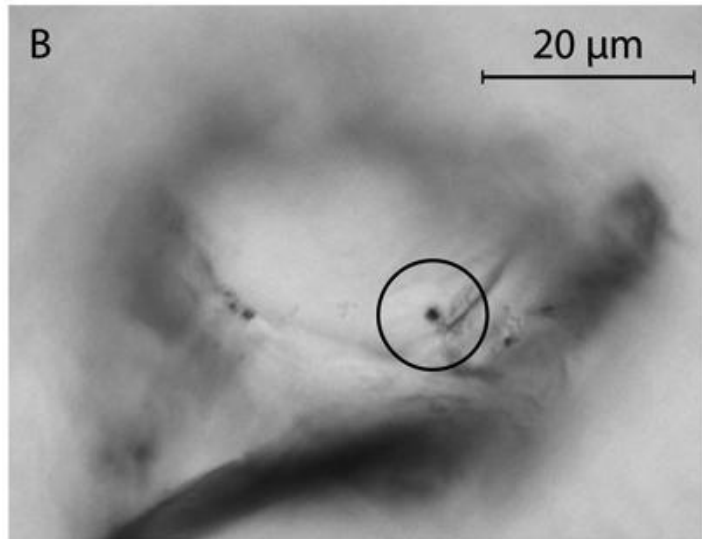
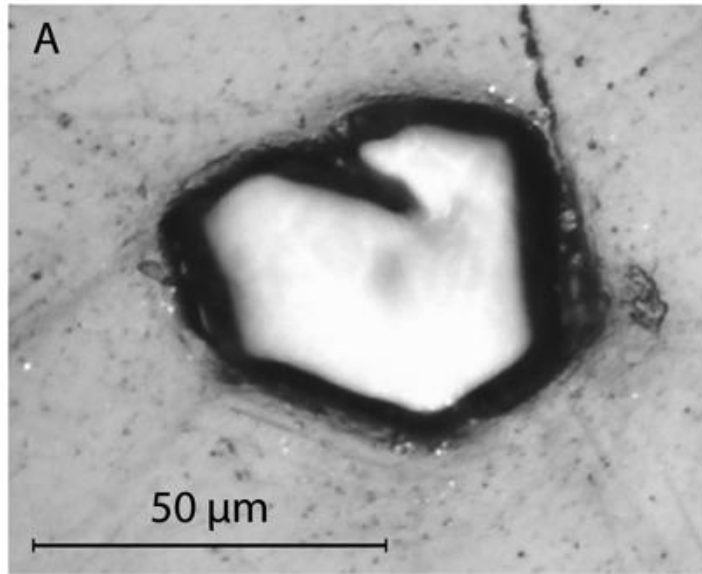
812 **Fig. 5.** Probability density functions of the $\Delta^{17}\text{O}$ values of silicates in comets and IDPs (n =
813 146, green) and CI chondrites (olivine and pyroxene, n = 41, orange; see text for references).

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 2σ . See text for details.



