



HAL
open science

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

Yves Marrocchi, Alizé Longeau, Rosa Lozano Goupil, Valentin Dijon, Gabriel Pinto, Julia Neukampf, Johan Villeneuve, Emmanuel Jacquet

► To cite this version:

Yves Marrocchi, Alizé Longeau, Rosa Lozano Goupil, Valentin Dijon, Gabriel Pinto, et al.. Isotopic evolution of the inner solar system revealed by size-dependent oxygen isotopic variations in chondrules. *Geochimica et Cosmochimica Acta*, 2024, 371, pp.52 - 64. 10.1016/j.gca.2024.03.001 . hal-04636887

HAL Id: hal-04636887

<https://hal.science/hal-04636887v1>

Submitted on 5 Jul 2024

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

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

3
4 Yves Marrocchi^{1,*}, Alizé Longeau¹, Rosa Lozano Goupil¹, Valentin Dijon¹,
5 Gabriel Pinto^{1,2,3}, Julia Neukampf¹, Johan Villeneuve¹ & Emmanuel Jacquet²

6
7 ¹Centre de Recherches Pétrographiques et Géochimiques (CRPG), CNRS, UMR 7358,
8 Nancy, France

9 ²Royal Belgian Institute of Natural Sciences, Geological Survey of Belgium, 1000, Brussels,
10 Belgium

11 ³Instituto de Ciencias de la Tierra, Universidad Austral de Chile, Valdivia, Chile.

12 ⁴Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie (IMPMC), Muséum
13 national d'Histoire naturelle, Sorbonne Université, CNRS, CP52,
14 57 rue Cuvier, 75005 Paris, France.

15
16 *corresponding author: yvesm@crpg.cnrs-nancy.fr

17
18
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.

41

42

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

44

45

46

47

48

49

50

51

52

53

54

55

56

57

58

59

60

61

62

63

64

65

66

67

68

69

70

71

72

73

74

75

76

77

78

79

80
81
82

1- Introduction

83

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.

158

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

536 **Acknowledgments**

537 We thank Nordine Bouden for help during sample preparation and isotopic analyses.
538 This work was funded by l'Agence Nationale de la Recherche through grant CASSYSS ANR-
539 18-CE31-0010-01 (PI Johan Villeneuve). We thank the three anonymous reviewers for
540 constructive comments and Associate Editor Frédéric Moynier for careful editing. This is
541 CRPG contribution #2851.

542

543 **Data Availability**

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

546

547

548

549

550

551

552

553

554 **References**

555
556 Baeza L (2019) Oxygen isotope systematics of ordinary chondrite chondrules: insights into
557 the inner solar system planetary reservoir. Master of Philosophy of the Australian National
558 University.

559 Bekaert D. V., Auro M., Shollenberger Q. R., Liu M.-C., Marschall H., Burton K. W.,
560 Jacobsen B., Brennecka G. A., McPherson G. J., von Mutius R., Sarafian A. and Nielsen S.
561 G. (2021) Fossil records of early solar irradiation and cosmolocation of the CAI factory: A
562 reappraisal. *Sci. Adv.* **7**, eabg8329.

563 Bollard J., Connelly J. N., Whitehouse M. J., Pringle E. A., Bonal L., Jørgensen J. K.,
564 Nordlund A, Moynier F. and Bizzarro M. (2017) Early formation of planetary building
565 blocks inferred from Pb isotopic ages of chondrules. *Science Advances* **3**, e1700407.

566 Borlina C. S., Weiss B. P., Bryson J. F. J., Bai X.-N., Lima E. A., Chatterjee N. and Mansbach
567 E. N. (2021) Paleomagnetic evidence for a disk substructure in the early solar system.
568 *Science Advances* **7**, eabj6928.

569 Bouden N., Villeneuve J., Marrocchi Y., Deloule E., Füre E., Gurenko A., Piani L., Thomassot
570 E., Peres P. and Fernandes F. (2021) Triple Oxygen Isotope Measurements by Multi-
571 Collector Secondary Ion Mass Spectrometry. *Frontiers in Earth Science* **8**, 9.

572 Brennecka G. A., Burkhardt C., Budde G., Kruijjer T. S., Nimmo F. and Kleine T. (2020)
573 Astronomical context of Solar System formation from molybdenum isotopes in meteorite
574 inclusions. *Science* **370**, 837–840.

575 Bryson J. F. J. and Brennecka G. A. (2021) Constraints on Chondrule Generation, Disk
576 Dynamics, and Asteroid Accretion from the Compositions of Carbonaceous Meteorites.
577 *The Astrophysical Journal* **912**, 163.

578 Burkhardt C., Dauphas N., Hans U., Bourdon B. and Kleine T. (2019) Elemental and isotopic
579 variability in solar system materials by mixing and processing of primordial disk
580 reservoirs. *Geochimica et Cosmochimica Acta* **261**, 145–170.

581 Burkhardt C., Spitzer F., Morbidelli A., Budde G., Render J. H., Kruijjer T. S. and Kleine T.
582 (2021) Terrestrial planet formation from lost inner solar system material. *Science Advances*
583 **7**, eabj7601.

584 Chaumard N., Defouilloy C., Hertwig A. T. and Kita N. T. (2021) Oxygen isotope systematics
585 of chondrules in the Paris CM2 chondrite: indication for a single large formation region
586 across snow line. *Geochimica et Cosmochimica Acta* **2021**, 199-218.

587 Clayton R. N., Mayeda T. K. and Goswami J. N. (1986) Oxygen and Silicon Isotopic
588 Variations in Chondrules. *Meteoritics*, **21**, 346.

589 Clayton R. N., Mayeda T. K. and Goswami J. N. (1991) Oxygen isotope studies of ordinary
590 chondrites. *Geochimica et Cosmochimica Acta* **55**, 2317–2337.

591 Cuzzi J. N. and Alexander C. M. O. (2006) Chondrule formation in particle-rich nebular
592 regions at least hundreds of kilometres across. *Nature* **441**, 483–485.

593 Desch S. J., Kalyaan A. and Alexander C. M. O. (2018) The Effect of Jupiter’s Formation on
594 the Distribution of Refractory Elements and Inclusions in Meteorites. *The Astrophysical*
595 *Journal* **238**, 11.

596 Dunham E. T., Sheikh A., Opara D., Matsuda N., Liu M. - C. and McKeegan K. D. (2023)
597 Calcium–aluminum- rich inclusions in non- carbonaceous chondrites: Abundances, sizes,
598 and mineralogy. *Meteoritics & Planetary Science* **58**, 643–671.

599 Ebert S. and Bischoff A. (2016) Genetic relationship between Na-rich chondrules and Ca,Al-
600 rich inclusions? – Formation of Na-rich chondrules by melting of refractory and volatile
601 precursors in the solar nebula. *Geochimica et Cosmochimica Acta* **177**, 182–204.

602 Ebert S., Nagashima K., Bischoff A., Berndt J. and Krot A. N. (2022) Mineralogy, petrology,
603 and oxygen isotopic compositions of aluminum-rich chondrules from unequilibrated

604 ordinary and the Dar al Gani 083 (CO3.1) chondrite. *Geochimica et Cosmochimica Acta*
605 336, 448–468.

606 Ebert S., Render J., Brennecka G. A., Burkhardt C., Bischoff A., Gerber S. and Kleine T.
607 (2018) Ti isotopic evidence for a non-CAI refractory component in the inner Solar System.
608 *Earth and Planetary Science Letters* 498, 257–265.

609 Friedrich J. M., Weisberg M. K., Ebel D. S., Biltz A. E., Corbett B. M., Iotzov I. V., Khan W.
610 S. and Wolman M. D. (2015) Chondrule size and related physical properties: A compilation
611 and evaluation of current data across all meteorite groups. *Geochemistry* 75, 419–443.

612 Fukuda K., Tenner T. J., Kimura M., Tomioka N., Siron G., Ushikubo T., Chaumard N.,
613 Hertwig A. T. and Kita N. T. (2022) A temporal shift of chondrule generation from the
614 inner to outer Solar System inferred from oxygen isotopes and Al-Mg chronology of
615 chondrules from primitive CM and CO chondrites. *Geochimica et Cosmochimica Acta*
616 322, 194–226.

617 Gerber S., Burkhardt C., Budde G., Metzler K. and Kleine T. (2017) Mixing and Transport of
618 Dust in the Early Solar Nebula as Inferred from Titanium Isotope Variations among
619 Chondrules. *The Astrophysical Journal* 841, L17.

620 Grant H., Tartèse R., Jones R., Piani L., Marrocchi Y., King A. and Rigaudier T. (2023) Bulk
621 mineralogy, water abundance, and hydrogen isotope composition of unequilibrated
622 ordinary chondrites. *Meteoritics & Planetary Science* 58, 1365–1381.

