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1 **Isotopic evolution of the inner solar system revealed by**
2 **size-dependent oxygen isotopic variations in chondrules**

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19 **Abstract**

20
21 The systematic isotopic difference between refractory inclusions and chondrules,
22 particularly for oxygen, has long indicated an isotopic evolution of the solar protoplanetary
23 disk. However, it remains underconstrained whether such evolution continued during the
24 chondrule formation epoch. Intrigued by past reports of the size-dependent oxygen isotopic
25 compositions of chondrules in ordinary chondrites (OC), we analyzed type I olivine-rich
26 chondrules of various sizes in two LL3 chondrites. Although most chondrules show positive
27 $\Delta^{17}\text{O}$ values comparable to bulk ordinary chondrites, a population of smaller (less than about
28 0.1 mm² in cross-section), including many isolated olivine grains (*sensu lato*), are ¹⁶O-
29 enriched (with $\Delta^{17}\text{O}$ values down to -13.2 ‰). Literature data allow the same observation for
30 R chondrites. All sub-TFL type I chondrules (i.e., $\Delta^{17}\text{O} < 0$) chondrules have Mg# > 97 mol%
31 while the supra-TFL type I chondrule olivines extend to the formal boundary with type II
32 chondrules (i.e., Mg# = 90 mol%). The sub-TFL chondrules are likely genetically related to
33 isotopically similar aluminum-rich chondrules described in the literature. They therefore must
34 have formed earlier than most OC and R chondrules when the inner disk was still sub-TFL.
35 This interpretation is supported by the presence of similarly ¹⁶O-rich relict grains in supra-

36 TFL OC and R chondrules that must be remains of this incompletely recycled precursor
37 material. The non-carbonaceous reservoir was thus still evolving isotopically towards ^{16}O -
38 poorer composition when chondrule formation began, whether by mixing with outer disk
39 material, late accretion streamers and/or an increase in the solid/gas ratio due to
40 magnetothermal disk winds.

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43 **Keywords:** chondrules, oxygen isotopes, protoplanetary disk, dust dynamic, relict olivines

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1- Introduction

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84 The solar protoplanetary disk (PPD) surrounded the Sun for a few million years (e.g.,
85 Weiss et al. 2021) and was the site of an efficient production of submillimeter- to centimeter-
86 scale solids that subsequently agglomerated into planetary bodies. The initial isotopic
87 composition of the PPD was recorded by early refractory condensates in the form of calcium-
88 aluminum-rich inclusions (CAIs) and ameboid olivine aggregates (AOAs). Their
89 characterization has revealed that the disk was initially enriched in oxygen-16 ($\Delta^{17}\text{O} = \delta^{17}\text{O} -$
90 $0.52 \times \delta^{17}\text{O} < -20 \text{ ‰}$; Krot, 2019; Marrocchi et al., 2019a) and neutron-rich *r*-process-
91 derived nuclides (e.g., ^{48}Ca , ^{50}Ti , ^{54}Cr , and Mo and Ni isotopes; Trinquier et al., 2007, 2009;
92 Burkhardt et al., 2021, Jansen et al., 2024, Torrano et al., 2024). Although CAIs and AOAs are
93 generally considered to have condensed within 1 astronomical unit (AU, the semimajor axis
94 of Earth's orbit, $\sim 150 \times 10^6$ km; Jacquet, 2019; Bekaert et al. 2021), both are mostly observed
95 in carbonaceous chondrites (CCs), whose parent bodies likely accreted in the outer solar
96 system (Kruijer et al., 2017). This has been attributed to viscous expansion of the disk (e.g.,
97 Jacquet et al., 2011; Pignatale et al., 2019) or a dust trap outside a Jupiter-carved gap (Desch
98 et al., 2018; Jongejan et al., 2023). Conversely, non-carbonaceous chondrites (NCs), thought
99 to have accreted in the inner solar system, are notably depleted in refractory inclusions
100 (Haugbølle et al., 2019; Dunham et al., 2023). Therefore, the isotopic compositions of CCs
101 were more influenced by early-formed solids during disk evolution (Warren, 2011; Bryson
102 and Brennecka, 2021; Marrocchi et al., 2023), yet also evidence a shift during PPD evolution.

103

104 Chondrules are submillimeter-scale igneous spheroids mostly composed of silicate
105 minerals, glassy mesostases, Fe-Ni metal beads and sulfides. They represent the most

106 abundant high-temperature material in all types of chondrites, implying that both the inner
107 and outer solar system were affected by their formation. Although the timing of the onset of
108 chondrule formation remains debated (Bollard et al., 2017; Fukuda et al., 2022; Piralla et al.,
109 2023), all chronometers indicate formation processes spanning several million years.
110 Chondrules thus represent powerful proxies for quantifying the temporal isotopic evolution of
111 the disk.

112 Recent characterizations of individual chondrules have revealed drastic differences
113 between NC and CC chondrules: NC chondrules have homogeneous $\epsilon^{50}\text{Ti}$ and $\epsilon^{54}\text{Cr}$ values
114 similar to those of their host chondrites (despite exceptions in EH3 chondrite Sahara 97096,
115 Zhu et al., 2020), whereas CC chondrules have more variable values that sometimes greatly
116 differ from those of their host chondrites while also being systematically lower than those of
117 refractory inclusions (Gerber et al., 2017; Schneider et al., 2020). Furthermore, ^{16}O -rich relict
118 olivine grains inherited from chondrule precursors are more abundant in CC chondrules
119 (outside the CRHB clan; Marrocchi et al. 2022) than in their OC or R counterparts (Tenner et
120 al., 2018; Marrocchi et al., 2018, 2019b; Regnault et al., 2022). This multi-isotope approach
121 indicates that CC chondrules must have derived from the recycling of nebular condensates
122 (Marrocchi et al., 2018, 2019b; Ebert et al., 2018, Williams et al., 2020), not from NC
123 chondrule precursors as concluded from single-element isotopic data (Olsen et al., 2016; van
124 Kooten et al., 2021). These results also imply that the isotopic composition of the disk
125 changed rapidly after CAI and AOA condensation, settling towards values close to those
126 sampled by NC chondrules (Brennecka et al., 2020). This could have occurred by intra-disk
127 mixing (e.g., Jacquet et al., 2019) and/or the late infall of isotopically distinct material from
128 the surrounding molecular cloud (Nanne et al., 2019; Burkhardt et al., 2019, 2021, Jansen et
129 al., 2024). In the latter scenario, the varied isotopic compositions of CC chondrules would
130 represent a mixture between (i) a CAI-AOA-like dust component recording the initial disk

131 composition and (ii) NC-like dust inherited from late infalling material from the molecular
132 cloud (Schneider et al., 2020).

133 Compared to CC chondrules, NC chondrules show a narrower range of
134 nucleosynthetic anomalies, suggesting they formed from different precursors and/or under
135 different conditions (Schneider et al., 2020). Although less abundant than in CC chondrules,
136 relict olivine grains with chemical signature similar to those in AOAs are observed in ordinary
137 chondrites (OCs, the most NCs; Kita et al., 2010; Piralla et al., 2021). This suggests that the
138 NC chondrules could have originated from AOA-like material that condensed after the
139 isotopic composition of the disk had shifted toward NC values (Brennecka et al., 2020; Piralla
140 et al., 2021). The preservation of early-formed refractory inclusions in the inner disk is not
141 trivial, however, as they would have either been lost to the Sun or rapidly accreted into
142 planetesimals (Spitzer et al., 2020). However, such early planetesimals would have melted
143 due to ^{26}Al decay, homogenizing their isotopic compositions and erasing any record of early
144 ^{16}O -rich material. The chondrule isotopic variability is not limited to rare relicts isotopically
145 similar to AOAs. Less ^{16}O -rich relict grains also occur and some OC chondrules are ^{16}O -
146 enriched relative to NC bulks (Kita et al., 2010; Piralla et al., 2021). In addition, separated
147 chondrules in unequilibrated OCs show systematically decreasing $\Delta^{17}\text{O}$ values with
148 decreasing chondrule size (Clayton et al., 1991). Therefore, the inner disk was certainly
149 populated by isotopically diverse dust populations that escaped sunward drift and melting in
150 planetesimals.

151 To better understand the nature of OC chondrule precursors, we selected variably sized
152 type I porphyritic olivine (PO) chondrules from the pristine OCs Piancaldoli (LL3.10;
153 Marrocchi et al., 2020) and NWA 5206 (LL3.05; Pape et al., 2019). We report a
154 comprehensive study of chondrule olivine grains combining petrological observations and
155 quantitative geochemical analyses. Based on our results, we discuss (i) the oxygen isotopic

156 variability of OC chondrule precursors, (ii) the conditions of OC chondrule formation, and
157 (iii) implications for dust dynamics and evolution in the inner solar system.

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159 **2- Material & methods**

160

161 We surveyed all type I chondrules and isolated olivine grains (IOGs; Jacquet et al.,
162 2021) in two sections of the OC LL3.1 Piancaldoli: thin section USNM 5649 from the
163 National Museum of Natural History, Smithsonian Institution (Washington D.C., USA) and a
164 thick section prepared at the Centre de Recherches Pétrographiques et Géochimiques (CRPG-
165 CNRS, Nancy, France) from a parent sample provided by the Dipartimento scienze della Terra
166 e del Mare (Palermo, Italy). We also examined three thick sections of OC LL3.05 NWA 5206
167 that were prepared in-house. Chondrules were examined microscopically under transmitted
168 and reflected light. Among all porphyritic type I chondrules examined, we selected 47 olivine-
169 rich chondrules and IOGs with crystals large enough for major, minor, and trace element as
170 well as O isotopic analyses. The exposed surface areas of the chondrules were measured using
171 ImageJ (Schindelin et al., 2012, Pinto et al., 2021).

172 Scanning electron microscope observations were performed at CRPG-CNRS using a
173 JEOL JSM-6510 with 3 nA primary beam at 15 kV. We also performed multi-element EDX
174 mapping (Mg, Si, Fe, Ni, S, Na, Ca, and Al) of selected chondrules (Figs 1–3). Quantitative
175 point analyses were performed at the CRPG-CNRS using a JEOL JXA-8230 electron
176 microprobe analyzer (EPMA) equipped with five wavelength-dispersive spectrometers
177 (WDS) and one silicon drift detector energy dispersive spectrometer. Quantitative analyses of
178 Al, Ti, Ca, Cr, Mn, Ni, Mg, Fe, and Si in all olivine grains large enough to be isotopically
179 characterized by SIMS were performed with an accelerating voltage of 20 kV, a probe current
180 of 10 nA and beam diameter of 1 μm . **We used different standards for tuning the EPMA:**

181 Springwater olivine (Mg, Si), fayalite (Fe), wollastonite (Ca), albite (Na, Al), rutile (Ti), Ni
182 metal (Ni), chromite (Cr) and rhodochrosite (Mn). The total peak + background counting time
183 was 200 ms for Al, Ti, Ca, Mn and Cr, and 20 ms for Mg, Fe and Si. Detection limits were
184 0.025 wt% (Mg), 0.025 wt% (Fe), 0.05 wt% (Si), 0.005 wt% (Ca), 0.02 wt% (Al), 0.005 wt%
185 (Ti), 0.015 wt% (Cr), and 0.008 wt% (Mn).