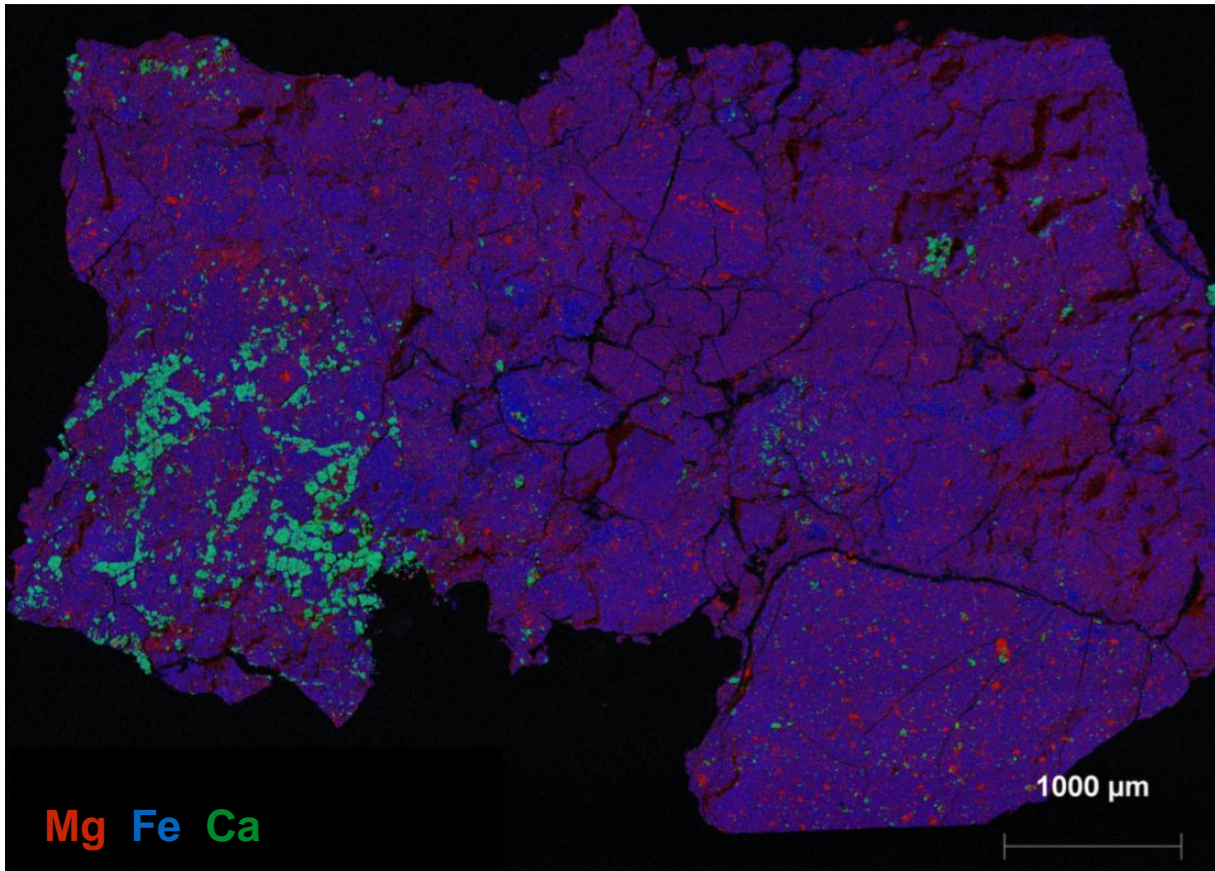
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Fig. 1

