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Origin of isolated olivine grains in carbonaceous chondrites

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27 ***Abstract***

28

29 We report microscopic, cathodoluminescence, chemical and O isotopic measurements
30 of FeO-poor isolated olivine grains (IOG) in the carbonaceous chondrites Allende (CV3),
31 Northwest Africa 5958 (C2-ung), Northwest Africa 11086 (CM2-an), Allan Hills 77307
32 (CO3.0). The general petrographic, chemical and isotopic similarity with *bona fide* type I
33 chondrules confirms that the IOG derived from them. The concentric CL zoning, reflecting a
34 decrease in refractory elements toward the margins, and frequent rimming by enstatite are taken
35 as evidence of interaction of the IOG with the gas as stand-alone objects. This indicates that
36 they were splashed out of chondrules when these were still partially molten. CaO-rich refractory
37 forsterites, which are restricted to $\Delta^{17}\text{O} < -4 \text{ ‰}$ likely escaped equilibration at lower
38 temperatures because of their large size and possibly quicker quenching. The IOG thus bear
39 witness to frequent collisions in the chondrule-forming regions.

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1. Introduction

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56 In their seminal description of Murchison, Fuchs et al. (1973) reported isolated olivine
57 grains (henceforth IOG), one of which earned the honor of their frontispiece. With the
58 condensation models in full swing since the fall of Allende (e.g., Grossman 1972; Marvin et al.
59 1970), it was but natural to interpret these grains, soon found in other carbonaceous chondrite
60 clans, as nebular condensation products, for which Olsen and Grossman (1974, 1978) adduced
61 morphological evidence such as large size, euhedral shapes and patterned crystal surfaces. Yet
62 chemical analogies of the IOG with chondrule olivine, as well as the presence of glass
63 inclusions chemically comparable to chondrule mesostases, suggested early that they were
64 chondrule fragments, possibly liberated after alteration of the mesostasis (e.g. McSween (;
65 Richardson and McSween 1978; Desnoyers (; Nagahara and Kushiro (; Jones (1992.

66 Still, Steele (1986) drew attention to a subset of IOG, both in carbonaceous and ordinary
67 chondrites, with particularly forsteritic and refractory mineral chemistries and bright blue
68 cathodoluminescence, which led him to entertain again a condensate origin (e.g. Steele (, Steele
69 , for which the SIMS (Secondary Ion Mass Spectrometer) measurements of Weinbruch et al.
70 (1993) found supporting evidence in their ^{16}O enrichment (that is, in the direction of refractory
71 inclusion compositions) and flat rare earth element (REE) patterns in olivine, inconsistent with
72 igneous partitioning. Yet, as further data confirmed, the oxygen isotopic composition of the
73 IOG largely overlap with chondrules in carbonaceous chondrites (e.g., Jones et al. (; Russell
74 et al. (; Ushikubo et al. 2012). Also, *in situ* laser ablation inductively coupled mass spectrometer
75 (LA-ICP-MS) analyses have revealed that IOG olivine has, in fact, quite fractionated REE, in
76 accordance with near-equilibrium igneous partitioning (e.g. Jacquet et al. (; Jacquet and
77 Marrocchi (; Pack et al. (2005. While the balance of evidence thus remains in favor of a genetic
78 link between IOG and chondrules, some systematic differences, at least among the forsteritic
79 ones, with the general population of chondrules remain to be understood so as to decide whether
80 they sample the same heating events. For instance, the relative ^{16}O enrichment of Tagish Lake
81 (C2-ung) IOG indicated “more primitive material than the forsterite-rich chondrules” to Russell
82 et al. (2010) while Jacquet and Marrocchi (2017) attempted to relate their coarse grain size and
83 incompatible element enrichment of IOG in Northwest Africa (NWA) 5958 (C2-ung) to longer
84 thermal processing. The IOG would be obviously important (olivine-rich) pieces of the puzzle

85 of chondrule origin, if only their exact formation context *vis-à-vis* mainstream chondrules could
86 be ascertained.