186 The oxygen isotopic compositions were measured by secondary ion mass spectrometry
187 (SIMS) using a CAMECA IMS 1270 E7 at CRPG-CNRS (Bouden et al., 2021; Morin et al.,
188 2022). $^{16}\text{O}^-$, $^{17}\text{O}^-$, and $^{18}\text{O}^-$ ions produced by a Cs^+ primary ion beam ($\sim 4 \mu\text{m}$, 500 pA) were
189 measured in multi-collection mode using two off-axis Faraday cups (FCs) for $^{16,18}\text{O}^-$ and the
190 axial electron multiplier (EM) for $^{17}\text{O}^-$. To remove $^{16}\text{OH}^-$ interference on the $^{17}\text{O}^-$ peak and
191 achieve maximum flatness atop the $^{16}\text{O}^-$ and $^{18}\text{O}^-$ peaks, the entrance and exit slits of the
192 central EM were adjusted to achieve a mass resolving power ($\text{MRP} = M/\Delta M$) of $\sim 6,000$ for
193 $^{17}\text{O}^-$. The OH contribution was negligible ($< 0.1 \%$) and the ^{16}OH interference was well
194 resolved. The multi-collection FCs were set on exit slit 1 ($\text{MRP} = 2,500$). The total
195 measurement duration was 260 s, comprising 60 s of pre-sputtering and 200 s of
196 measurement. Three terrestrial standard materials (San Carlos olivine, Ipanko spinel, and
197 Rockport fayalite) were used to define the instrumental mass fractionation (IMF) line for the
198 three oxygen isotopes and correct for IMF due to matrix effects in olivine. To monitor any
199 instrumental drift and to achieve good precision, the San Carlos olivine was reanalyzed after
200 every 10 sample analyses. Oxygen isotopic compositions are expressed in δ notation as
201 $\delta^{17,18}\text{O} = ([^{17,18}\text{O}/^{16}\text{O}]_{\text{sample}}/[^{17,18}\text{O}/^{16}\text{O}]_{\text{V-SMOW}} - 1) \times 1,000\%$, where V-SMOW refers to the
202 Vienna Standard Mean Ocean Water. Samples related by mass-dependent fractionation to the
203 V-SMOW composition plot along a line with a slope of 0.52, defining the terrestrial
204 fractionation line (TFL), whereas mass-independent variations are described by $\Delta^{17}\text{O} = \delta^{17}\text{O} -$
205 $0.52 \times \delta^{18}\text{O}$, representing vertical deviations from the TFL in a triple oxygen isotope diagram.

206 Typical 2σ uncertainties, accounting for internal errors on each measurement and the external
207 reproducibility of the standard, were estimated to be $\sim 0.4\text{‰}$ for $\delta^{18}\text{O}$, $\sim 0.7\text{‰}$ for $\delta^{17}\text{O}$, and
208 $\sim 0.8\text{‰}$ for $\Delta^{17}\text{O}$. The error on $\Delta^{17}\text{O}$ was calculated by quadratically summing the errors on
209 $\delta^{17}\text{O}$ and $\delta^{18}\text{O}$. All SIMS analytical spots were checked thoroughly by SEM, and any spots
210 near fractures, in the mesostasis, or not completely within olivine grains were excluded from
211 the data set (see Supplementary Materials).

212

213 **3- Results**

214

215 The 47 selected type I PO chondrules and IOGs have surface areas ranging from 0.005
216 to 0.87 mm^2 . All contained euhedral to rounded olivine crystals with varying sizes in the
217 range 15–75 μm (Figs. 1–3). Many of the smaller chondrules were IOGs in the broader sense
218 of [Jacquet et al. \(2021\)](#), i.e., their olivines were dominated by a single crystal, even though
219 they may also have contained other phases such as glassy mesostasis (Figs. 1–2). Larger *bona*
220 *fide* chondrules (i.e., non-IOG) show palissadic textures (e.g., Libourel and Portail, 2018;
221 Marrocchi et al., 2019b) with larger olivine grains occurring in the chondrule edges and
222 smaller olivine crystals in chondrule interiors (Fig. 3). Punctual analyses **in olivine** show large
223 variations in minor and trace elements, with Al_2O_3 ranging from 0.01 to 0.54 wt%, Cr_2O_3
224 from 0.03 to 1.01 wt%, MnO from 0.01 to 0.66 wt%, CaO from 0.03 to 0.81 wt% and TiO_2
225 from 0.01 to 0.16 wt% (Table S1). The Mg# ($=\text{Mg}/(\text{Mg} + \text{Fe})$ in mol%) of olivine grains
226 range from 89.4 to 99.5 mol% (Table S1).

227 Olivine O-isotopic compositions vary widely, with $\delta^{18}\text{O}$ and $\delta^{17}\text{O}$ values ranging from
228 -19‰ to 8.4‰ and from -23.4‰ to 6.1‰ , respectively ([Figs 4–5, S1–S41](#)). Our set of
229 data defines two isotopic populations that plot parallel to either the TFL (Fig. 5, average $\Delta^{17}\text{O}$
230 $= 0.8 \pm 0.6\text{‰}$) or the Primary Chondrule Minerals **line**, (Fig.5; Ushikubo et al., 2012; $\Delta^{17}\text{O} =$

231 -3.3 ± 2.8 ‰). Hereafter, we will refer to these populations as the *supra-TFL* and *sub-TFL*
232 populations, respectively. Individual chondrules generally showed uniform $\Delta^{17}\text{O}$, consistent
233 with the paucity of isotopically identifiable relicts reported by Piralla et al. (2021) in their
234 LL3 chondrites. **The smallest supra-TFL chondrules (in 2D) show the isotopically lightest**
235 **mean compositions; possibly they were sectioned close to their surface as Piralla et al. (2021)**
236 **reported the isotopically heaviest spots to be concentrated in chondrule interiors.** However,
237 half (6/12) of the sub-TFL chondrules show standard deviations (largely) beyond the
238 analytical precision (Fig. 6).

239 Our data define orthogonal relationships between $\Delta^{17}\text{O}$ and Mg# (Fig. 6A), as well as
240 minor and trace element concentrations (Figs. 6B–F). This trend is somewhat reminiscent of
241 the anticorrelation between $\Delta^{17}\text{O}$ and Mg# in CC chondrules (e.g., **Chaumard et al., 2021,**
242 **Tenner et al., 2015, 2018 and references therein**); here, however, the trend is not monotonic,
243 but rather the concatenation of two trends, one vertical and one horizontal, **with no Mg# gap**
244 **(Fig. 6).** All sub-TFL (i.e., $\Delta^{17}\text{O} < 0$) chondrules are restricted to Mg# > 97 mol% but span a
245 large range of $\Delta^{17}\text{O}$ values, whereas supra-TFL type I chondrule olivines span only a small
246 range of $\Delta^{17}\text{O}$ values but extend to the formal boundary with type II chondrules at Fo₉₀ (i.e.,
247 Mg# = 90 mol%; Fig. 6A). **This is in line with the relative Mn and Cr enrichment of supra-**
248 **TFL olivine, since these elements correlate positively with Fa content in chondrules in OCs**
249 **and elsewhere (e.g., Jacquet et al. 2015a).** The highly magnesian compositions of the sub-TFL
250 chondrules are consistent with the observation of Pack et al. (2004) that 10 of 18 refractory
251 forsterites (cathodoluminescent, with Mg# > 98.3 mol%) in unequilibrated OCs (UOCs) were
252 sub-TFL.

253 Average $\Delta^{17}\text{O}$ values per chondrule also correlate with chondrule size; small
254 chondrules (especially below about 0.1 mm² in cross section) tend to be more enriched in ¹⁶O
255 than larger chondrules (Fig. 7; Table S1). This is also true of literature data: we measured the

256 exposed areas of chondrules shown in published figures for both ordinary (Pack et al., 2004;
257 Kita et al., 2010; Piralla et al., 2021; Ebert et al., 2022; Fig. 8) and Rumuruti chondrites (Pack
258 et al., 2004; Regnault et al., 2022; Fig. 9), including Al-rich chondrules (Russell et al., 1996,
259 1998; Ebert et al., 2022). These results are consistent with the systematic decrease of $\Delta^{17}\text{O}$
260 values with **decreasing chondrule size** reported by Clayton et al. (1991) in H chondrites,
261 although $\Delta^{17}\text{O}$ values remained positive for their finest size fractions (100-250 μm or 100-300
262 μm , about 0.01-0.1 mm^2). Presumably, supra-TFL compositions remain dominant **even for**
263 **such size fractions** (Fig. 7B).

264 Texturally, 4 out of our 12 sub-TFL chondrules are IOGs (Figs. 1,2); this is also true
265 for 4 of the 14 (9 if considering only those with at least two analyses) sub-TFL chondrules of
266 the extensive OC study of Baeza (2019). Earlier, Saxton et al. (1998) reported 5 sub-TFL
267 compositions out of 7 coarse forsterites (IOG *sensu lato*) in the Julesburg L3.6 chondrite,
268 consistent with the overrepresentation of IOGs among sub-TFL compositions.

269

270 **4- Discussion**

271

272 **4.1 ^{16}O -rich OC chondrules: witnesses of a distinctive chondrule-forming reservoir**

273

274 The most striking finding of our study is the correlation between chondrule size and
275 oxygen isotopic composition (Figs 7–9). This leads us to consider two main types of
276 chondrules: (i) sub-TFL chondrules (i.e., $\Delta^{17}\text{O} < 0 \text{ ‰}$) and (ii) supra-TFL chondrules (i.e.,
277 $\Delta^{17}\text{O} \geq 0 \text{ ‰}$). Straightforwardly, this correlation cannot be due to parent-body metamorphism
278 because the smallest chondrules, which are more susceptible to re-equilibration, are
279 isotopically furthest from the supra-TFL bulk OC chondrites/matrices (Clayton et al., 1991;
280 Greenwood et al., 2020). It has been first suggested that all OC chondrules underwent

281 exchange with an ^{16}O -rich gas, and that the larger chondrules lagged nearer to their assumed
282 (supra-TFL) starting isotopic composition due to their smaller surface/volume ratio (Clayton
283 et al., 1986). However, these chondrules show little internal variation in the $\Delta^{17}\text{O}$ values of
284 different phases (Kita et al., 2010), let alone olivine grains (with only 3 % of isotopically
285 identifiable relicts; Piralla et al., 2021). Although sub-TFL aluminum-rich chondrules (ARCs)
286 show isotopically distinctive mesostases, those mesostases are supra-TFL and were likely
287 affected by incipient aqueous alteration, whereas other minerals mostly show
288 indistinguishable $\Delta^{17}\text{O}$ values (Russell et al., 2000; Ebert et al., 2022). Therefore, there is no
289 evidence for the mass-independent isotopic evolution of chondrule melts during the
290 crystallization sequence of large supra-TFL chondrules. **Conversely, the mass-dependent**
291 **isotopic variations recorded by supra-TFL chondrules likely result from kinetic effects**
292 **induced by evaporation/recondensation processes during the gas–melt interactions, possibly**
293 **accumulated through successive recycling events (Piralla et al., 2021).**

294 The fact remains that half of our sub-TFL olivine-rich type I chondrules show
295 significant internal $\Delta^{17}\text{O}$ variations (Figs 4–8). The limited mass-dependent fractionation
296 (with $\Delta^{18}\text{O} = \delta^{18}\text{O} - \delta^{17}\text{O} = 2.9 \pm 1.0$ ‰, Table S1) shown by sub-TFL chondrule suggests the
297 heating timescale was much longer than the condensation timescale of oxygen, as maximum
298 kinetic fractionation of order $[\text{m}(\text{H}_2^{17}\text{O})/\text{m}(\text{H}_2^{16}\text{O})]^{1/2} - 1 = 27$ ‰/amu could be produced
299 (Cuzzi and Alexander, 2006). Consequently, the ambient gas should have significantly
300 exchanged with sub-TFL chondrule melts before the onset of the crystallization of their host
301 olivine grains.