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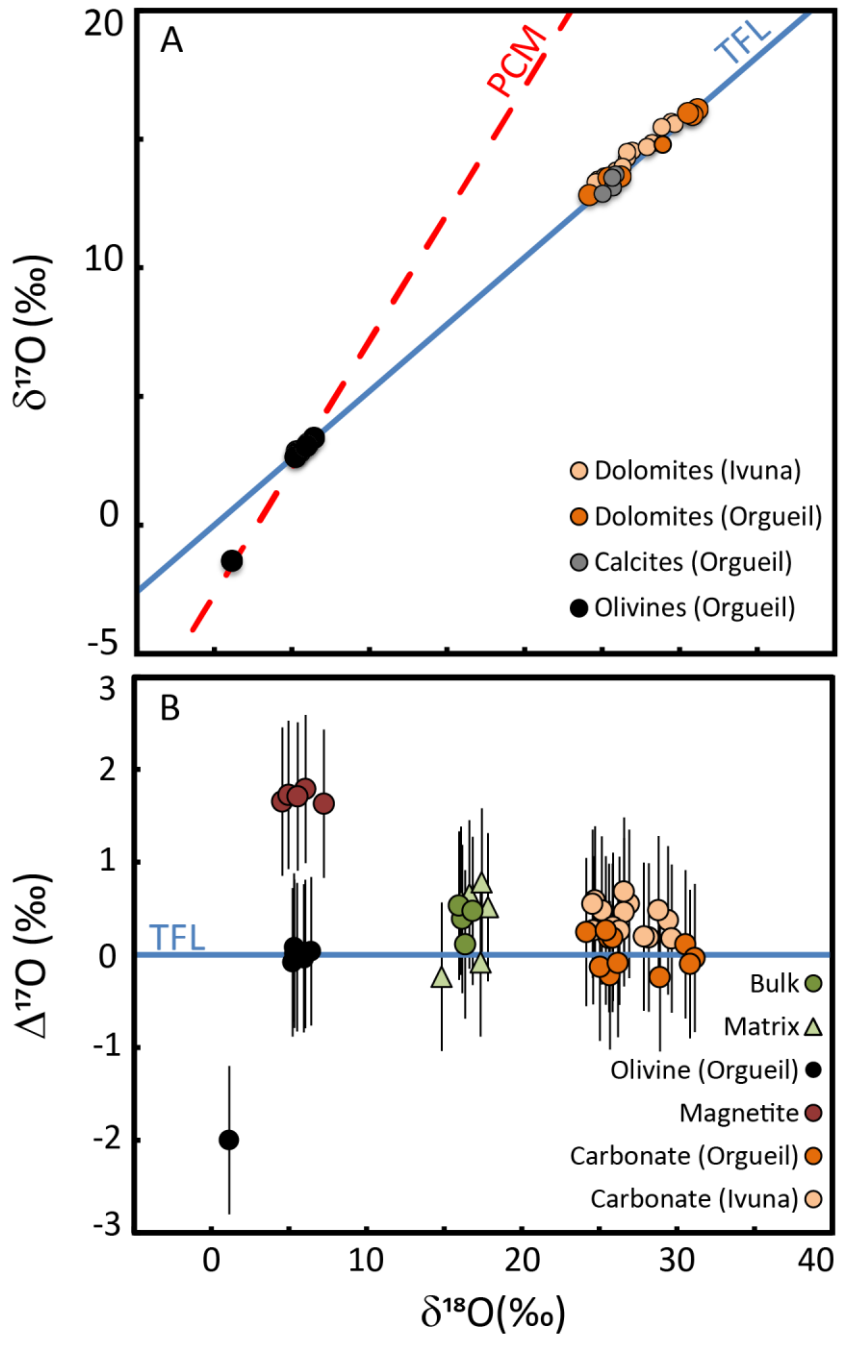
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Fig. 2

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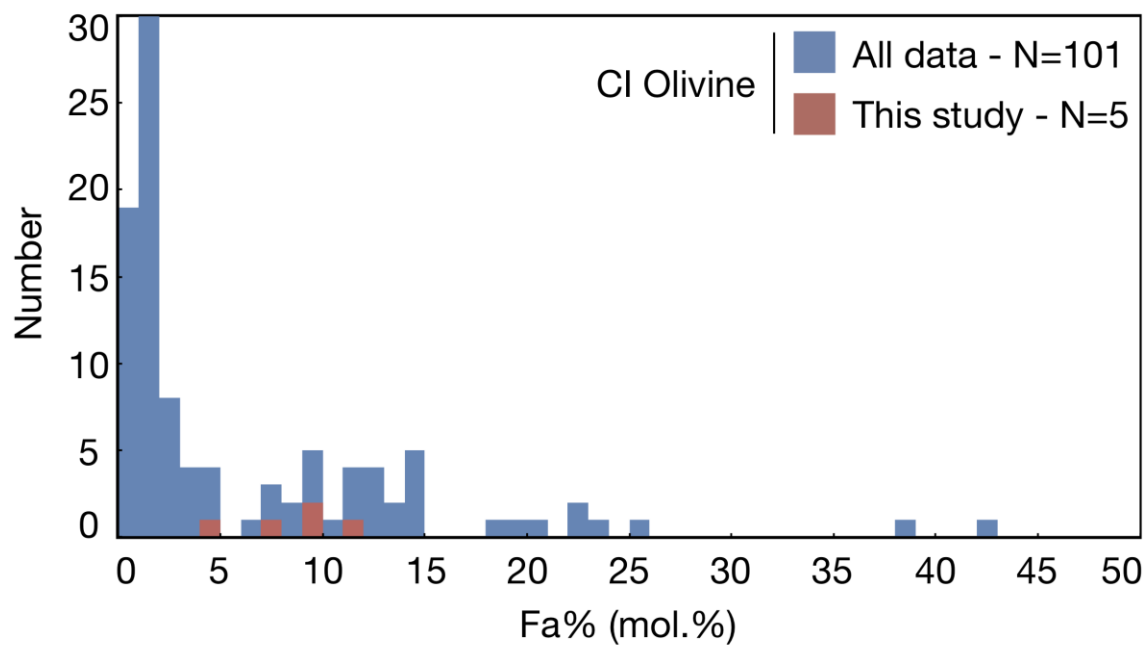