623 Greenwood R. C., Burbine T. H. and Franchi I. A. (2020) Linking asteroids and meteorites to
624 the primordial planetesimal population. *Geochimica et Cosmochimica Acta* 277, 377–406.

625 Grossman L., Ganapathy R. and Davis A. M. (1977) Trace elements in the Allende
626 meteorite—III. Coarse-grained inclusions revisited. *Geochimica et Cosmochimica Acta* 41,
627 1647–1664.

628 Grossman L., Ganapathy R., Methot R. L. and Davis A. M. (1979) Trace elements in the
629 Allende meteorite—IV. Amoeboid olivine aggregates. *Geochimica et Cosmochimica Acta*
630 43, 817–829.

631 Haugbølle T., Weber P., Wielandt D. P., Benítez-Llambay P., Bizzarro M., Gressel O. and
632 Pessah M. E. (2019) Probing the Protosolar Disk Using Dust Filtering at Gaps in the Early
633 Solar System. *The Astrophysical Journal* 158, 55.

634 Jacquet E. (2019) Beryllium-10 production in gaseous protoplanetary disks and implications
635 for the astrophysical setting of refractory inclusions. *Astronomy and Astrophysics* 624,
636 A131-12.

637 Jacquet E., Alard O. and Gounelle M. (2015a) Trace element geochemistry of ordinary
638 chondrite chondrules: The type I/type II chondrule dichotomy. *Geochimica et*
639 *Cosmochimica Acta* 155, 47–67.

640 Jacquet E., Alard O. and Gounelle M. (2015b) The formation conditions of enstatite
641 chondrites: Insights from trace element geochemistry of olivine-bearing chondrules in
642 Sahara 97096 (EH3). *Meteoritics & Planetary Science* 50, 1624–1642.

643 Jacquet E., Fromang S. and Gounelle M. (2011) Radial transport of refractory inclusions and
644 their preservation in the dead zone. *Astronomy and Astrophysics* 526, L8-4.

645 Jacquet E., Gounelle M. and Fromang S. (2012) On the aerodynamic redistribution of
646 chondrite components in protoplanetary disks. *Icarus* 220, 162–173.

647 Jacquet E., Pignatale F. C., Chaussidon M. and Charnoz S. (2019) Fingerprints of the
648 Protosolar Cloud Collapse in the Solar System. II. Nucleosynthetic Anomalies in
649 Meteorites. *The Astrophysical Journal* 884, 32–11.

650 Jacquet E., Piralla M., Kersaho P. and Marrocchi Y. (2021) Origin of isolated olivine grains in
651 carbonaceous chondrites. *Meteoritics & Planetary Science* 56, 13–33.

652 Jacquet E. and Robert F. (2013) Water transport in protoplanetary disks and the hydrogen
653 isotopic composition of chondrites. *Icarus* 223, 722–732.

654 Jansen C. A., Burkhardt C., Marrocchi Y., Schneider J. M., Wölfer E. and Kleine T. (2024)
655 Condensate evolution in the solar nebula inferred from combined Cr, Ti, and O isotope
656 analyses of amoeboid olivine aggregates. *Earth and Planetary Science Letters* **627**, 118567.

657 Jones R. H. (2012) Petrographic constraints on the diversity of chondrule reservoirs in the
658 protoplanetary disk. *Meteoritics & Planetary Science* **47**, 1176–1190.

659 Jongejan S., Dominik C. and Dullemond C. P. (2023) The effect of Jupiter on the CAI storage
660 problem. *Astronomy and Astrophysics* **679**, A45.

661 Kita N. T., Nagahara H., Tachibana S., Tomomura S., Spicuzza M. J., Fournelle J. H. and
662 Valley J. W. (2010) High precision SIMS oxygen three isotope study of chondrules in LL3
663 chondrites: Role of ambient gas during chondrule formation. *Geochimica et Cosmochimica*
664 *Acta* **74**, 6610–6635.

665 van Kooten E., Schiller M., Moynier F., Johansen A., Haugbølle T. and Bizzarro M. (2021)
666 Hybrid Accretion of Carbonaceous Chondrites by Radial Transport across the Jupiter
667 Barrier. *The Astrophysical Journal* **910**, 70.

668 Krot A. N. (2019) Refractory inclusions in carbonaceous chondrites: Records of early solar
669 system processes. *Meteoritics & Planetary Science* **54**, 1647–1691.

670 Krot A. N., Petaev M. I., Russell S. S., Itoh S., Fagan T. J., Yurimoto H., Chizmadia L.,
671 Weisberg M. K., Komatsu M., Ulyanov A. A. and Keil K. (2004) Amoeboid olivine
672 aggregates and related objects in carbonaceous chondrites: records of nebular and asteroid
673 processes. *Chemie der Erde - Geochemistry* **64**, 185–239.

674 Kruijer T. S., Burkhardt C., Budde G. and Kleine T. (2017) Age of Jupiter inferred from the
675 distinct genetics and formation times of meteorites. *Proc Natl Acad Sci USA*, 201704461.

676 Kuznetsova A., Bae J., Hartmann L. and Low M.-M. M. (2022) Anisotropic Infall and
677 Substructure Formation in Embedded Disks. *The Astrophysical Journal* **928**, 92.

678 Libourel G. and Krot A. N. (2007) Evidence for the presence of planetesimal material among
679 the precursors of magnesian chondrules of nebular origin. *Earth and Planetary Science*
680 *Letters* **254**, 1–8.

681 Libourel G., Nagashima K., Portail M. and Krot A. N. (2023) On the significance of oxygen-
682 isotope variations in chondrules from carbonaceous chondrites. *Geochimica et*
683 *Cosmochimica Acta* **346**, 102–120.

684 Libourel G. and Portail M. (2018) Chondrules as direct thermochemical sensors of solar
685 protoplanetary disk gas. *Science Advances* **4**, eaar3321-32.

686 Marrocchi Y., Bonal L., Gattacceca J., Piani L., Beck P., Greenwood R., Eschrig J., Basque
687 A., Nuccio P. M. and Martin F. F. (2020) The Piancaldoli meteorite: A forgotten primitive
688 LL3.10 ordinary chondrite. *Meteoritics & Planetary Science* **55**, 1924-1935.

689 Marrocchi Y. and Chaussidon M. (2015) A systematic for oxygen isotopic variation in
690 meteoritic chondrules. *Earth and Planetary Science Letters* **430**, 308–315.

691 Marrocchi Y., Euverte R., Villeneuve J., Batanova V., Welsch B., Ferrière L. and Jacquet E.
692 (2019b) Formation of CV chondrules by recycling of amoeboid olivine aggregate-like
693 precursors. *Geochimica et Cosmochimica Acta* **247**, 121–141.

694 Marrocchi Y., Piralla M., Regnault M., Batanova V., Villeneuve J. and Jacquet E. (2022)
695 Isotopic evidence for two chondrule generations in CR chondrites and their relationships to
696 other carbonaceous chondrites. *Earth and Planetary Science Letters* **593**, 117683.

697 Marrocchi Y., Piralla M. and Tissot F. L. H. (2023) Iron Isotope Constraints on the Structure
698 of the Early Solar System. *The Astrophysical Journal* **954**, L27.

699 Marrocchi Y., Villeneuve J., Batanova V., Piani L. and Jacquet E. (2018) Oxygen isotopic
700 diversity of chondrule precursors and the nebular origin of chondrules. *Earth and Planetary*
701 *Science Letters* **496**, 132–141.

702 Marrocchi Y., Villeneuve J., Jacquet E., Piralla M. and Chaussidon M. (2019a) Rapid
703 condensation of the first Solar System solids. *Proceedings of the National Academy of*
704 *Sciences of the United States of America* 116, 23461–23466.

705 Morin G. L. F., Marrocchi Y., Villeneuve J. and Jacquet E. (2022) ^{16}O -rich anhydrous silicates
706 in CI chondrites: Implications for the nature and dynamics of dust in the solar accretion
707 disk. *Geochimica et Cosmochimica Acta* 332, 203–219.

708 Nakashima D., Nakamura T., Zhang M., Kita N. T., Mikouchi T., Yoshida H., Enokido Y.,
709 Morita T., Kikuri M., Amano K., Kagawa E., Yada T., Nishimura M., Nakato A., Miyazaki
710 A., Yogata K., Abe M., Okada T., Usui T., Yoshikawa M., Saiki T., Tanaka S., Nakazawa
711 S., Terui F., Yurimoto H., Noguchi T., Yabuta H., Naraoka H., Okazaki R., Sakamoto K.,
712 Watanabe S., Tachibana S. and Tsuda Y. (2023) Chondrule-like objects and Ca-Al-rich
713 inclusions in Ryugu may potentially be the oldest Solar System materials. *Nature*
714 *Communications* 14, 532.