302 **The kinetics of oxygen isotopic exchange between CAI-like melts and O-bearing gas**
303 **appear dominated by water vapor (Yamamoto et al., 2021, 2022). Using the 90 % exchange**
304 **timescales reported for a 1 cm diameter molten CAI at 1540 °C (Yamamoto et al., 2021), we**
305 **can correct for the size dependence for sub-mm molten chondrules (if we consider CAI melts**

306 as suitable analogs of dry basaltic melts; Yamamoto et al. 2021). Beyond a critical water
307 partial pressure (proportional to size) of ~ 0.1 Pa, the exchange timescale of a 0.5 mm
308 diameter droplet (an upper bound for chondrules in this study) should be diffusion-limited at
309 ~ 0.1 h (proportional to the square of the size). This is shorter than the day-scale cooling times
310 estimated for type I chondrules (e.g., Jacquet et al. 2015a). Although the partial pressure of
311 water in the OC chondrule-forming regions is not known, it appears that (i) the ratio to the
312 total (H_2 -dominated) pressure should be roughly enhanced by two orders of magnitude (e.g.,
313 Tenner et al. 2015) above the solar ratio ($\sim 10^{-3}$; Yamamoto et al. 2021) and (ii) the total
314 pressure would be ~ 1.4 Pa at 1 AU for a Minimum Mass Solar Nebula (Hayashi 1981). Thus,
315 a value near the aforementioned critical value of ~ 0.1 Pa seems likely.

316 The host olivine crystals of sub-TFL chondrules thus record the average O-isotopic
317 composition of their native reservoir, as do larger supra-TFL chondrules. This also implies
318 that isotopically deviant grains in sub-TFL chondrules must be considered relicts from
319 chondrule precursors, as are isotopically distinct grains in more classical NC and CC
320 chondrules (e.g., Ruzicka et al., 2007; Ushikubo et al., 2012; Schrader et al., 2013; Tenner et
321 al., 2013; Marrocchi et al., 2018, 2019b, 2022; Piralla et al., 2021; Schnuriger et al., 2022).
322 However, the relict grains reported herein are not as ^{16}O -rich as AOAs (i.e., $\Delta^{17}O < -20\%$;
323 Krot et al., 2004), instead forming a cluster of only relatively ^{16}O -rich compositions (Figs. 6–
324 9). This indicates that the relicts in question must be relicts of pre-existing chondrules of the
325 same general ^{16}O -rich reservoir (or, depending on how one delineates a reservoir, the set of
326 ^{16}O -rich reservoirs sampled by sub-TFL chondrules). These ^{16}O -rich reservoir(s) must have
327 spatially and/or temporally varied in composition for relicts to be isotopically manifest in
328 recycled chondrules.

329 Therefore, we conclude that sub-TFL chondrules originated from a ^{16}O -rich reservoir,
330 spatio-temporally distinct from that which formed most supra-TFL OC chondrules. We note

331 that the relicts found by (Piralla et al., 2021) in large supra-TFL chondrules span the same
332 range of isotopic compositions and minor element abundances as the sub-TFL chondrules
333 identified herein (with two isotopically AOA-like exceptions; Fig. 10). These relicts thus
334 likely stem from preexisting sub-TFL chondrules. These must thus have been present in the
335 OC-forming reservoir not only upon agglomeration, but also during the chondrule-melting
336 events that produced larger supra-TFL chondrules. Because the reverse situation (*i.e.*, supra-
337 TFL relicts within sub-TFL chondrules) is not observed, the sub-TFL chondrule-forming
338 reservoir may have preceded the supra-TFL chondrule-forming (and accreting) one, consistent
339 with the chronology suggested by Clayton et al., 1991. This is reminiscent of our results on
340 CR chondrites supporting two generations of type I chondrules of distinct size distributions,
341 further suggesting an evolution toward ^{16}O -poorer compositions (as exhibited by larger
342 objects; Marrocchi et al., 2022). **While Al-Mg dating on sub-TFL chondrules could test this
343 inference, we found an initial $^{26}\text{Al}/^{27}\text{Al}$ estimate for only one such chondrule in the literature
344 (c27 in NWA 8649; Siron et al. 2022), which, with $(7.52 \pm 0.88) \times 10^{-6}$, fits in the cluster of
345 UOC chondrule ages, but its $\Delta^{17}\text{O} = -0.94 \text{ ‰}$ is only marginally in sub-TFL territory.**

346

347 **4.2 Significance of the small size of the *sub-TFL* chondrules**

348

349 Because sub-TFL chondrules originate from a different reservoir than most OC
350 chondrules, the difference in size compared to the latter may not be surprising as different
351 chondrite groups show distinctive average sizes (e.g., Jones, 2012; Friedrich et al., 2015).
352 However, it not obvious to what extent the observed size of these sub-TFL chondrules reflects
353 that of their native population, because their sizes may have been biased by transport
354 processes. Larger grains tend to drift faster than small grains (e.g., Jacquet et al., 2012);
355 therefore the present-day sub-TFL chondrules may represent only the small end of a broader

356 size distribution, which could have diffused more efficiently across reservoirs (or have been
357 depleted more slowly from a given evolving region of the disk).

358 As noted previously, we observed an overrepresentation of IOGs among the sub-TFL
359 population (this study, Saxton et al., 1998; Pack et al., 2004). It was recently proposed that
360 many IOGs showing evidence of interaction with a gas as single entities were expelled from
361 colliding partially molten chondrules (Jacquet et al., 2021). A natural consequence of this
362 process is to produce IOGs that are smaller than their parent chondrules (Jacquet et al., 2021).
363 Such collisions could also occasionally expel olivine-free melt fragments that would solidify
364 as spherical chondrules enriched in incompatible elements such as aluminum, calcium and
365 sodium. Interestingly, most OC ARCs have sub-TFL compositions with similar size
366 distributions and $\Delta^{17}\text{O}$ ranges as ferromagnesian sub-TFL chondrules (Fig. 8). Notably, **Ebert**
367 **and Bischoff (2016)** mentioned splashing as a possible formation mechanism of ARCs. They
368 nevertheless inferred that ARC had CAI-like precursors, drawing some analogy from their
369 volatility-fractionated rare earth element patterns (Grossman et al., 1977, 1979). However, the
370 REE patterns of ARCs are similar to those of enstatite chondrule mesostases, which were
371 ascribed to igneous partitioning and are not diagnostic of CAIs (Jacquet et al., 2015b). In
372 addition, OC CAIs have ^{50}Ti -rich compositions distinct from those of ARCs (Ebert et al.,
373 2018). This lack of the hypothesized ARC CAI precursors is a serious issue given the
374 nonnegligible abundance of ARCs. The latter may indeed be very roughly estimated from the
375 (i) 30 ARCs in 15 OC sections studied by **Ebert and Bischoff (2016)** and (ii) an average
376 apparent area of 0.14 mm^2 (Ebert et al., 2022). Assuming 1 cm^2 total area for each section
377 (which will do for an order-of-magnitude estimate), this yields a modal abundance of $30 \times$
378 $0.14 \text{ mm}^2 / (15 \times 1 \text{ cm}^2) = 0.3 \%$. Assuming 90 % recycling (i.e., an upper limit of the
379 chondrule fraction in OCs) of these ARC-like CAIs exclusively in the form of ARCs, we
380 would still expect $\sim 0.03 \%$ of isotopically ARC-like CAIs. This estimate is comparable to the

381 total abundance of CAIs reported in OCs (0.0088 %; Dunham et al., 2023), of which
382 isotopically ARC-like ones must however be a very minor fraction, if at all extant.

383 Therefore, it is unlikely that CAI precursors can explain the composition of OC ARCs
384 (unlike for CCs; Ebert et al., 2018), whereas their crystallization from residual melts enriched
385 in incompatible elements is a much more generic process. We conversely hypothesize that
386 most sub-TFL chondrules, whether ferromagnesian or aluminum-rich, represent melt splashes
387 from partially molten droplets. Cold fragmentation is also conceivable because the correlation
388 between size and isotopic compositions holds for fragments (Clayton et al., 1991). Sub-TFL
389 chondrules may thus represent the smaller end of a broader size distribution of early-formed
390 ^{16}O -rich chondrules, unless collisions from which they formed destroyed all larger droplets in
391 their immediate region.

392 **It is noteworthy that** H chondrites have the smallest chondrules among UOCs, as well
393 as the lowest bulk $\Delta^{17}\text{O}$ ($0.69 \pm 0.07 \text{ ‰}$ vs. $0.96 \pm 0.11 \text{ ‰}$ in L chondrites and $1.08 \pm 0.13 \text{ ‰}$
394 in LL chondrites; Clayton et al., 1991). As already suggested by Clayton et al. (1991), this
395 may indicate a higher fraction (by a few percent) of small sub-TFL chondrules, resulting of
396 differential size sorting in the OC-forming reservoir. Our observations are limited to LL
397 chondrites, but we note that more abundant ARCs (mostly sub-TFL) were observed in H (22
398 in 8 thin sections) than in L and LL (8 in 7 thin sections) chondrites (Ebert and Bischoff,
399 2016), consistent with this expectation.

400

401 **4.3 A carbonaceous chondrite provenance?**

402

403 **As chondrules in carbonaceous chondrites are generally ^{16}O -rich compared to their NC**
404 **counterparts (e.g., Kita et al., 2010, Ushikubo et al. 2012; Tenner et al. 2015; Marrocchi et al.**
405 **2018, 2019, 2022, Piralla et al., 2021, Siron et al., 2022), this raises the question whether the**

406 sub-TFL OC chondrules formed in the outer CC reservoir. There is some potential evidence
407 for transport of NC chondrules to the C reservoir (e.g., Schrader et al. 2020; Williams et al.
408 2020), so migration in the other direction is conceivable. Al-Mg dating does not suggest CC
409 chondrule formation to predate OC chondrule formation (e.g., Fukuda et al., 2022, Piralla et
410 al. 2023), so there may be little prospect of significant drift of CC chondrules before the
411 formation of the main population (supra-TFL) of OC chondrules. Yet Zhu et al. (2020)
412 reported 2 chondrules with CC-like Cr isotopic composition out of 9 analyzed in EH3
413 chondrite Sahara 97096, although measurement of other isotopic systems would be desirable
414 to exclude the possibility of a “lost component” in the inner disk (Burkhardt et al., 2021).