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Fig. 3



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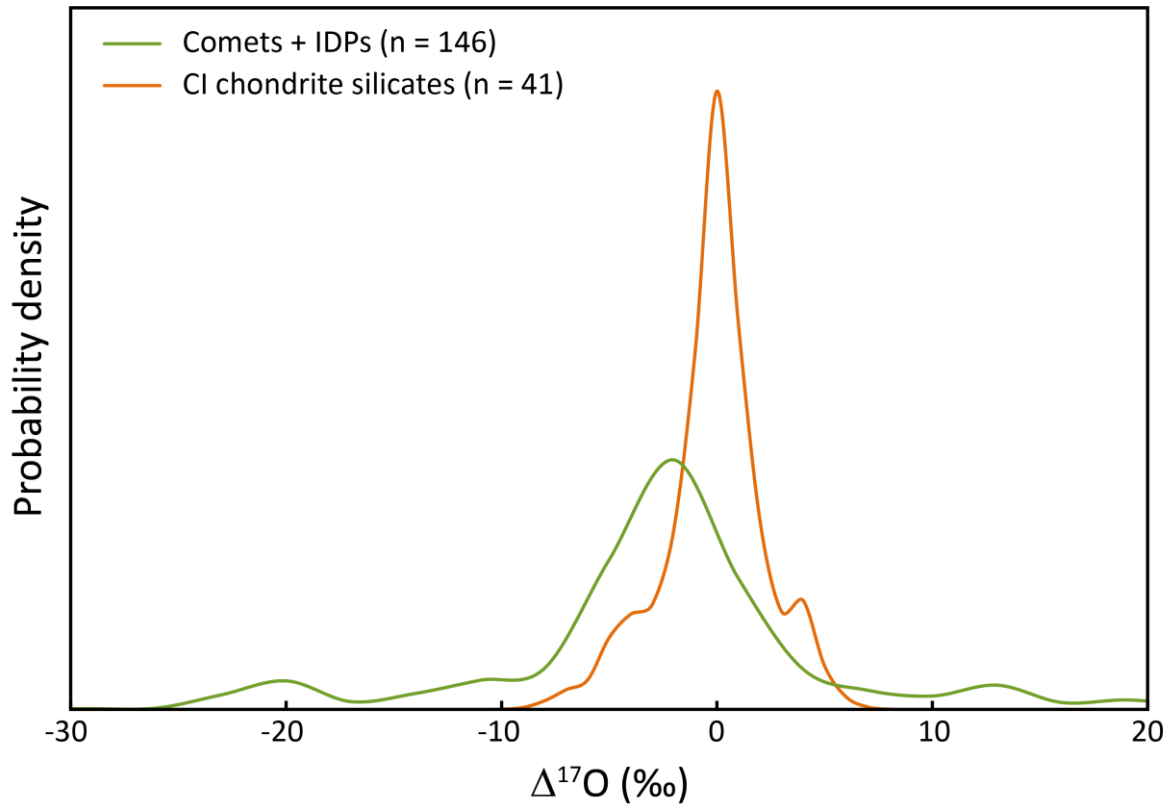
Fig. 4