715 Nanne J. A. M., Nimmo F., Cuzzi J. N. and Kleine T. (2019) Origin of the non-carbonaceous–
716 carbonaceous meteorite dichotomy. *Earth and Planetary Science Letters* 511, 44–54.

717 Olsen M. B., Wielandt D., Schiller M., Van Kooten E. M. M. E. and Bizzarro M. (2016)
718 Magnesium and ^{54}Cr isotope compositions of carbonaceous chondrite chondrules –
719 Insights into early disk processes. *Geochimica et Cosmochimica Acta* 191, 118–138.

720 Pack A., Yurimoto H. and Palme H. (2004) Petrographic and oxygen-isotopic study of
721 refractory forsterites from R-chondrite Dar al Gani 013 (R3.5–6), unequilibrated ordinary
722 and carbonaceous chondrites. *Geochimica et Cosmochimica Acta* 68, 1135–1157.

723 Pape J., Mezger K., Bouvier A.-S. and Baumgartner L. P. (2019) Time and duration of
724 chondrule formation: Constraints from ^{26}Al - ^{26}Mg ages of individual chondrules.
725 *Geochimica et Cosmochimica Acta* 244, 416–436.

726 Pascucci I., Cabrit S., Edwards S., Gorti U., Gressel O. and Suzuki T. (2022) The Role of
727 Disk Winds in the Evolution and Dispersal of Protoplanetary Disks. *Protostars and Planets*
728 *VII, ASP Conference Series, Vol. 534, Proceedings of a conference held 10-15 April 2023*
729 *at Kyoto, Japan. Edited by Shu-ichiro Inutsuka, Yuri Aikawa, Takayuki Muto, Kengo*
730 *Tomida, and Motohide Tamura. San Francisco: Astronomical Society of the Pacific, 2023,*
731 *p.567*

732 Piani L., Marrocchi Y., Vacher L. G., Yurimoto H. and Bizzarro M. (2021) Origin of hydrogen
733 isotopic variations in chondritic water and organics. *Earth and Planetary Science Letters*
734 567, 117008.

735 Pignatale F. C., Jacquet E., Chaussidon M. and Charnoz S. (2019) Fingerprints of the
736 Protosolar Cloud Collapse in the Solar System. I. Distribution of Presolar Short-lived ^{26}Al .
737 *The Astrophysical Journal* 884, 31–8.

738 Pinto G. A., Marrocchi Y., Morbidelli A., Charnoz S., Varela M. E., Soto K., Martínez R., and
739 Olivares F. (2021). Constraints on Planetesimal Accretion Inferred from Particle-size
740 Distribution in CO Chondrites. *The Astrophysical Journal Letters* 917:L25.

741 Pinto G. A., Jacquet E., Corgne A., Olivares F., Villeneuve J. and Marrocchi Y. (2024)
742 Deciphering recycling processes during solar system evolution from magnesium-rich relict
743 olivine grains in type II chondrules. *Geochimica et Cosmochimica Acta* 364, 65–78.

744 Piralla M., Villeneuve J., Batanova V., Jacquet E. and Marrocchi Y. (2021) Conditions of
745 chondrule formation in ordinary chondrites. *Geochimica et Cosmochimica Acta* 313, 295–
746 312.

747 Piralla M., Villeneuve J., Schnuriger N., Bekaert D. V. and Marrocchi Y. (2023) A unified
748 chronology of dust formation in the early solar system. *Icarus* 394, 115427.

749 Regnault M., Marrocchi Y., Piralla M., Villeneuve J., Batanova V., Schnuriger N. and Jacquet
750 E. (2022) Oxygen isotope systematics of chondrules in Rumuruti chondrites: Formation

751 conditions and genetic link with ordinary chondrites. *Meteoritics & Planetary Science* 57,
752 122–135.

753 Russell S. S., MacPherson G. J., Leshin L. A. and McKeegan K. D. (2000) ^{16}O enrichments in
754 aluminum-rich chondrules from ordinary chondrites. *Earth and Planetary Science Letters*
755 184, 57–74.

756 Russell S. S., Srinivasan G., Huss G. R., Wasserburg G. J. and MacPherson G. J. (1996)
757 Evidence for Widespread ^{26}Al in the Solar Nebula and Constraints for Nebula Time Scales.
758 *Science* 273, 757–762.

759 Ruzicka A., Hiyagon H., Hutson M. and Floss C. (2007) Relict olivine, chondrule recycling,
760 and the evolution of nebular oxygen reservoirs. *Earth and Planetary Science Letters* 257,
761 274–289.

762 Saxton J. M., Lyon I. C. and Turner G. (1998) Oxygen isotopes in forsterite grains from
763 Julesburg and Allende: Oxygen-16-rich material in an ordinary chondrite. *Meteoritics &*
764 *Planetary Science* 33, 1017–1027.

765 Schindelin J., Arganda-Carreras I., Frise E., Kaynig V., Longair M., Pietzsch T., Preibisch S.,
766 Rueden C., Saalfeld S., Schmid B., Tinevez J.-Y., White D. J., Hartenstein V., Eliceiri K.,
767 Tomancak P. and Cardona A. (2012) Fiji: an open-source platform for biological-image
768 analysis. *Nat Methods* 9, 676–682.

769 Schneider J. M., Burkhardt C., Marrocchi Y., Brennecka G. A. and Kleine T. (2020) Early
770 evolution of the solar accretion disk inferred from Cr-Ti-O isotopes in individual
771 chondrules. *Earth and Planetary Science Letters* 551, 116585.

772 Schnuriger N., Cartier C., Villeneuve J., Batanova V., Regnault M. and Marrocchi Y. (2022)
773 Spinel in CV chondrules: Investigating precursor legacy and chondrule thermal histories.
774 *Meteoritics & Planetary Science* 57, 1018–1037.

775 Schrader D. L., Connolly Jr H. C., Lauretta D. S., Nagashima K., Huss G. R., Davidson J. and
776 Domanik K. J. (2013) The formation and alteration of the Renazzo-like carbonaceous
777 chondrites II: Linking O-isotope composition and oxidation state of chondrule olivine.
778 *Geochimica et Cosmochimica Acta* 101, 302–327.

779 **Schrader D. L., Nagashima K., Davidson J., McCoy T. J., Ogliore R. C. and Fu R. R. (2020)**
780 **Outward migration of chondrule fragments in the early Solar System: O-isotopic evidence**
781 **for rocky material crossing the Jupiter Gap? *Geochimica et Cosmochimica Acta* 282, 133–**
782 **155.**

783 Spitzer F., Burkhardt C., Budde G., Kruijer T. S., Morbidelli A. and Kleine T. (2020) Isotopic
784 Evolution of the Inner Solar System Inferred from Molybdenum Isotopes in Meteorites.
785 *The Astrophysical Journal* 898, L2.

786 Tenner T. J., Nakashima D., Ushikubo T., Kita N. T. and Weisberg M. K. (2015) Oxygen
787 isotope ratios of FeO-poor chondrules in CR3 chondrites: Influence of dust enrichment and
788 H_2O during chondrule formation. *Geochimica et Cosmochimica Acta* 148, 228–250.

789 Tenner T. J., Ushikubo T., Kurahashi E., Kita N. T. and Nagahara H. (2013) Oxygen isotope
790 systematics of chondrule phenocrysts from the CO3.0 chondrite Yamato 81020: Evidence
791 for two distinct oxygen isotope reservoirs. *Geochimica et Cosmochimica Acta* 102, 226–
792 245.

793 Tenner, T. J., Ushikubo, T., Nakashima, D., Schrader, D. L., Weisberg, M. K., Kimura, M., &
794 Kita, N. T. (2018). Oxygen Isotope Characteristics of Chondrules from Recent Studies by
795 Secondary Ion Mass Spectrometry. In *Chondrules: Records of Protoplanetary Disk*
796 *Processes*. Cambridge University Press, pp. 196–246.

797 **Torrano Z. A., Alexander C. M. O., Carlson R. W., Render J., Brennecka G. A. and Bullock E.**
798 **S. (2024) A common isotopic reservoir for amoeboid olivine aggregates (AOAs) and**
799 **calcium-aluminum-rich inclusions (CAIs) revealed by Ti and Cr isotopic compositions.**
800 ***Earth and Planetary Science Letters* 627, 118551.**

801 Trinquier A., Birck J. and Allegre C. J. (2007) Widespread ^{54}Cr Heterogeneity in the Inner
802 Solar System. *The Astrophysical Journal* 655, 1179–1185.

803 Trinquier A., Elliott T., Ulfbeck D., Coath C., Krot A. N. and Bizzarro M. (2009) Origin of
804 Nucleosynthetic Isotope Heterogeneity in the Solar Protoplanetary Disk. *Science* 324, 374–
805 376.

806 Ushikubo T., Kimura M., Kita N. T. and Valley J. W. (2012) Primordial oxygen isotope
807 reservoirs of the solar nebula recorded in chondrules in Acfer 094 carbonaceous chondrite.
808 *Geochimica et Cosmochimica Acta* 90, 242–264.