87 Our recent Secondary Ion Mass Spectrometer (SIMS)/electron microprobe (EMP) work
88 on type I chondrules (that is, reduced, with Fo₉₀₋₁₀₀ olivine) with porphyritic olivine (PO)
89 textures in carbonaceous chondrites has increased our understanding of the formation of olivine
90 in these objects (Marrocchi et al. (, Marrocchi et al. We have identified Al-Ti-poor cores in
91 central olivine grains whose oxygen isotopic deviation from the host revealed their relict nature
92 and likely derivation from amoeboid olivine aggregate (AOA)-like precursors. These are
93 overgrown by olivine enriched in incompatible elements and crystallized from melt produced
94 during initial heating. Finally, palisadic olivine grains near the chondrule margins show
95 evidence for gas-assisted outward growth, as also beautifully depicted by the
96 cathodoluminescence (CL) maps of Libourel and Portail (2018). These different generations of
97 olivine multiply our opportunities to find suitable analogs for IOG, at least for the magnesian
98 population which dominates in carbonaceous chondrites. We have thus set to extend our work
99 from type I chondrules to their isolated olivine counterparts (which we will also refer to as “type
100 I IOG”) in carbonaceous chondrites, where such reduced compositions dominate their high-
101 temperature fraction. Specifically, we present in this paper combined petrographic (CL),
102 chemical (EMP), oxygen isotopic (SIMS) data for magnesian IOG and chondrules in CM, CO
103 and CV chondrites. The comparison will afford new insights on the genesis of these IOG.

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105 **2. Samples and methods**

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107 We surveyed the polished sections NWA 5958-1 and NWA 5958-4 of Northwest Africa
108 (NWA) 5958, the thin section 3181lm4 (or 3181-4) of Allende (all three from the Muséum
109 national d’Histoire naturelle, Paris, France), the thin section ALH 77307,96 of Allan Hills
110 (ALH) 77307 (from the NASA Antarctic Search for Meteorites program) and a thick section of
111 NWA 11086 prepared at CRPG. Allende is an oxidized Allende-like CV sub-group classified
112 as a CV3.6 by Bonal et al. (2006). ALH 77307 is a CO3.0 chondrite (Grossman and Brearley
113 2005; Bonal et al. 2007; Busemann et al. 2007). Opaque assemblages, chondrule mesostases,
114 olivine textures and compositions of chondrules show no indication of alteration and diffusional
115 exchange. NWA 5958 corresponds to a C2-ung CM-like chondrite with type II chondrule
116 olivine Cr content and opaque petrography indicating minimal thermal metamorphism (<

117 300°C) but chondrule mesostases bearing witness to significant aqueous alteration (Jacquet et
118 al., 2016; Jacquet and Marrocchi 2017). NWA 11086 has been classified as a CM-an due to ¹⁶O
119 enrichment and the absence of phyllosilicates according to X-ray diffraction (Gattacceca et al.
120 2019), but significant aqueous alteration is indicated by presence of PCP (Poorly Characterized
121 Phases, also known as tochilinite cronstedtite intergrowths; see e.g. Lentfort et al., this issue),
122 conversion of chondrule mesostasis to « spinach » (Fuchs et al. 1973) and paucity of Fe-Ni
123 metal, so it might be a plain, if somewhat ¹⁶O-rich, CM2 chondrite. In the plots and the
124 discussion, data from these latter two chondrites and ALH 77307 will be subsumed in the
125 CM/CO clan.

126 Secondary Electron Microscope (SEM) imaging was performed on a JEOL JSM-6510
127 SEM equipped with a Genesis EDX detector at the Centre de Recherches Pétrographiques et
128 Géochimiques (CRPG-CNRS, Nancy, France) using 3 nA electron beam accelerated at 15 kV.
129 Modal abundances of high-temperature components were also estimated by manual point
130 counting on BSE maps using the JMicrovision software (N. Roduit;
131 <https://jmicrovision.github.io>; last accessed in December 2019) with 4000 points (randomly
132 chosen by the software) per section (except for ALHA 77307, 2000 points). An object was
133 considered an IOG if its interior was dominated by one olivine crystal. During closer BSE
134 observation, only IOG large enough for SIMS analyses (~15 µm spot; this is also roughly the
135 scale over which the BSE image allowed assignment to IOG in the point counting) were
136 selected for further study. The effective radius of the IOG or the coarsest olivine of each studied
137 chondrule was calculated as the radius of the equal area disk, from back-scattered electron
138 (BSE) images. Cathodoluminescence (CL) imaging of chondrules was performed using (i) an
139 RGB CL-detection unit attached to a field emission gun secondary electron microscope JEOL
140 J7600F at the Service Commun de Microscopie Électronique (SCMEM, Nancy, France) and
141 (ii) a field-gun ZEISS Supra 55 VP equipped with an OPEA catholuminescence device
142 (imaging and spectral) with a high-tech parabolic mirror. Quantitative chemical compositions
143 of olivine grains were obtained using a Cameca SX Five electron microprobe (EMP) at the
144 Université Pierre et Marie Curie (UPMC, Camparis, Paris, France) using a 150 nA focused
145 beam (about ~2 µm in diameter) accelerated to 15 kV. We analysed Na, Mg, Si, Al, K, Ca, Fe,
146 Ti, Cr, and Mn in olivine grains. The high beam current allowed detection limits for silicates of
147 100 ppm for Al, Ca, and Ti, 150 ppm for Mn and Si, and 200 ppm for Na, K, Cr, Fe, and Mg.
148 The PAP software was used for matrix corrections.