415 A first objection stems from the abundance of refractory inclusions in carbonaceous
416 chondrites where a refractory inclusion/chondrule modal ratio ~ 0.1 is commonly reported
417 (e.g., Jacquet et al. 2016; Ebel et al. 2016). Consequently, if some chondrules immigrated
418 from the C reservoir, refractory inclusions must as well, especially since, being generally
419 smaller than chondrules (May et al. 1999), they should not be preferentially filtered out by
420 any size sorting, e.g. at a hypothetical pressure maximum (e.g., Haugbølle et al. 2019).
421 However, the 0.001 vol% CAIs reported in subtype 3.00-3.2 UOCs (Dunham et al., 2023)
422 provides an upper limit for the abundance of CAIs (+rare AOAs) coming from the C outer
423 disk (and having escaped chondrule melting) there. Thus, using the refractory
424 inclusion/chondrule ratio above, that of C-sourced chondrules cannot exceed ~ 0.01 vol%. The
425 summed area of sub-TFL chondrules found in the 5 sections of this study is 1.4 mm^2 and the
426 summed area of the said sections is 5.8 cm^2 , thus representing an abundance of at least 0.2
427 vol% for our LL chondrites. This is a conservative lower bound, for our survey was not
428 exhaustive (e.g., in being restricted to analyzable objects). Thus, sub-TFL chondrules appear
429 too numerous to be cogenetic with any CC provenance subset of OC refractory inclusions.

430 Piralla et al. (2021) argued against a C provenance of sub-TFL relicts found in OC
431 chondrules because their average $\Delta^{18}\text{O}$ was distinctly lower than CC host grains measured in
432 the same laboratory (2.1 vs. 3.4 ‰). Our sub-TFL chondrules exhibit an average $\Delta^{18}\text{O}$ of 2.9
433 ‰, which, although consistent with the previous results, is no longer conclusive.
434 Nevertheless, if we are right in associating sub-TFL ferromagnesian chondrules to sub-TFL
435 ARC, the clearly NC isotopic composition of the latter (Ebert et al. 2018) also ties them to the
436 NC superclan. We thus conclude that sub-TFL chondrules mostly formed in the NC reservoir,
437 at an earlier time than most OC chondrules, although measurements of nucleosynthetic
438 signatures in their ferromagnesian subset would be worthwhile.

439

440 **4.4 Thermal, chemical and isotopic dynamics of the inner disk**

441

442 The sub-TFL chondrules (including IOGs) studied herein commonly show **diverse** O
443 isotopic compositions (Figs. 5–7). We propose that these chondrules correspond to the
444 remains of a lost population of ^{16}O -rich large chondrules that initially populated the inner
445 disk. Today, they are largely under-represented in OCs probably due to the sunward drift of
446 most of their larger counterparts. Others were presumably recycled into supra-TFL
447 chondrules, as attested by the sub-TFL nature of their relict olivine grains (Piralla et al., 2021;
448 Fig. 10). This implies that recycling processes were highly efficient in the inner disk, as
449 evidenced by the large modal abundance of supra-TFL chondrules in OCs. However, our
450 study shows that some sub-TFL objects escaped remelting in supra-TFL environments,
451 implying that both OC and R chondrites are composed of several genetically related
452 chondrule populations formed at different times during the evolution of the inner disk.
453 Furthermore, this attests of the nebular origin of OC and R chondrules because large planetary
454 precursors (such as envisioned by Libourel and Krot 2007) would have been isotopically

455 homogenized by heat released from ^{26}Al -decay, which is inconsistent with the varied $\Delta^{17}\text{O}$
456 values observed in sub-TFL chondrules and IOGs (Fig. 9). Interestingly, similar
457 interpretations were recently proposed in the CC reservoirs based on (i) the chemical
458 characteristics and oxygen isotopic compositions of relict olivine grains in type I CC
459 chondrules (Marrocchi et al., 2018, 2019b), (ii) the size-dependent oxygen isotope variations
460 observed in CR chondrules (Marrocchi et al., 2022), (iii) the presence of small ^{16}O -rich
461 chondrule-like objects in the Ryugu samples (Nakashima et al., 2023) and (iv) the similar
462 $\Delta^{17}\text{O}$ values observed between type I olivine phenocrysts and Mg-rich relict olivine grains in
463 type II chondrules (Pinto et al., 2024). Altogether, **this** evidence shows that the nebular dust
464 has been constantly reprocessed and recorded the evolving isotopic composition of the disk.

465 Combined with literature data, our results have profound implications on our
466 understanding of the evolution of the inner solar system. Assuming our "splash" hypothesis
467 for the formation of OC ARCs, the lost generation of inner disk chondrules were not
468 characterized by ^{50}Ti excesses. This supports models in which the initially *r*-process-rich
469 composition of the inner disk changed rapidly after being recorded by CAI condensation
470 (Nanne et al., 2019; Brennecka et al., 2020). Our data also show that sub-TFL chondrules and
471 IOGs have higher Mg# compared to supra-TFL chondrules (Figs 6–8). This highlights that
472 the isotopic composition and the redox state of the inner disk was still evolving from initially
473 ^{16}O -rich compositions and reduced conditions toward ^{16}O -poor, more oxidizing conditions
474 during chondrule formation. This interpretation is in line with the high modal abundance of
475 supra-TFL type II chondrules in ordinary chondrites (Tenner et al., 2018; **Zanda et al. 2006**).
476 In this regard, the presence of both reduced and oxidized supra-TFL chondrules within the
477 same OC does not imply their formation in two distinct spatial reservoirs.

478 The identification of the sub-TFL chondrule population attests to the isotopic
479 evolution of the inner disk. Because these objects recorded the isotopic compositions of their

480 formation reservoirs, the loss of some of them (e.g. the largest) by sunward drift should not
481 have generated any isotopic changes. This therefore suggests the addition of ^{16}O -poor material
482 after the condensation of ^{16}O -rich refractory inclusions. Such an addition might correspond to
483 ^{16}O -poor water ice resulting from CO self-shielding processes, either in the outer disk or in
484 the molecular cloud (e.g., Young et al. 2008). This water ice may have been enriched in
485 deuterium (Jacquet and Robert, 2013), which might account for the high D/H ratios of
486 minimally altered UOCs (Piani et al., 2021; Grant et al., 2023). Interestingly, the average $\epsilon^{50}\text{Ti}$
487 of OC ARCs (i.e., -1.0 ± 0.1 , Ebert et al., 2018), which are mostly sub-TFL, is slightly lower
488 than the bulk compositions of OCs (i.e., -0.66 ; Burkhardt et al., 2021). This may suggest a
489 late ^{50}Ti -rich admixture into the inner disk, which might correspond to CI-like dust. Indeed,
490 this scenario has recently been proposed to account for (i) bulk meteorite compositions
491 (Bryson & Brennecka, 2021, Yap and Tissot, 2023) and (ii) the evolution from type I-CO to
492 type I-CR chondrules in CR chondrites (Marrocchi et al., 2022).

493 Whatever the exact nature of the admixture, several astrophysical source can be
494 envisioned, such as (i) late accretion streamers onto the disk (e.g., Kuznetsova et al., 2022) or
495 (ii) material from the outer disk, whether drifting/diffusing freely or filtered by a gap carved
496 by Jupiter, with an outer pressure bump preventing the arrival of the coarsest solids (e.g.,
497 Haugbølle et al., 2019). Still, the mere change in composition of the solid assemblage cannot
498 intrinsically explain the trend toward more oxidized chondrule formation conditions, unless
499 invoking water-rich admixtures. This trend could be related to the solid/gas ratio (Tenner et
500 al., 2015; Marrocchi and Chaussidon, 2015), which might be increased by photoevaporation
501 of the gas-rich surface layers of the disk (e.g., Pascucci et al. 2022) or magnetized disk winds
502 (e.g., Borlina et al., 2021). Paleomagnetic records indicate the dissipation of the inner disk by
503 ~ 4 Ma after CAI formation (e.g., Borlina et al., 2021); associated effects on the solid/gas ratio

504 may thus have been appreciable by the time the later supra-TFL generation of NC chondrules
505 formed.

506

507 **5- Conclusions**

508

509 We performed a petrographic, chemical and isotopic survey of type I porphyritic
510 olivine-rich chondrules of various sizes in the unequilibrated ordinary chondrites Piancaldoli
511 and NWA 5206. Our main results are:

512 1- Chondrules show large oxygen isotopic variations with $\delta^{18}\text{O}$ and $\delta^{17}\text{O}$ values
513 ranging from -19‰ to 8.4‰ and from -23.4‰ to 6.1‰ , respectively.

514 2- Among the studied chondrules, we observed two isotopic populations: small
515 chondrules with heterogeneous mass-independent isotopic variations characterized by $\Delta^{17}\text{O} <$
516 0‰ ($n = 12$) and their larger counterparts with homogeneous supra-TFL compositions (i.e.,
517 $\Delta^{17}\text{O} \geq 0\text{‰}$, $n = 35$).

518 3- Small chondrules have similar Mg# and minor element concentrations as relict
519 olivine grains present within large supra-TFL chondrules.

520 From these observations, we propose the following interpretations:

521 1- Small sub-TFL chondrules (including aluminum-rich chondrules) originate from a
522 ^{16}O -rich reservoir spatiotemporally distinct from that where supra-TFL OC chondrules
523 formed. Sub-TFL chondrules thus represent a lost generation of chondrules that predates the
524 formation of large supra-TFL chondrules.

525 2- Sub-TFL chondrules represent the small end of a broader size distribution of early-
526 formed ^{16}O -rich chondrules. Their small size might result from their formation as fragments
527 of collisions between partially molten ^{16}O -rich chondrules and/or differential loss by radial
528 drift of their larger cogenetic counterparts.

529 3- Large supra-TFL chondrules formed through the recycling of this sub-TFL
530 population in combination with gas-melt interactions.

531 4- The inner disk sampled by non-carbonaceous chondrites was still isotopically
532 evolving toward ^{16}O -poorer composition at the onset of chondrule formation This could have
533 resulted from the (i) injection of material from the molecular cloud or the outer disk or
534 (ii) enhancement of the solid/gas ratio due to magnetothermal disk winds.

535

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542

543 **Data Availability**

544 Original data from this study are available on the Ordar database:
545 <https://doi.org/10.24396/ORDAR-123>.

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Figure caption

862 **Fig 1:** (A, C, E) Backscattered electron images of small olivine-rich porphyritic chondrules in
863 chondrite NWA 5206. The average $\Delta^{17}\text{O}$ values of each chondrule are reported. (B, D, F)
864 Compiled EDX maps of Si and Mg in the same three PO chondrules, revealing the
865 mineralogy of the different chondrule silicate phases: olivines (orange), low-Ca pyroxenes
866 (pale green), and mesostasis (dark green).

867

868 **Fig 2:** (A, C, E) Backscattered electron images of isolated olivine grains (IOGs) and small
869 olivine-rich porphyritic chondrules in chondrites NWA 5206 and Piancaldoli. The average
870 $\Delta^{17}\text{O}$ values of each chondrule are reported. (B, D, F) Compiled EDX maps of Si and Mg in
871 the the IOG and two PO chondrules, revealing the mineralogy of the different chondrule
872 silicate phases: olivines (orange), low-Ca pyroxenes (pale green), and mesostasis (dark
873 green).