809 Warren P. H. (2011) Stable-isotopic anomalies and the accretionary assemblage of the Earth
810 and Mars: A subordinate role for carbonaceous chondrites. *Earth and Planetary Science*
811 *Letters* 311, 93–100.

812 Weiss B. P., Bai X.-N. and Fu R. R. (2021) History of the solar nebula from meteorite
813 paleomagnetism. *Sci. Adv.* 7, eaba5967.

814 Williams C. D., Sanborn M. E., Defouilloy C., Yin Q.-Z., Kita N. T., Ebel D. S., Yamakawa A.
815 and Yamashita K. (2020) Chondrules reveal large-scale outward transport of inner Solar
816 System materials in the protoplanetary disk. *Proc Natl Acad Sci USA* 117, 23426–23435.

817 Yamamoto D., Kawasaki N., Tachibana S., Kamibayashi M. and Yurimoto H. (2021) An
818 experimental study on oxygen isotope exchange reaction between CAI melt and low-
819 pressure water vapor under simulated Solar nebular conditions. *Geochimica et*
820 *Cosmochimica Acta* 314, 108–120.

821 Yamamoto D., Kawasaki N., Tachibana S., Kamibayashi M. and Yurimoto H. (2022) Oxygen
822 isotope exchange kinetics between CAI melt and carbon monoxide gas: Implication for
823 CAI formation in the earliest Solar System. *Geochimica et Cosmochimica Acta* 336, 104–
824 112.

825 Yap T. E. and Tissot F. L. H. (2023) The NC-CC dichotomy explained by significant addition
826 of CAI-like dust to the Bulk Molecular Cloud (BMC) composition. *Icarus* 405, 115680.

827 Young E. D., Kuramoto K., Marcus R. A., Yurimoto H. and Jacobsen S. B. (2008) Mass-
828 independent Oxygen Isotope Variation in the Solar Nebula. *Reviews in Mineralogy and*
829 *Geochemistry* 68, 187–218.

830 Zanda B., Hewins R., Bourot-Denise M., Bland P. and Alabarède F. (2006) Formation of solar
831 nebula reservoirs by mixing chondritic components. *Earth and Planetary Science Letters*
832 248, 650–660.

833 Zhu K., Moynier F., Schiller M. and Bizzarro M. (2020) Dating and Tracing the Origin of
834 Enstatite Chondrite Chondrules with Cr Isotopes. *The Astrophysical Journal* 894, L26.

835
836
837
838
839
840
841
842
843
844
845
846
847
848
849
850

851
852
853
854
855
856
857
858
859
860
861

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).