149 We measured the oxygen isotopic compositions of chondrule olivine and isolated
150 olivine grains, where CL and EMP had shown fairly homogeneous compositions (typically near

151 the center), with a CAMECA ims 1270 E7 at CRPG-CNRS (Nancy, France). $^{16}\text{O}^-$, $^{17}\text{O}^-$, and
152 $^{18}\text{O}^-$ ions produced by a Cs^+ primary ion beam ($\sim 15\ \mu\text{m}$, $\sim 4\ \text{nA}$) were measured in multi-
153 collection mode with two off-axis Faraday cups (FC) for $^{16,18}\text{O}^-$ and the axial FC for $^{17}\text{O}^-$. The
154 FC had $10^{11}\ \Omega$ amplifiers. To remove $^{16}\text{OH}^-$ interference on the $^{17}\text{O}^-$ peak and to maximize
155 flatness atop the $^{16}\text{O}^-$ and $^{18}\text{O}^-$ peaks, the entrance and exit slits of the central FC were adjusted
156 to obtain mass resolution power of ~ 7000 for $^{17}\text{O}^-$. As an additional safeguard against $^{16}\text{OH}^-$
157 interference, a N_2 trap was used to reduce the pressure in the analysis chamber to $< 5 \times 10^{-9}$
158 mbar. The multicollection FCs were set on exit slit 1 (MRP = 2500). Total measurement times
159 were 240 s (180 s measurement + 60 s pre-sputtering). We used three terrestrial standard
160 materials (San Carlos olivine, magnetite and diopside) to define the instrumental mass
161 fractionation line for the three oxygen isotopes and correct for instrumental mass fractionation
162 for olivine. To obtain good precision on analytical measurements, we analyzed, in order, 4
163 standards, 8 chondrule olivine crystals and 4 standards. Typical count rates obtained on the San
164 Carlos olivine standards were 2.5×10^9 cps for ^{16}O , 1.0×10^6 cps for ^{17}O , and 5.4×10^6 cps for
165 ^{18}O . The isotopic compositions are expressed in standard δ -notation, relative to Vienna standard
166 mean ocean water (VSMOW): $\delta^{18}\text{O} = (^{18}\text{O}/^{16}\text{O})_{\text{sample}} / (^{18}\text{O}/^{16}\text{O})_{\text{VSMOW}} - 1$ and $\delta^{17}\text{O} =$
167 $(^{17}\text{O}/^{16}\text{O})_{\text{sample}} / (^{17}\text{O}/^{16}\text{O})_{\text{VSMOW}} - 1$ (both to be expressed in ‰). 2σ measurement errors,
168 accounting for internal errors on each measurement and the external reproducibility of the
169 standard, were estimated to be $< 1\%$ for $\delta^{18}\text{O}$, $\delta^{17}\text{O}$ and $\Delta^{17}\text{O} \equiv \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$ (representing
170 deviation from the Terrestrial Fractionation Line = TFL).