874

875 **Fig 3:** (A, C, E) Backscattered electron images of large olivine-rich porphyritic chondrules in
876 chondrite NWA 5206. The average $\Delta^{17}\text{O}$ values of each chondrule are reported. (B, D, F)
877 Compiled EDX maps of Si and Mg of PO chondrules, revealing the mineralogy of the
878 different chondrule silicate phases: olivines (orange), low-Ca pyroxenes (pale green), and
879 mesostasis (dark green).

880

881 **Fig 4:** Oxygen three-isotope diagrams for different type I chondrules of varying sizes in the
882 OC chondrites Piancaldoli and NWA 5206. Chondrules show (i) homogenous supra-TFL (A)
883 and sub-TFL (B) isotopic compositions, (ii) mass-independent sub-TFL isotopic variations (C
884 & D) and (iii) mass-dependent supra-TFL isotopic variations (E & F). The Terrestrial
885 Fractionation Line (TFL, blue line) and Primitive Chondrule Minerals line (PCM, red line)
886 are shown for reference. Errors are 2SD.

887 **Fig 5:** (A) Oxygen isotopic compositions of all olivine crystals measured in the 47 selected
888 chondrules and IOGs in chondrites NWA 5206 and Piancaldoli. (B) Oxygen isotopic
889 compositions of olivine grains in chondrules with average $\Delta^{17}\text{O} < 0\text{‰}$ (red circles) and those
890 with $\Delta^{17}\text{O} > 0\text{‰}$ (black circles). The former show mass-independent isotopic variations
891 whereas the latter display mass-dependent isotopic variations. The Terrestrial Fractionation
892 Line (TFL, blue line) and Primitive Chondrule Minerals line (PCM, red line) are shown for
893 reference. Errors are 2SD.

894

895 **Fig 6:** Oxygen isotopic compositions (expressed as $\Delta^{17}\text{O}$) reported as a function of (A) Mg#
896 and incompatible element concentrations: (B) Al, (C) Cr, (D) Mn, (E) Ca, and (F) Ti.
897 Chondrules with high Mg# and low concentrations of incompatible elements have variable,
898 and commonly negative, $\Delta^{17}\text{O}$ values.

899

900 **Fig 7:** Average oxygen isotopic compositions of chondrules in chondrites NWA 5206 and
901 Piancaldoli as functions of their (A) average Mg# and (B) surface areas.

902

903 **Fig 8:** Average $\Delta^{17}\text{O}$ values of chondrules as a function of their surface areas in chondrites
904 NWA 5206, Piancaldoli, Semarkona, Bishunpur, Chainpur, Dar al Gani 369, Dar al Gani 378,

905 Dar al Gani 327, and Dar al Gani. Data from this study, Russell et al., 1996, 1998; Pack et al.,
906 2004; Kita et al., 2010; Ebert et al., 2018; Piralla et al., 2021; [Siron et al. 2022](#).

907

908 **Fig 9:** Average $\Delta^{17}\text{O}$ values of chondrules as a function of their respective surface areas in
909 ordinary and Rumuruti chondrites. Data from this study, Pack et al., 2004; Kita et al., 2010;
910 Ebert et al., 2018; Piralla et al., 2021; Regnault et al., 2022.

911

912 **Fig 10:** Oxygen isotopic compositions (expressed as $\Delta^{17}\text{O}$) of *sub-TFL* (open black circles)
913 and *supra-TFL* (open red circles) chondrules reported as a function of (A) Mg# and
914 incompatible element concentrations: (B) Al, (C) Cr, (D) Mn, (E) Ca, and (F) Ti. Also
915 reported are host (open blue diamonds) and relict (open green diamonds) olivine grains from
916 larger OC chondrules (Piralla et al., 2021).