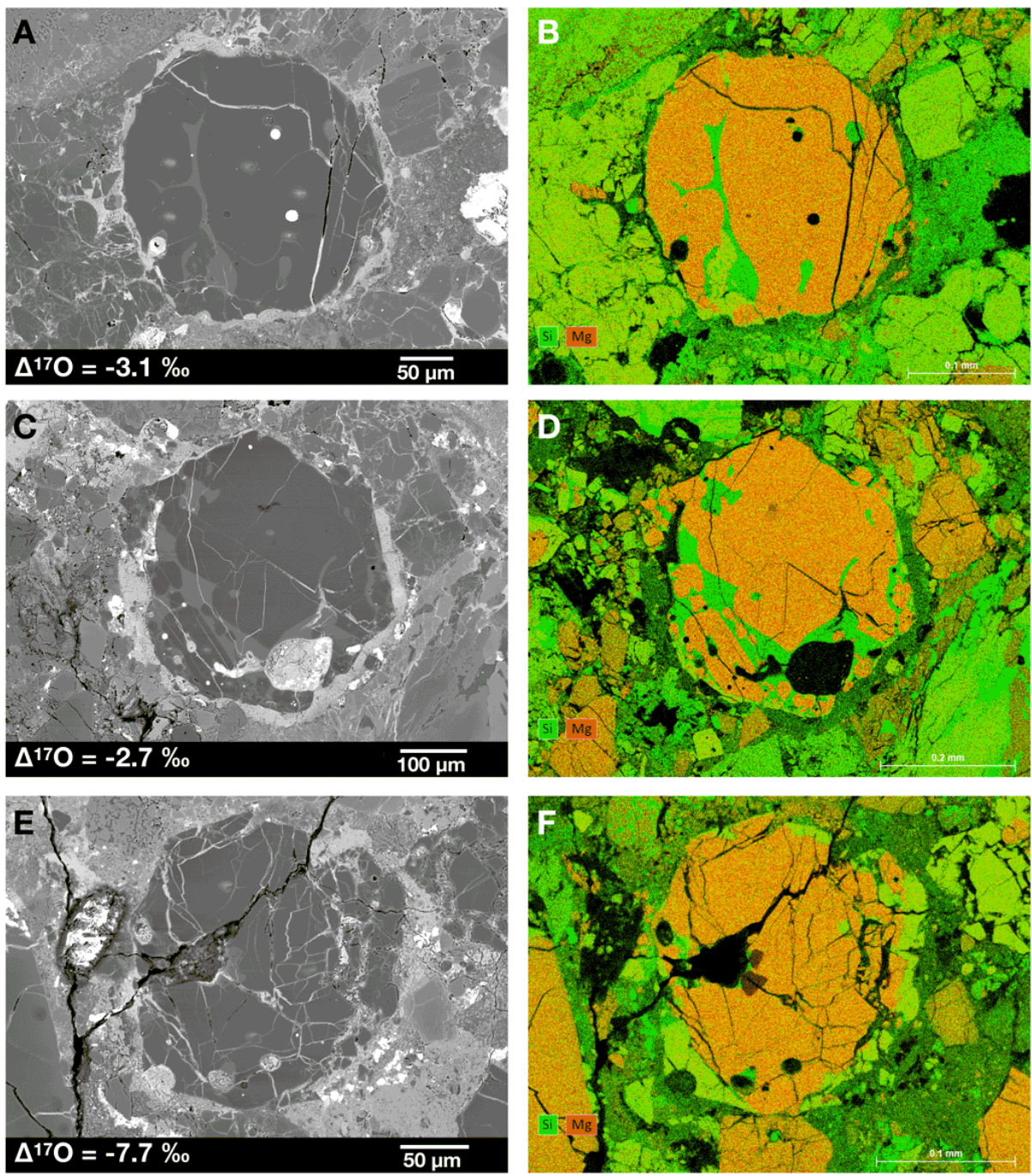


Fig. 1

917
 918
 919
 920
 921
 922
 923
 924
 925
 926
 927
 928
 929

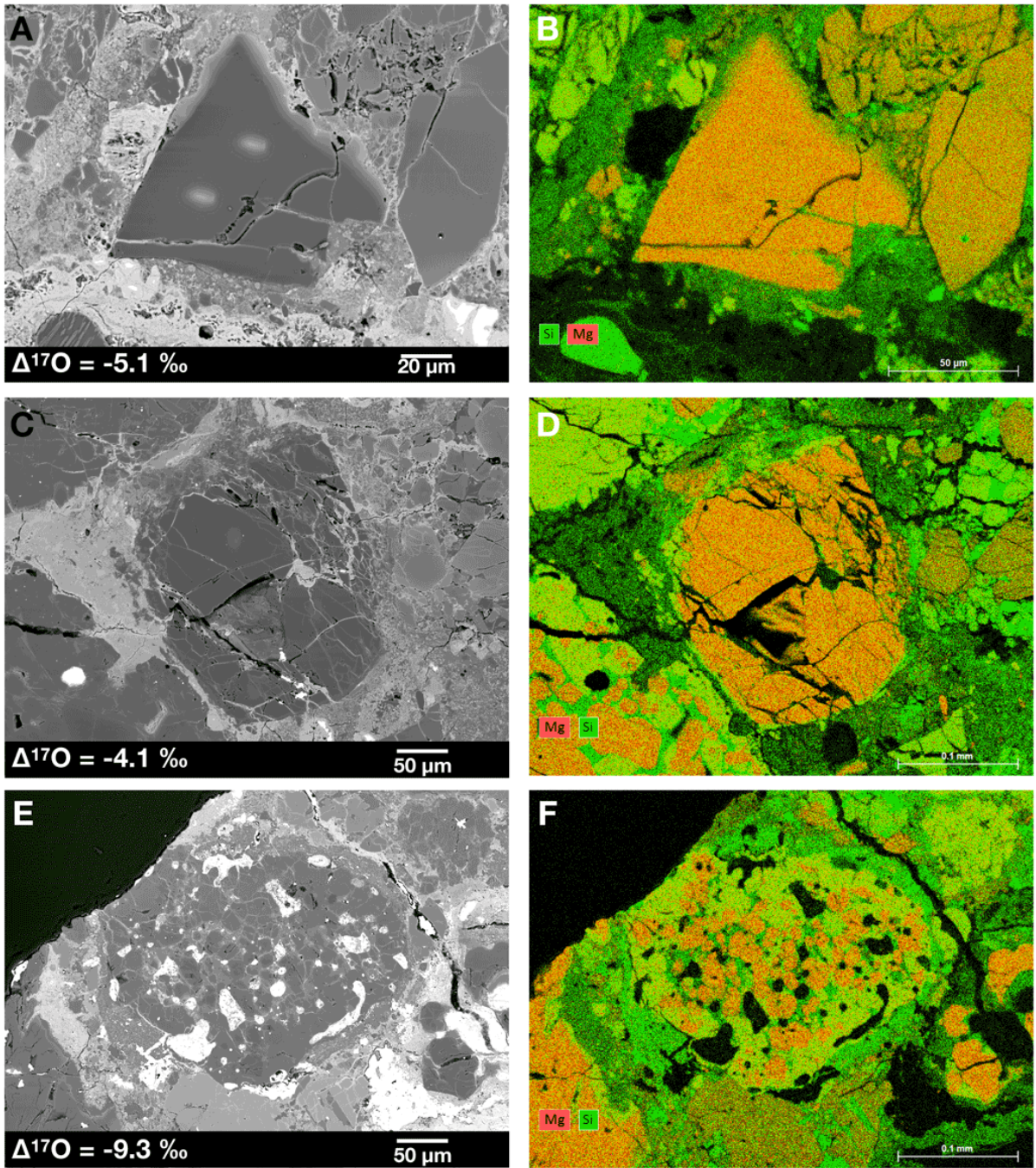
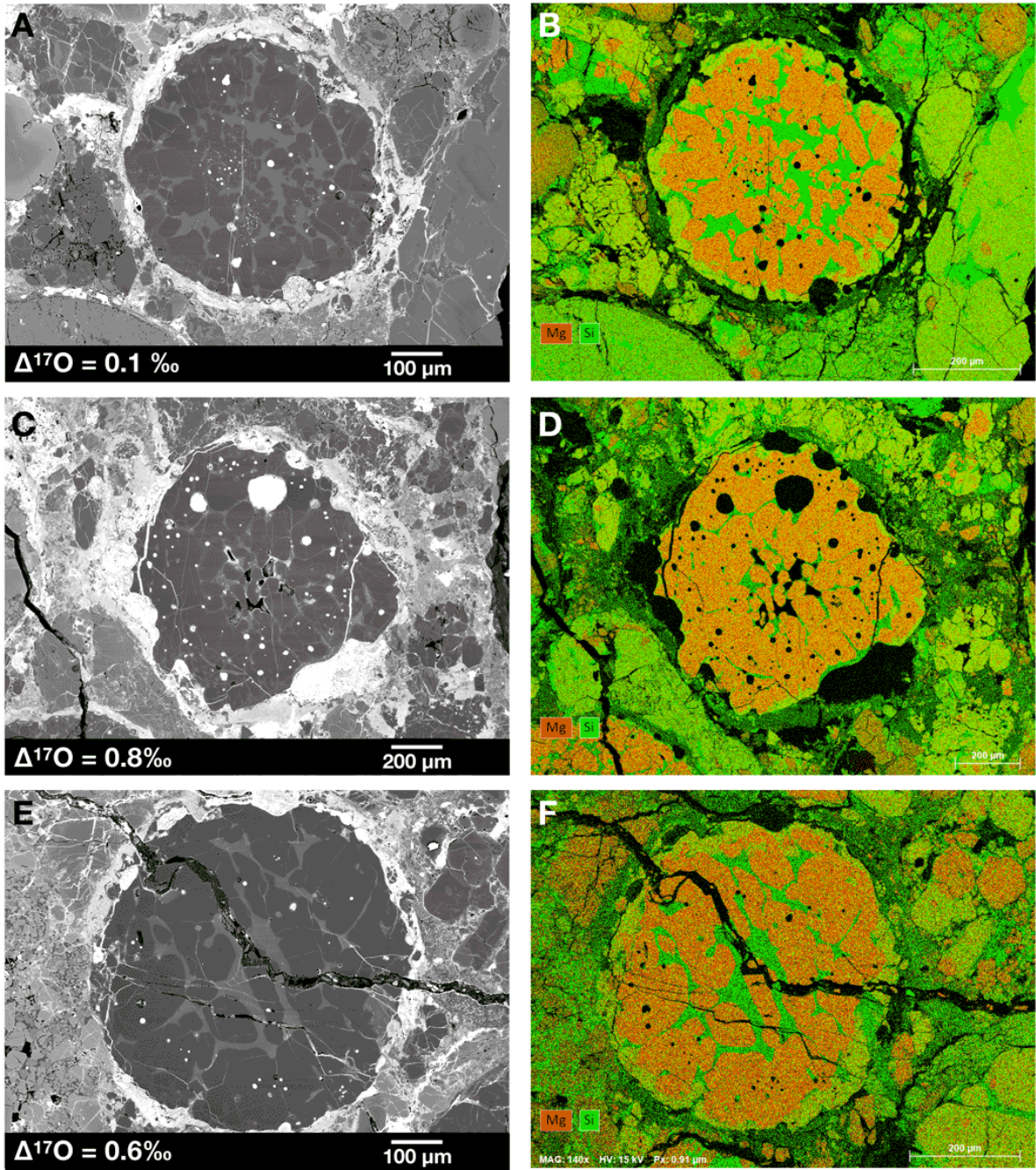