171

172 **3. Results**

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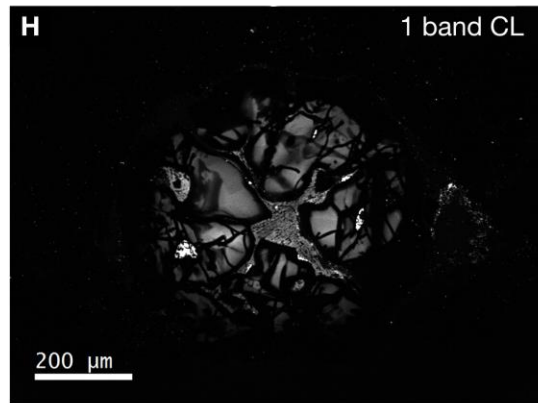
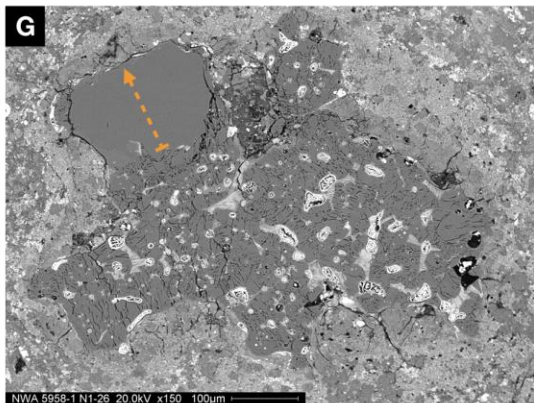
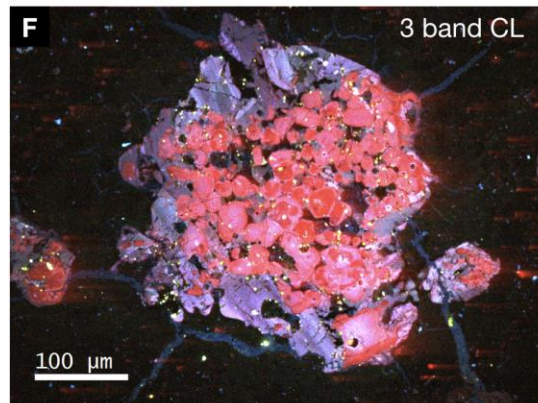
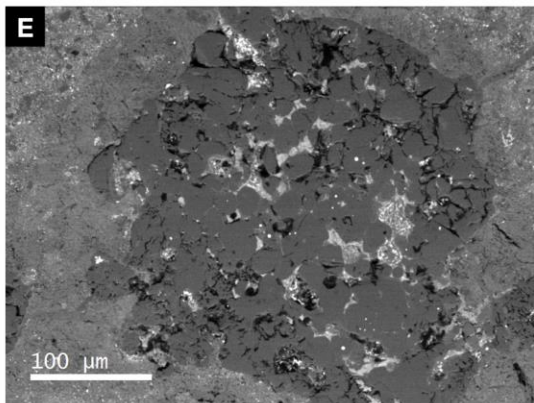
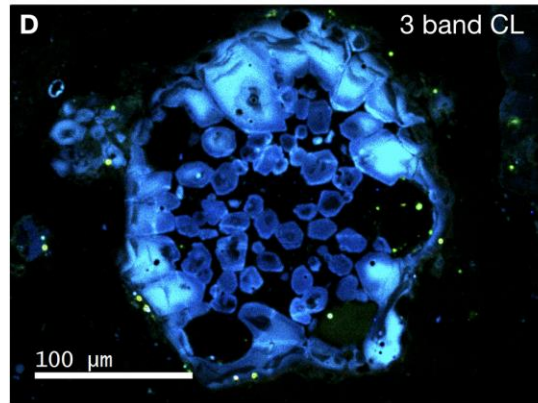
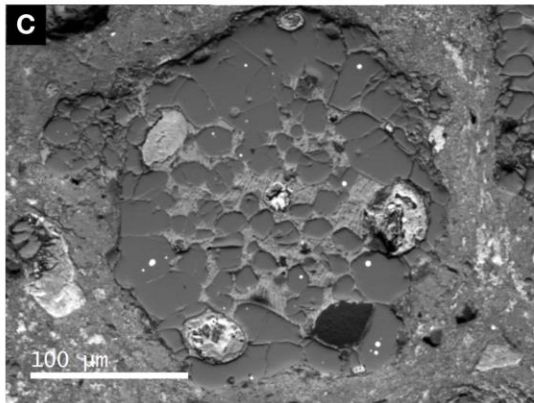
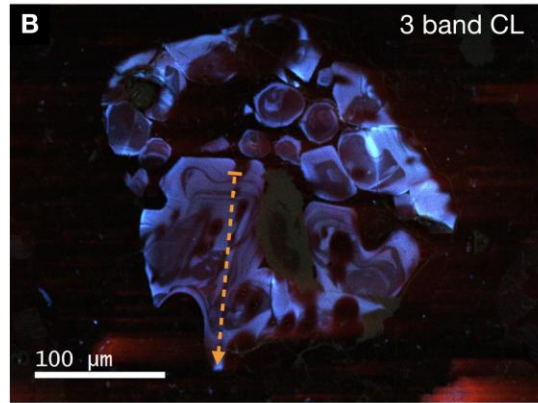
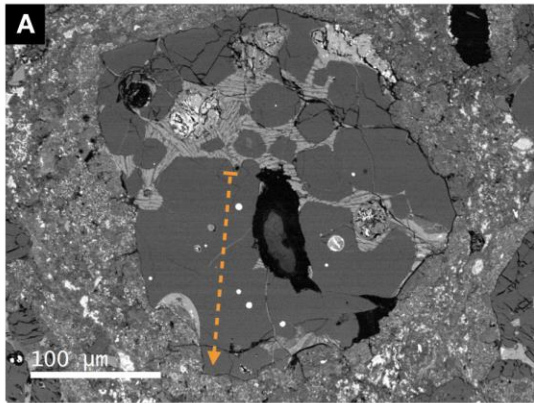
174 **Petrography**