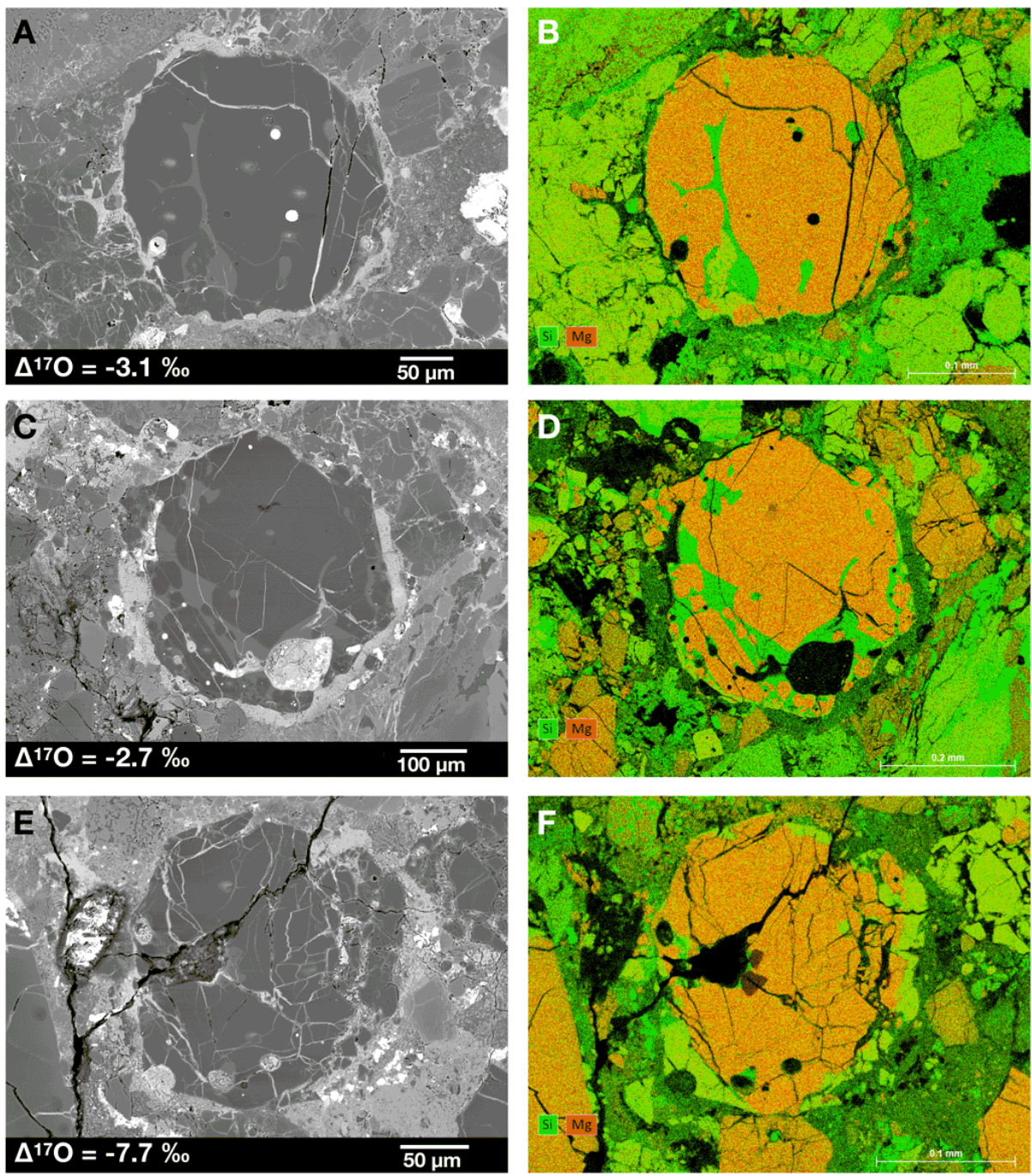


Fig. 1

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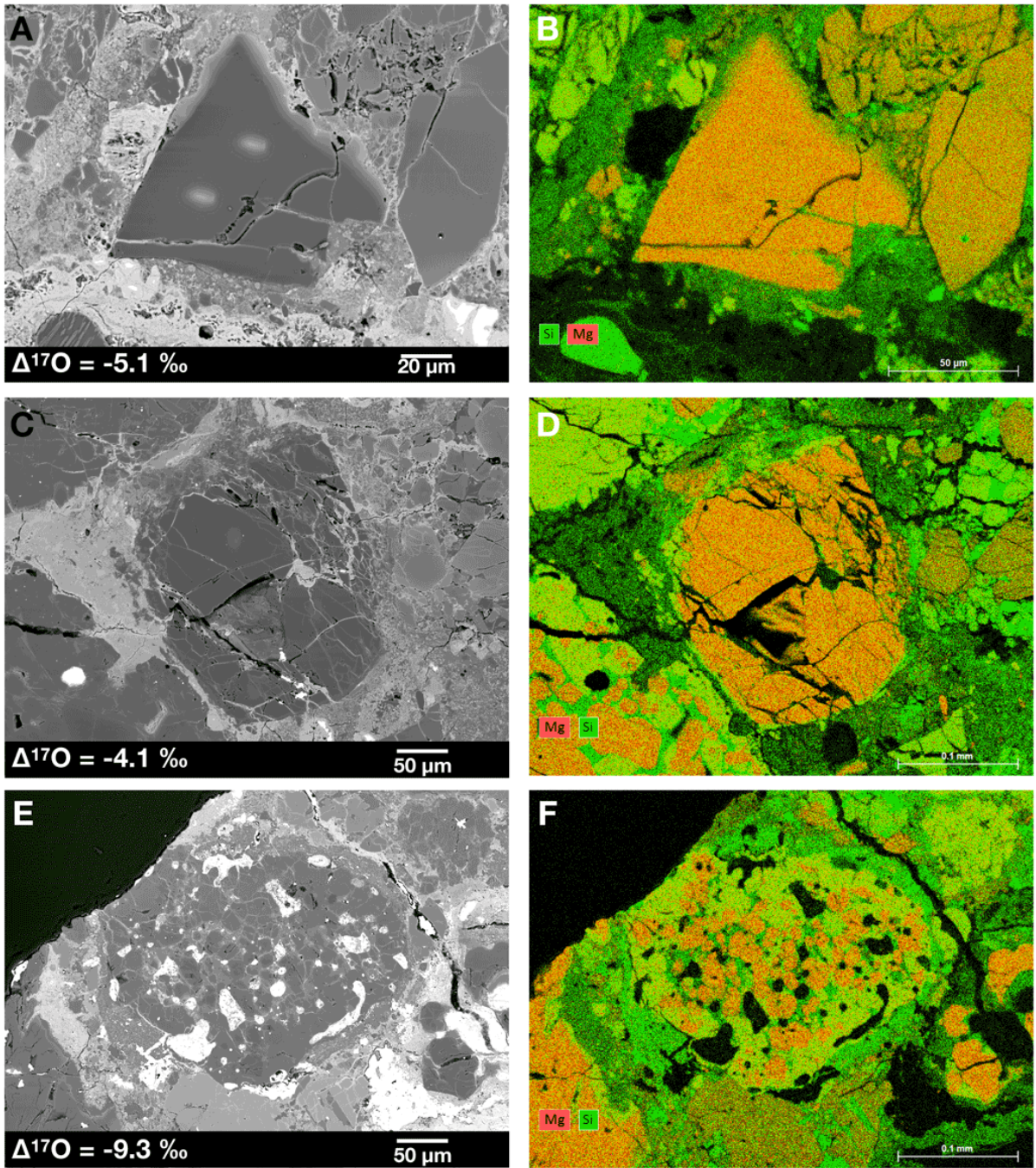
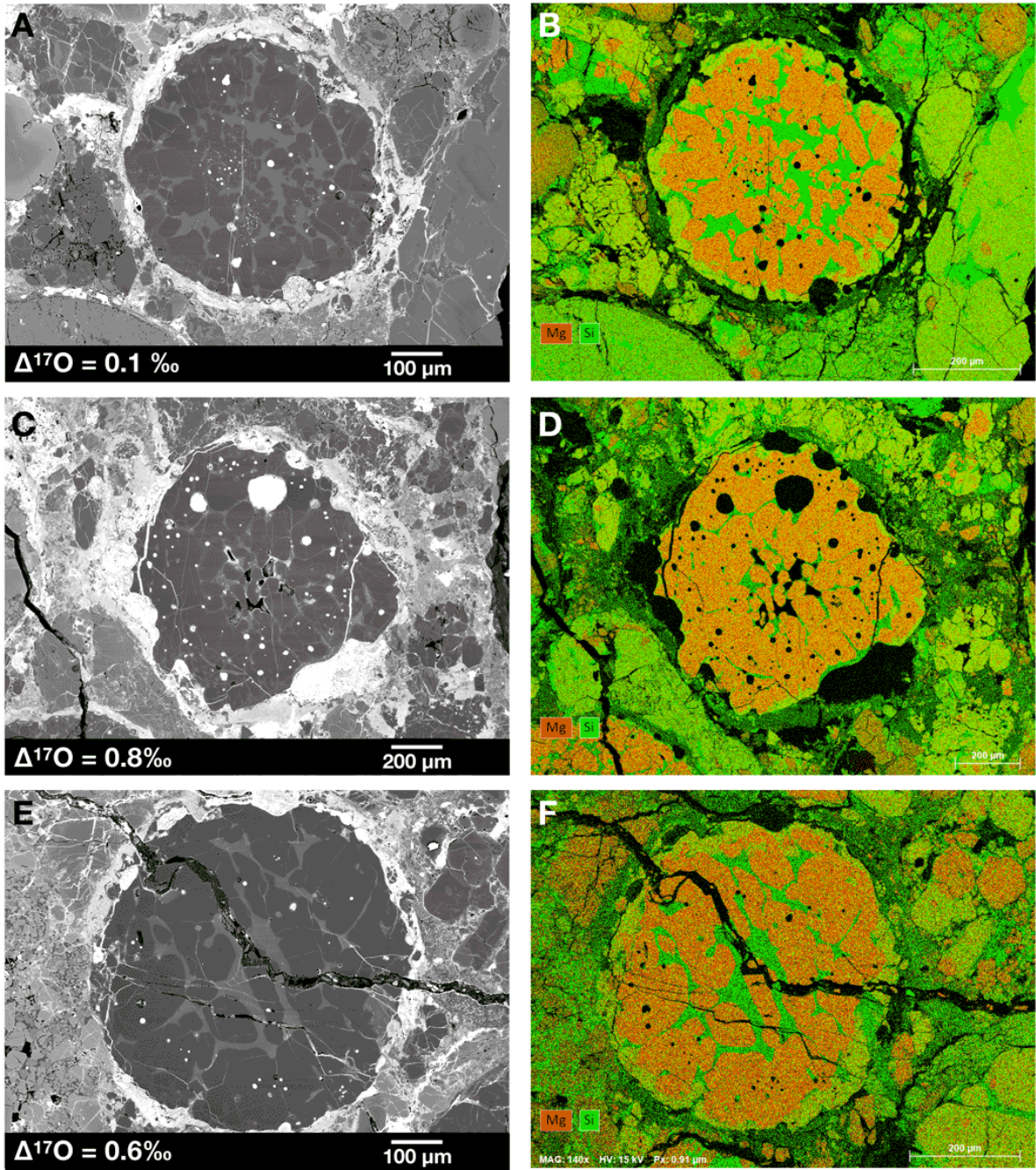


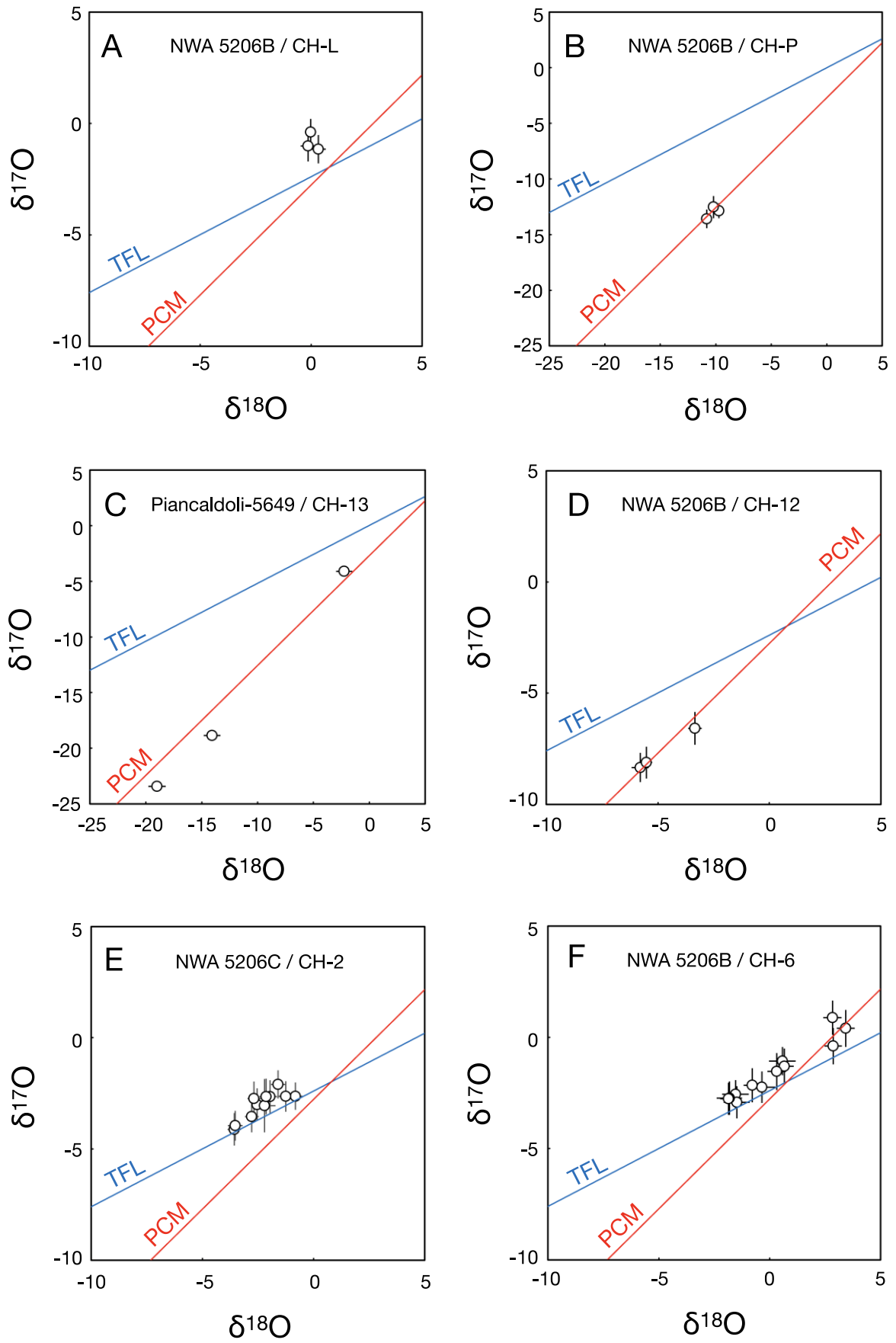
Fig. 2

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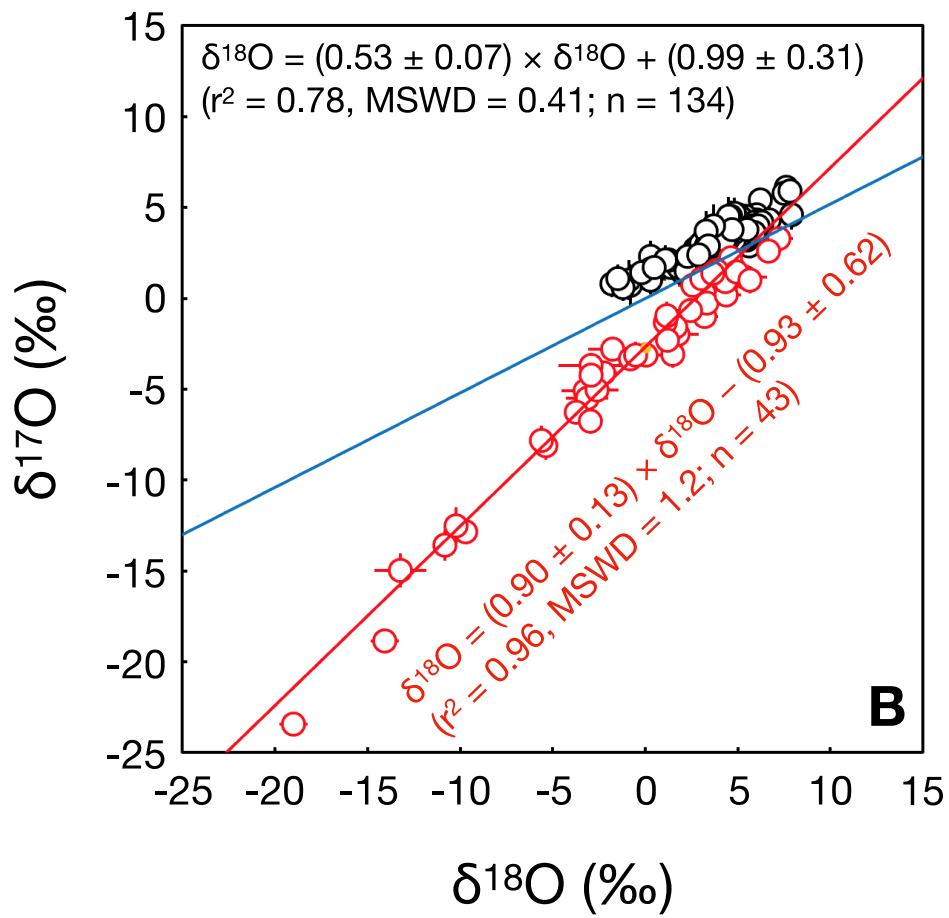
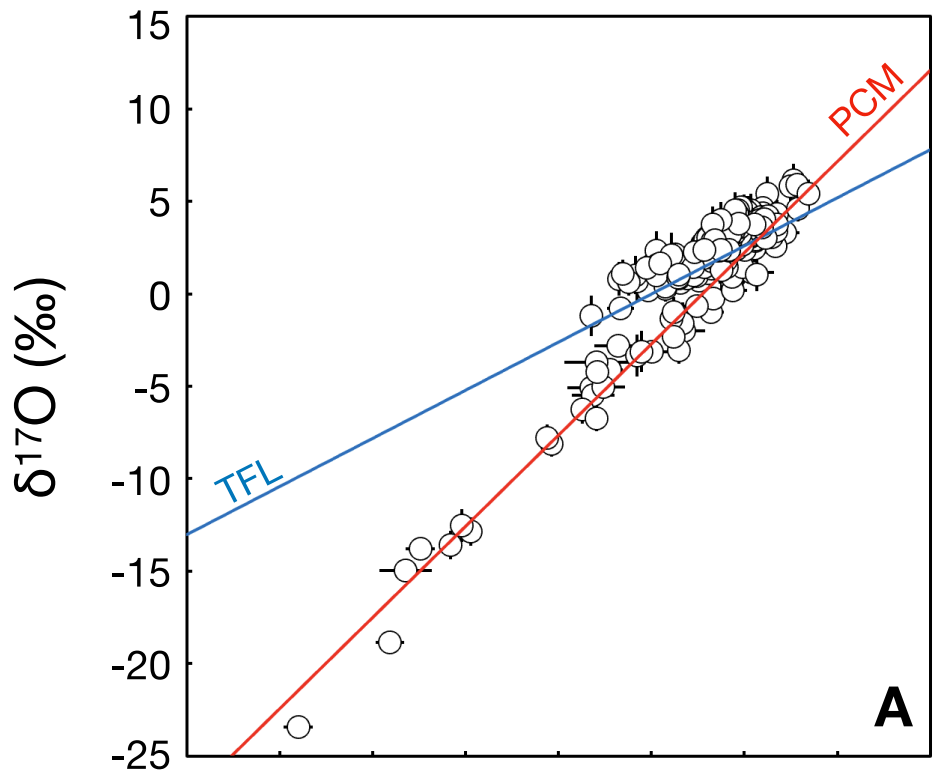
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Fig. 3