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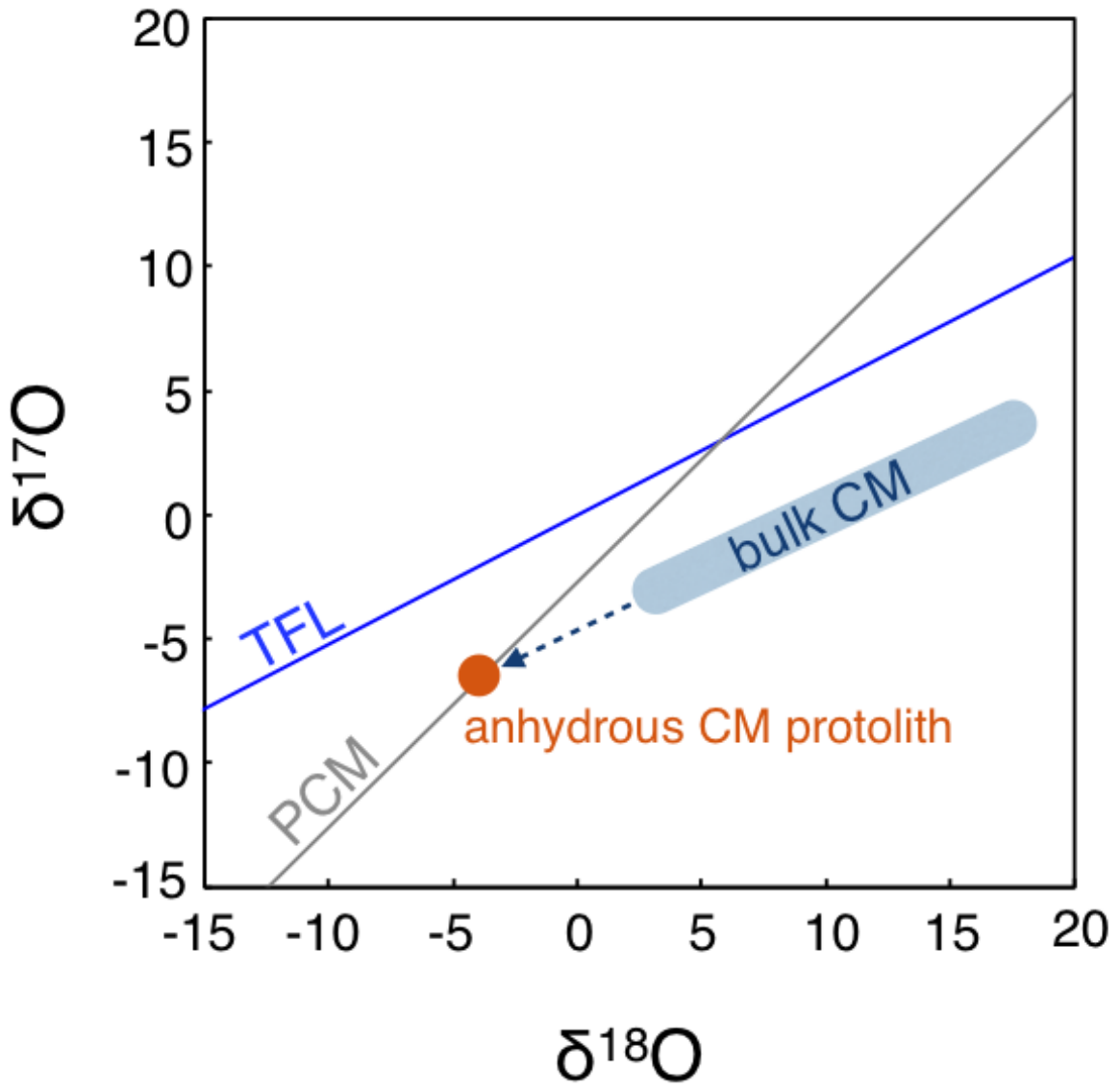
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Fig. 5