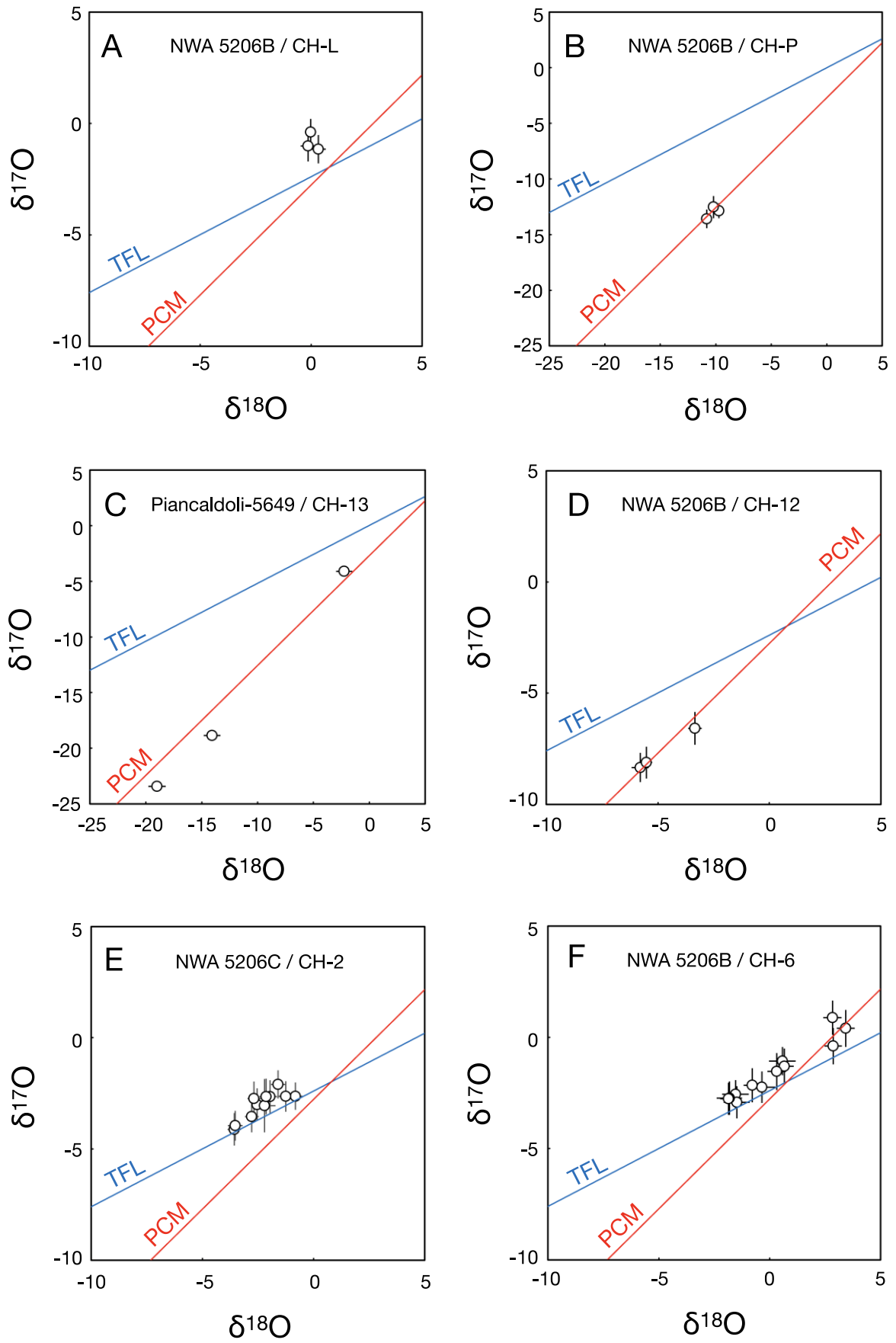
Fig. 2

930
 931
 932
 933
 934
 935
 936
 937
 938
 939
 940
 941
 942
 943



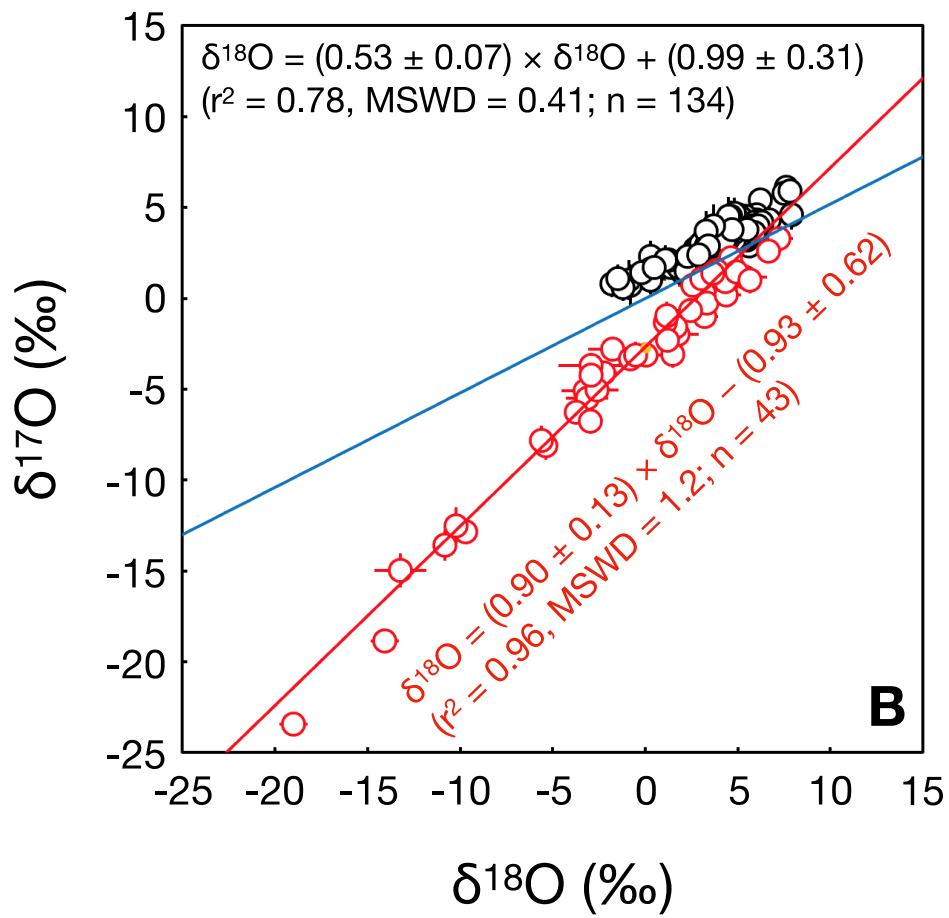
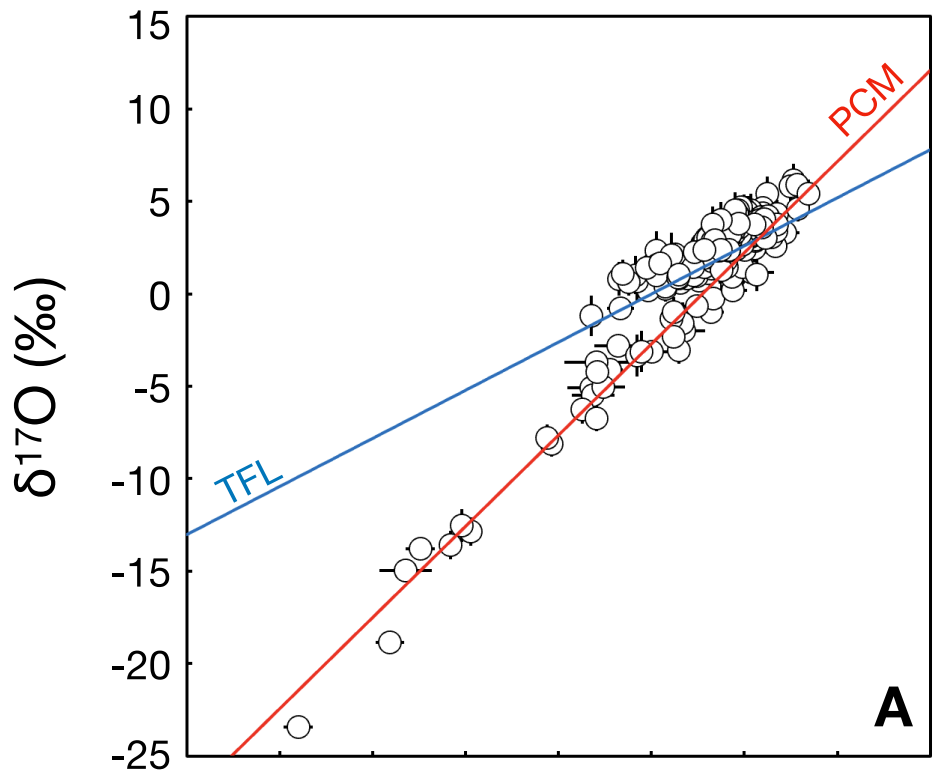
944
 945
 946
 947
 948
 949
 950