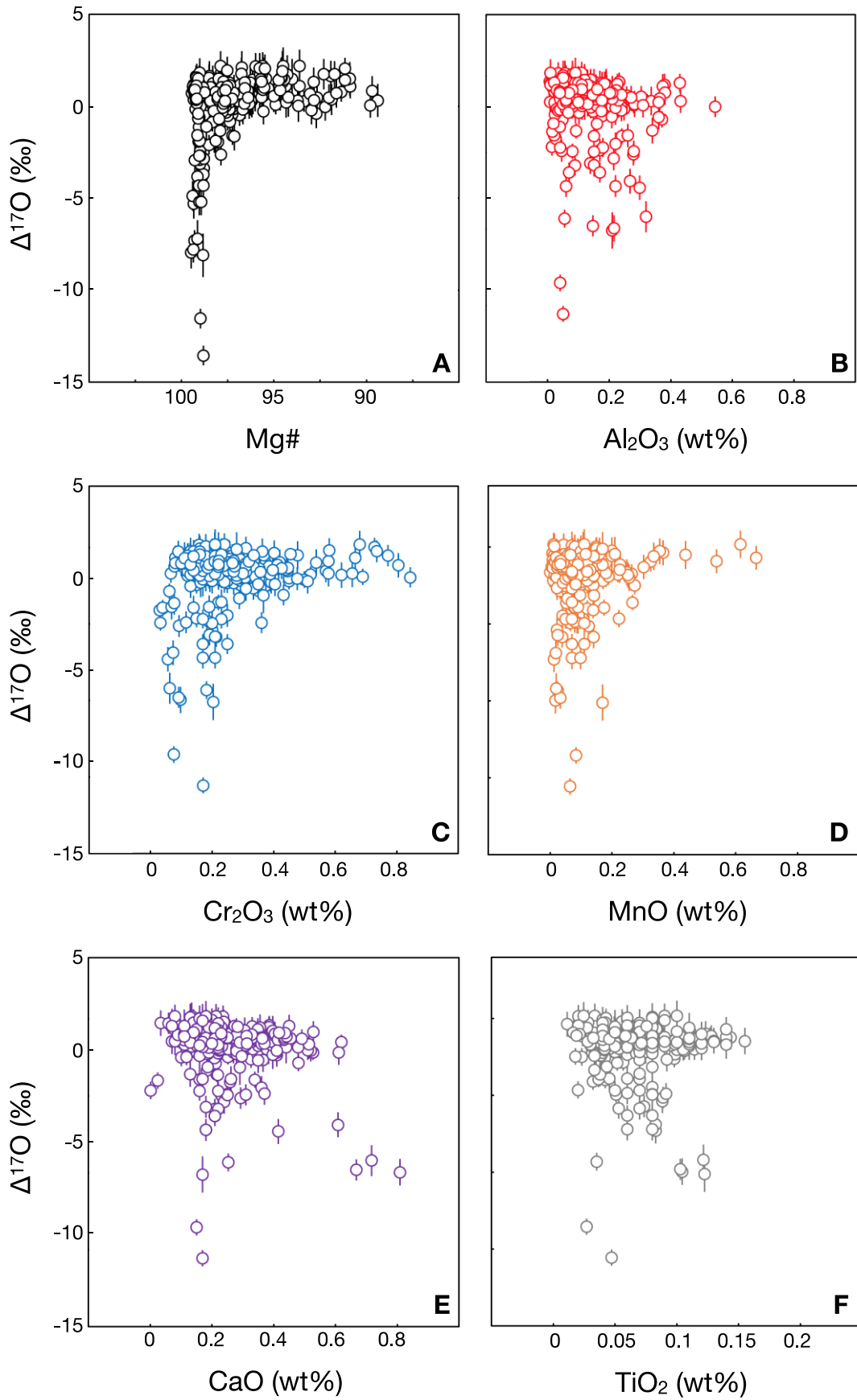
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Fig. 4



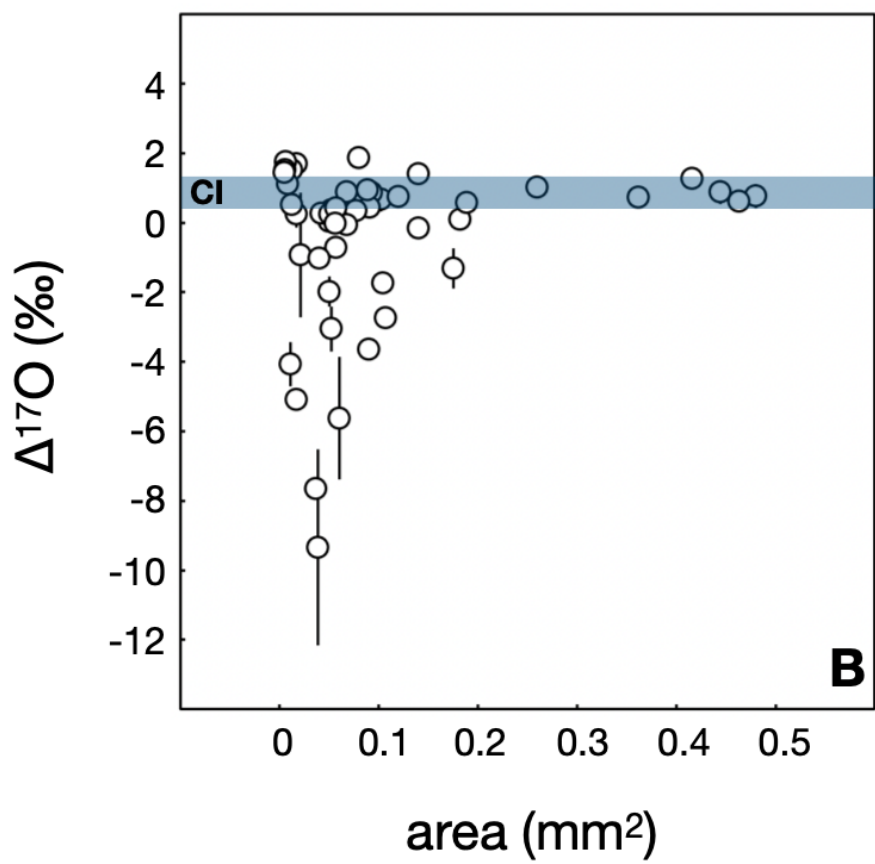
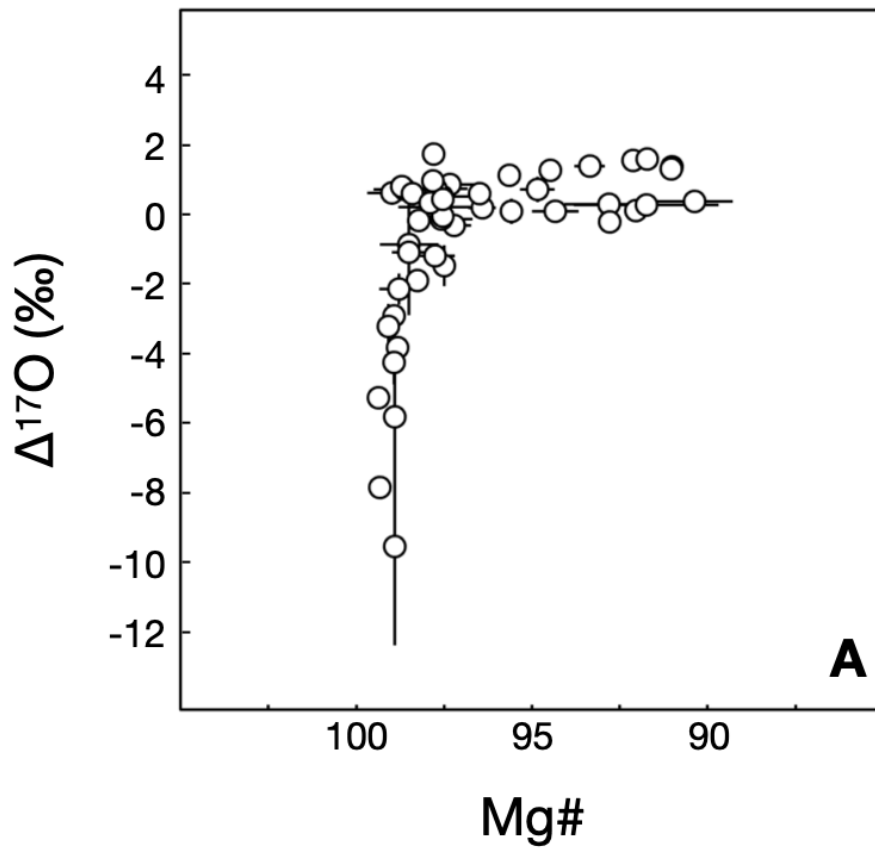
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Fig. 5