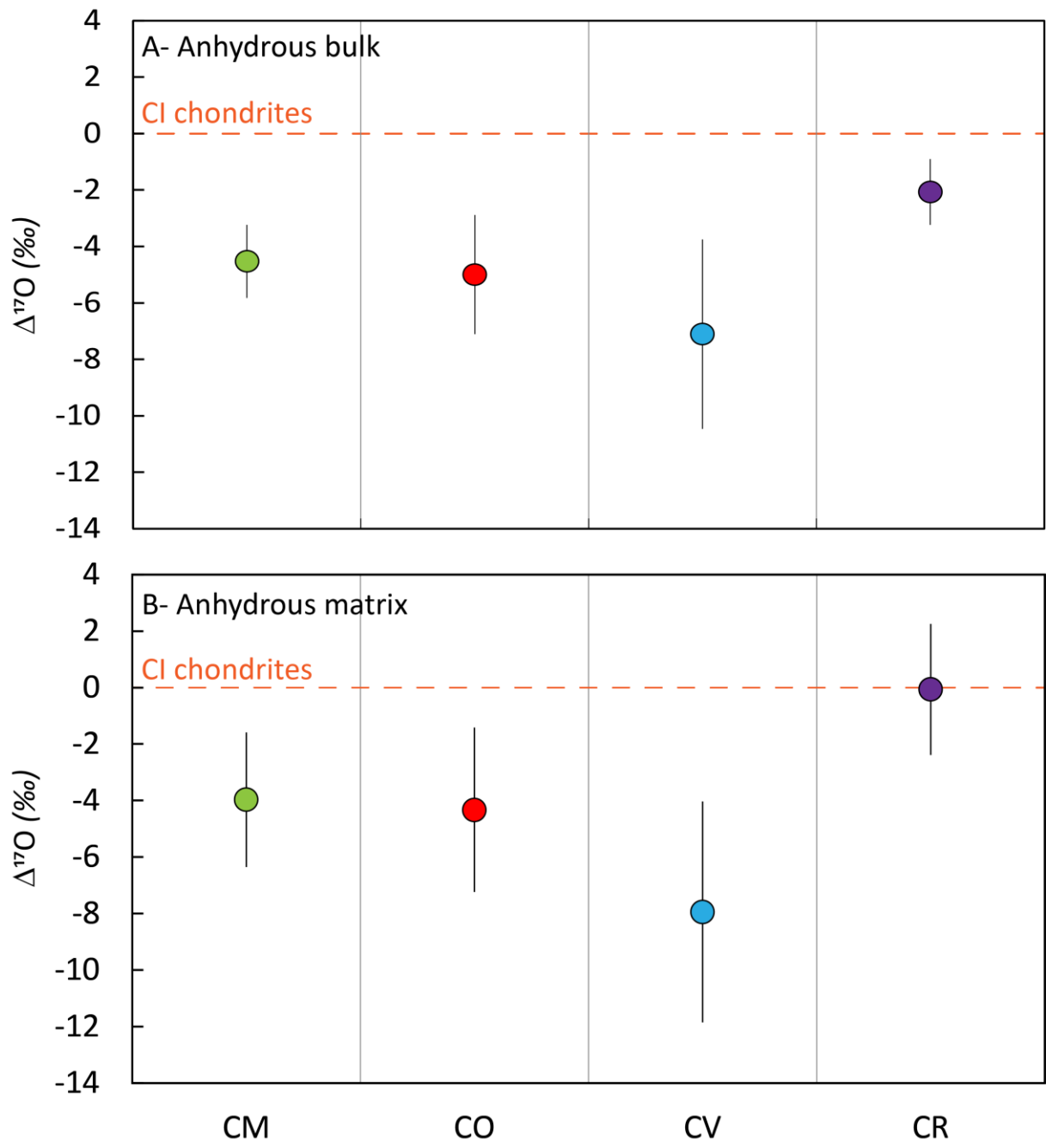
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Fig. 6

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Fig. 7

Meteorite	Mineralogy	Sample	SiO ₂	MgO	FeO	Al ₂ O ₃	CaO	Cr ₂ O ₃	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 ₂ O	SiO ₂	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	$\delta^{18}\text{O}$	2σ	$\delta^{17}\text{O}$	2σ	$\Delta^{17}\text{O}$	2σ
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).

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		$\delta^{18}\text{O}$	2σ	$\delta^{17}\text{O}$	2σ	$\Delta^{17}\text{O}$	2σ
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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