Fig. 3



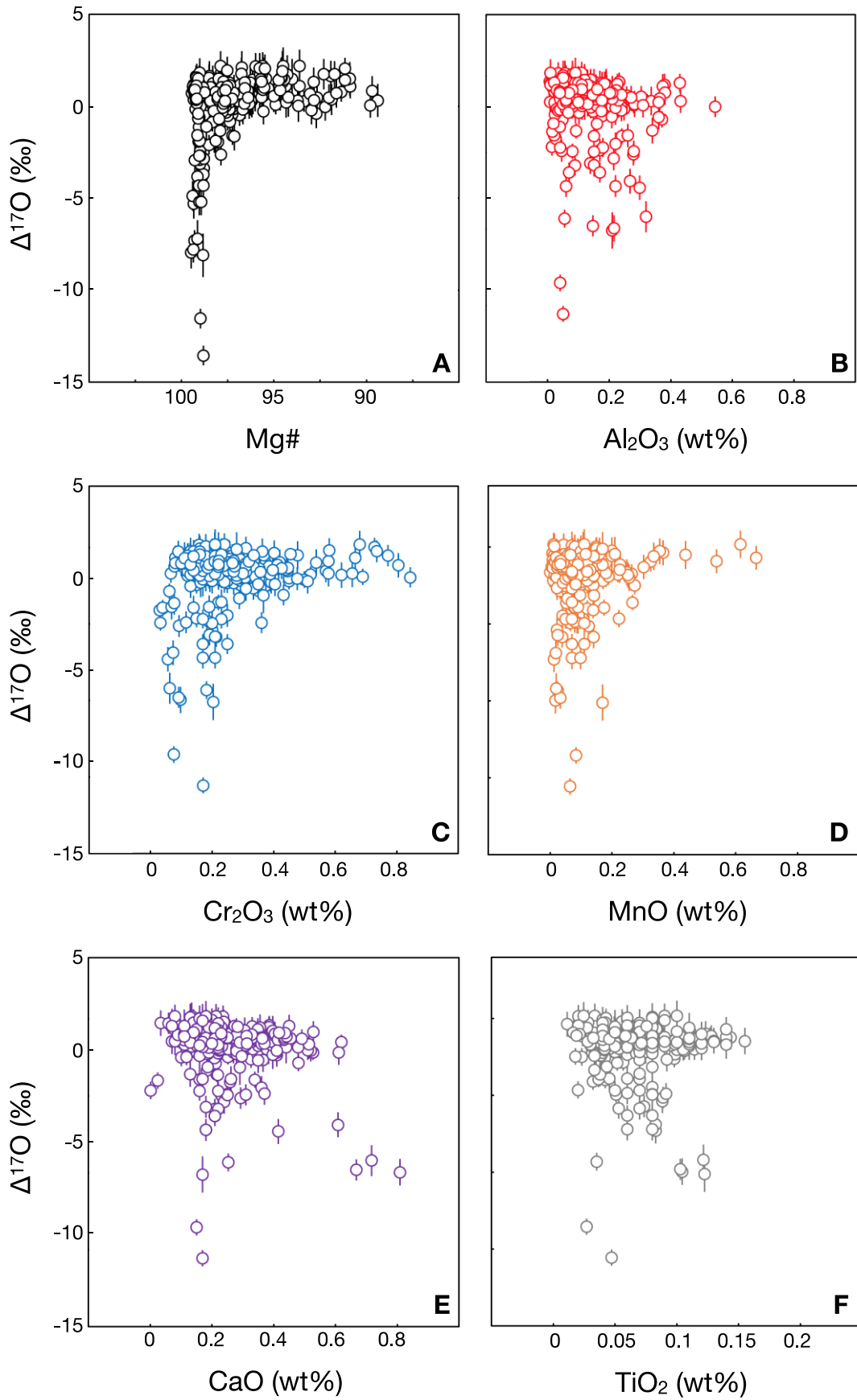
951
952

Fig. 4



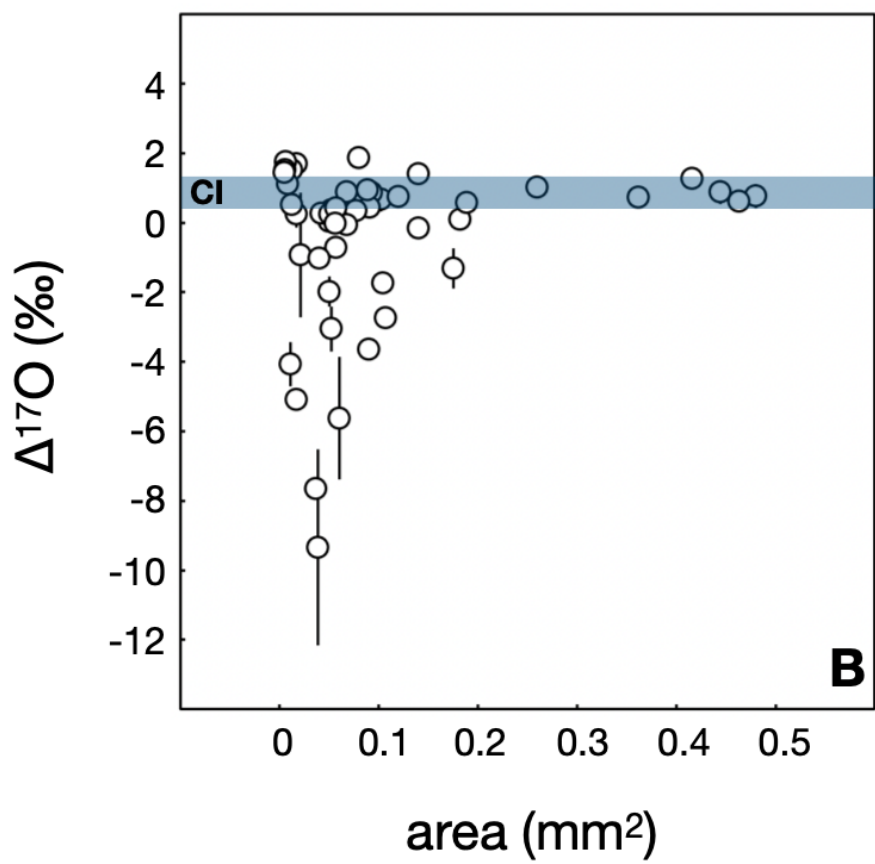
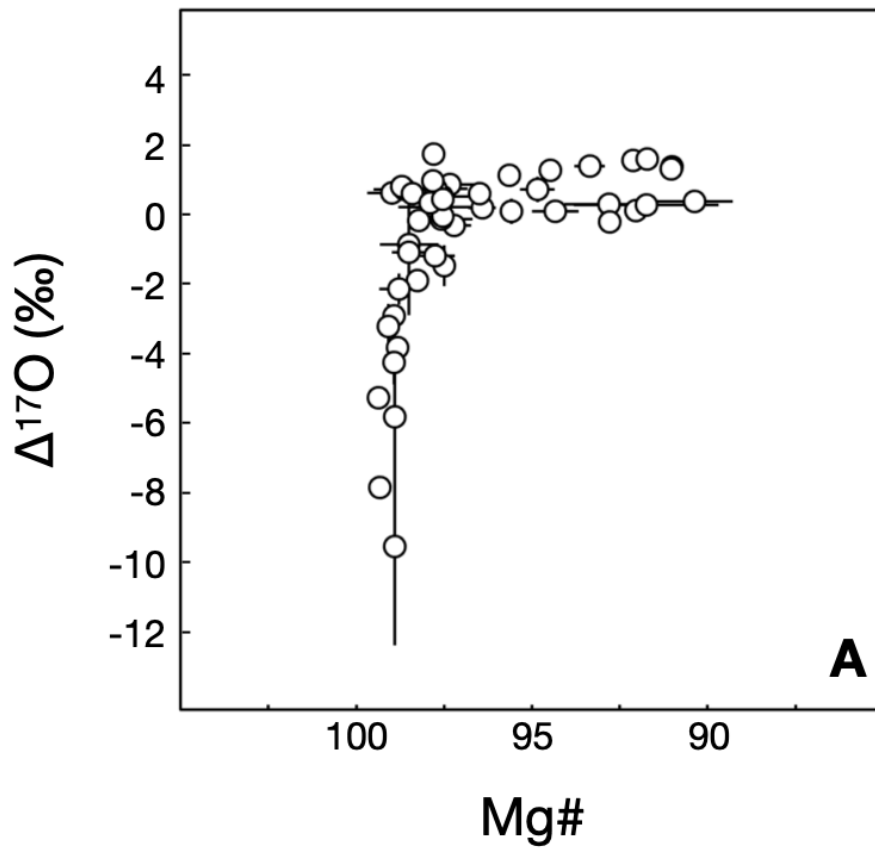
953
 954
 955