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Fig. 6



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Fig. 7

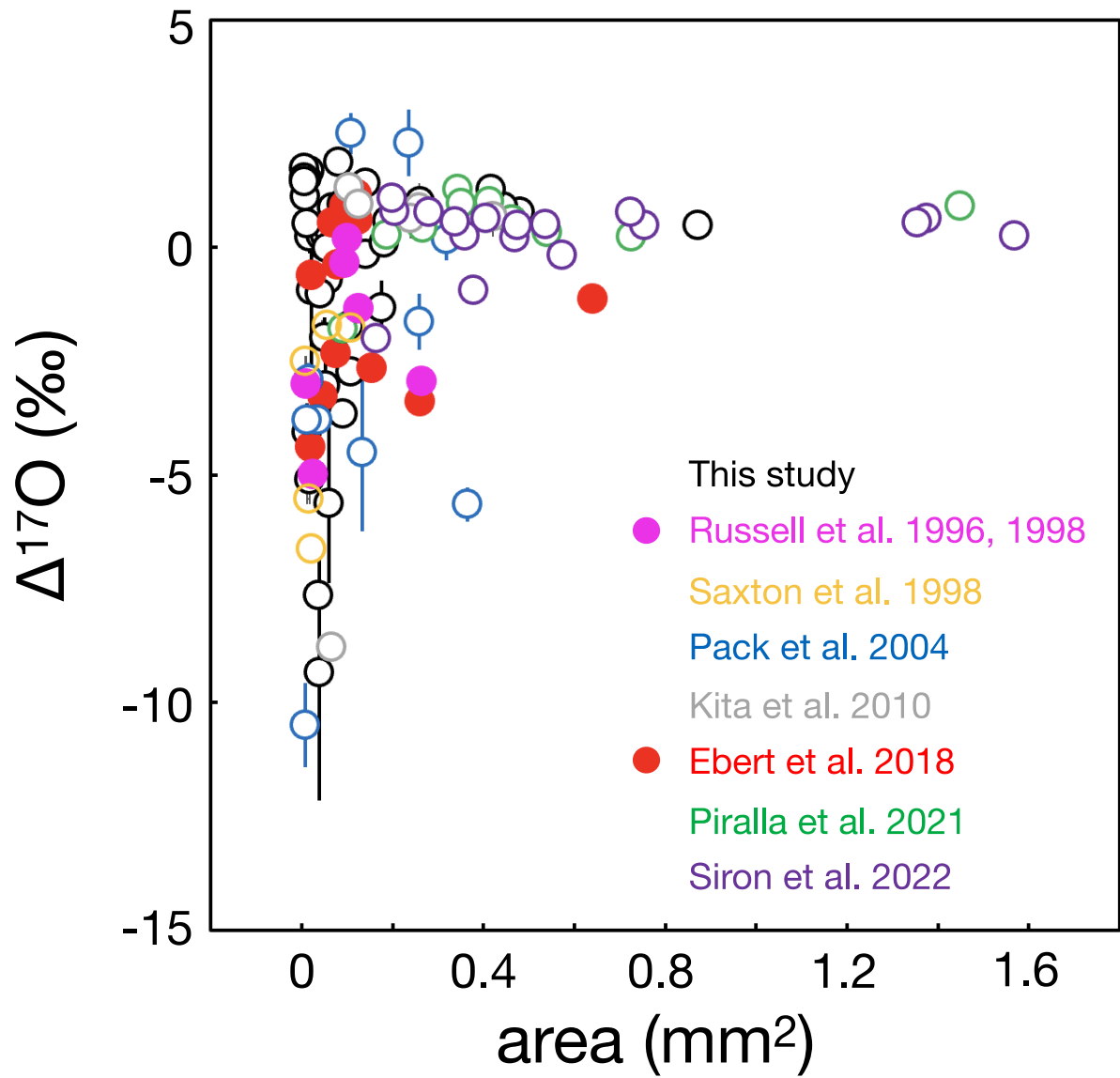
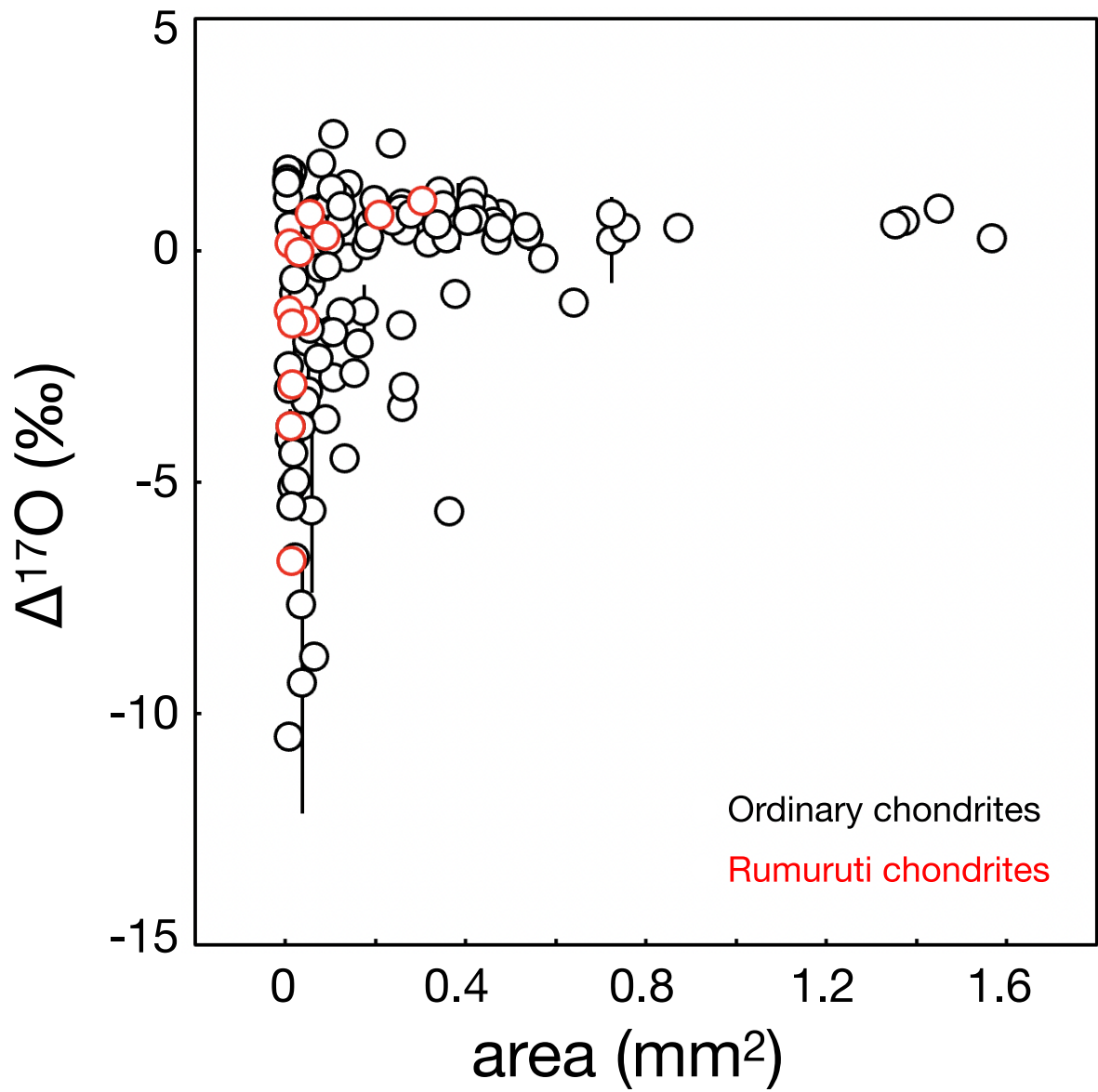


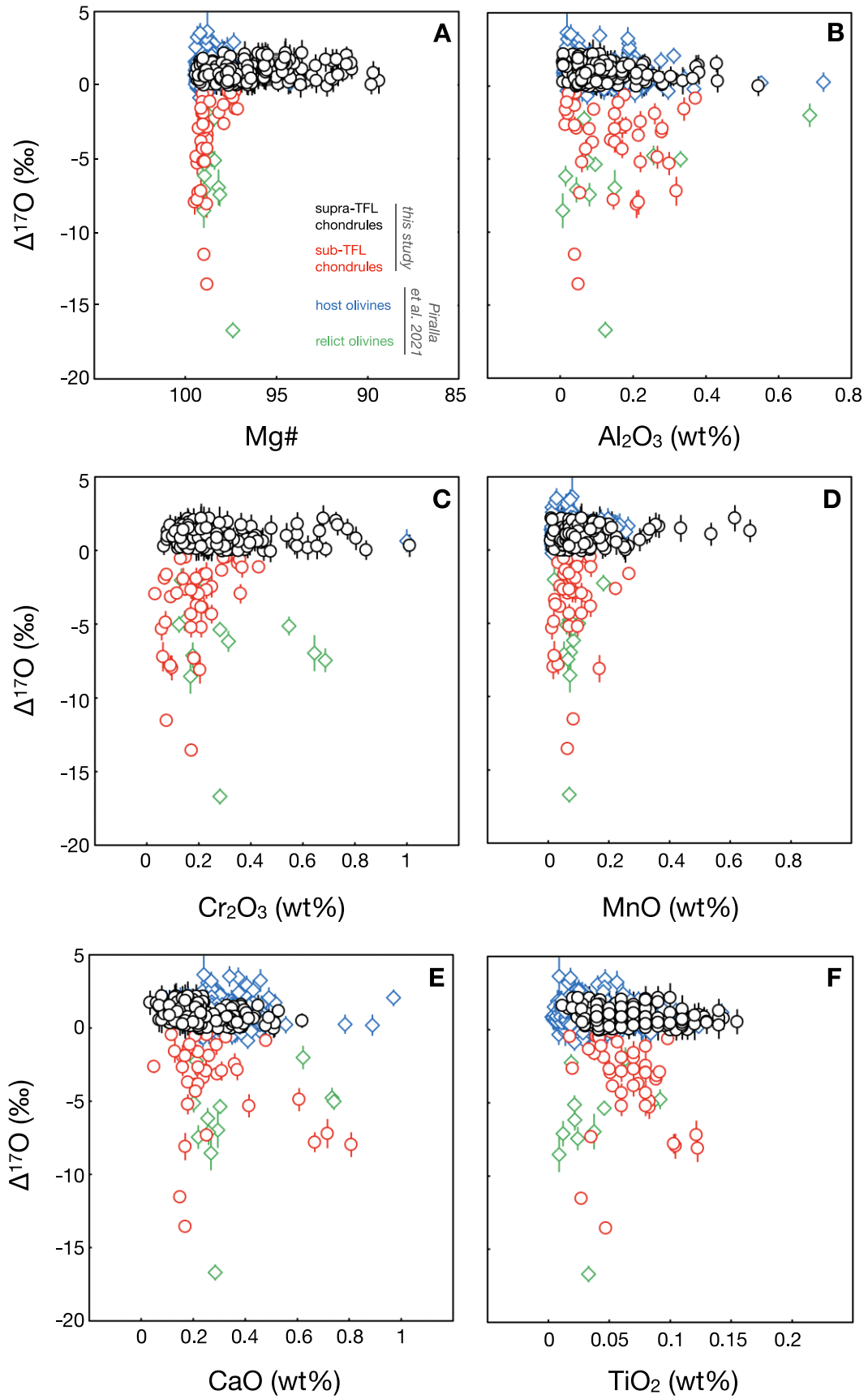
Fig. 8

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Fig. 9



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Fig. 10