Fig. 5



956
957

Fig. 6



958
959

Fig. 7

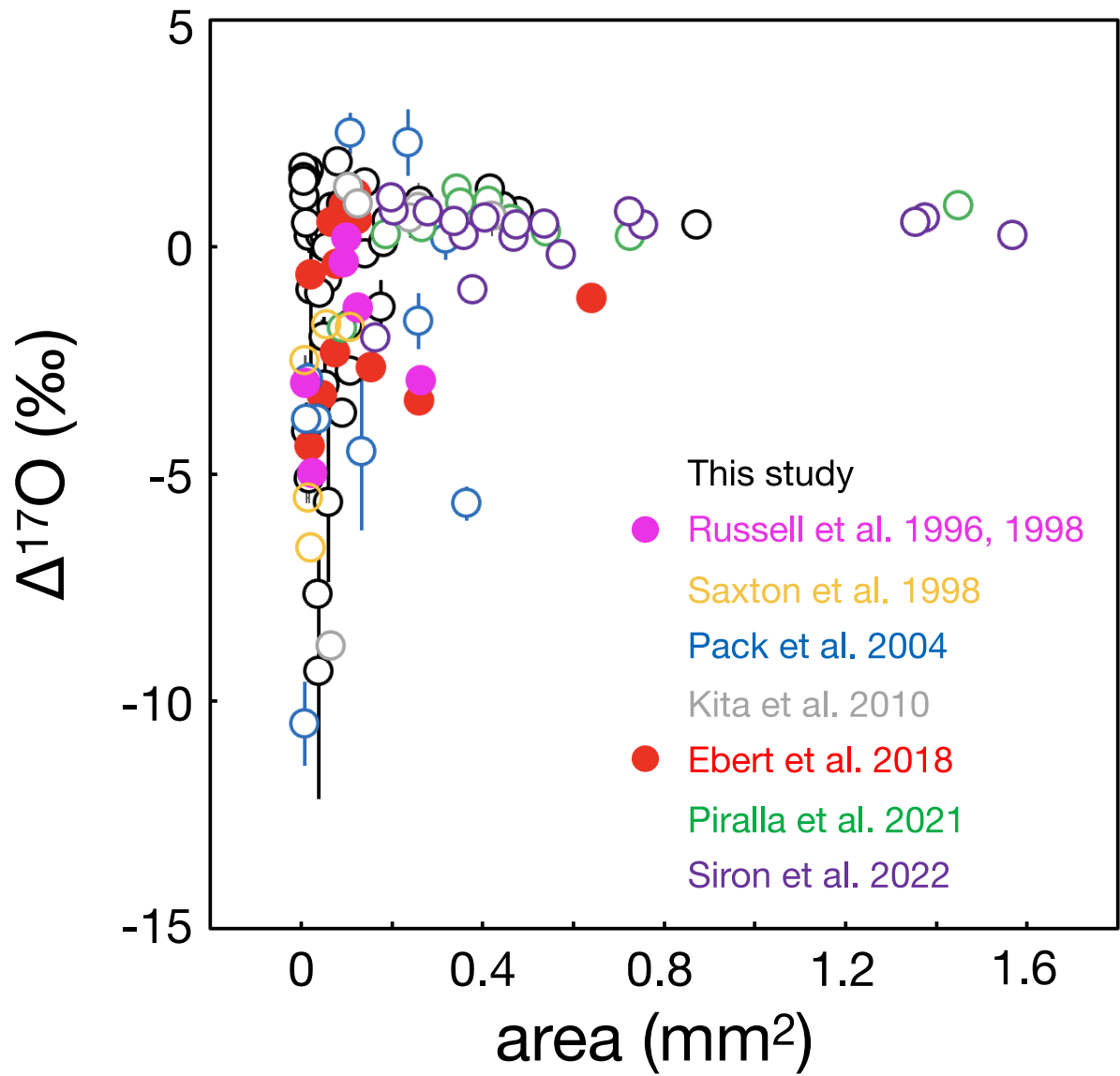
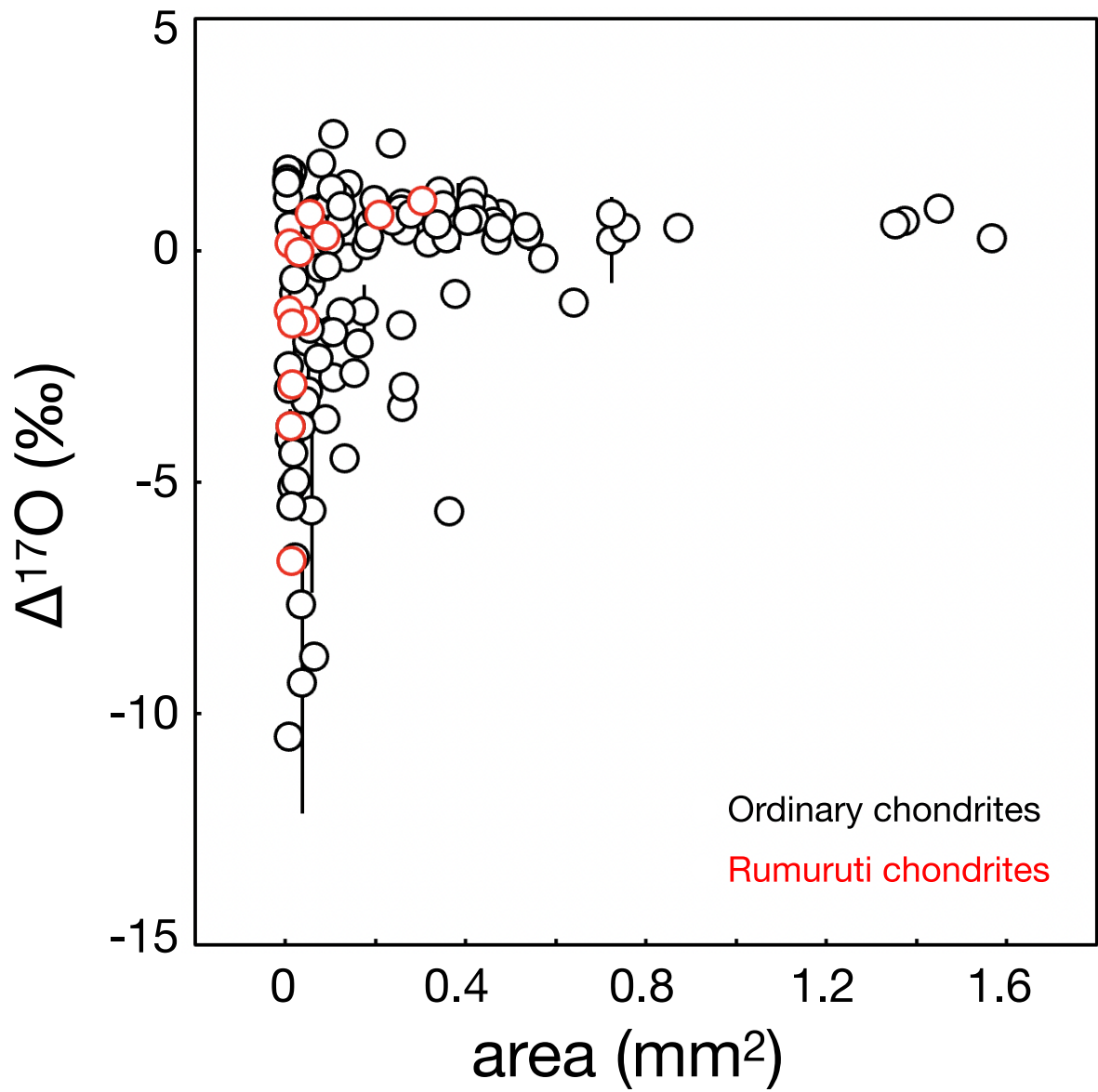


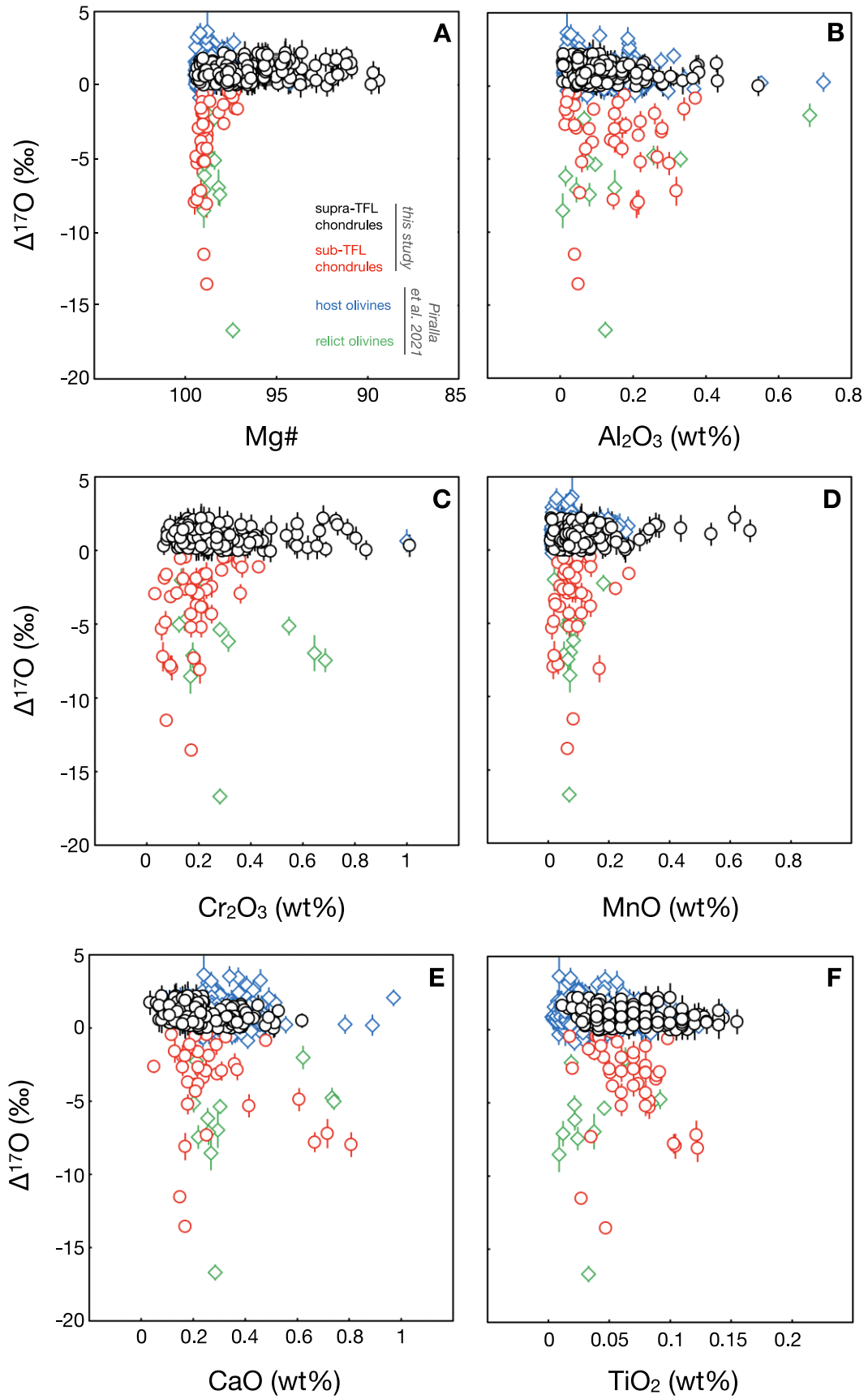
Fig. 8

960
 961
 962
 963
 964
 965
 966
 967
 968
 969
 970
 971



972
973
974
975

Fig. 9



976
 977
 978
 979

Fig. 10