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176 Type I chondrules present the usual mineralogical zoning (Fig. 1) of olivine phenocrysts
177 dominating in the interior and enstatite laths, poikilitically enclosing olivine chadacrysts, near
178 the outside (e.g., Friend et al. (2006). The mesostasis is altered in the CM
179 chondrites. When viewed in CL, interior olivine grains often display dark cores (Fig. 1) that
180 correspond to relict grains for those chondrules investigated by Marrocchi et al. (2018, 2019),
181 surrounded by brighter overgrowths (identified to the *in situ* crystallized host). The “palisadic”

182 olivine grains near the outside of PO chondrules (Marrocchi et al. 2018, 2019; Libourel and
183 Portail 2018) have brightest CL in their inner edge and darken toward the exterior (Fig. 1D).

184



186 **Figure 1:** Back-scattered electron and cathodoluminescence images of representative
187 chondrules (left and right, respectively, unless otherwise noted). Orange dashed lines show
188 location of profiles shown in Fig. 6. A) and B) Chondrule CHA in section NWA 5958-4. The
189 ovoid chondrule contains euhedral olivine grains with little discernible pyroxene and relatively
190 abundant altered mesostasis. Its lower half is occupied by a large olivine apparently embayed
191 near the central hole, and has concentric and oscillatory zoning in blue CL (as the other olivine
192 phenocrysts). Many of the oval dark-CL spots trace metal inclusions. C) and D) PO chondrule
193 CH9 in section NWA 5958-1. Dark-CL spots in interior olivine grains likely correspond to
194 relicts. Brightest blue CL is seen in the coarser palisadic olivine grains but CL intensity declines
195 with some oscillations near the outer edge. A small (~50 μm diameter) PO chondrule is attached
196 to the upper left of the object. E) and F) POP chondrule CHA in NWA 11086. Olivine grains
197 luminesce in red in the chondrule interior, with those closest to the center exhibiting large
198 darker-CL (presumably relict) cores. In the periphery, enstatite oikocrysts have only weak,
199 bluish CL (note that left panel is somewhat more zoomed than right panel). G) Chondrule N1-
200 26 in NWA 5958 (BSE only). This irregularly shaped chondrule is dominated by pyroxene
201 crystals, with $<20 \mu\text{m}$ olivine grains, in addition to altered mesostasis and metal grains.
202 However, one large (0.2 mm across) olivine crystal (analyzed by Jacquet and Marrocchi 2017)
203 is visible on the upper left edge of the chondrule, partly surrounded by a thin layer of enstatite
204 and must have been an IOG fused with the chondrule while still plastic (otherwise it would
205 have been replaced by pyroxene to the same extent as the remainder of the chondrule margins).
206 H) Chondrule CH1 in Allende (CL only) shows coarse, bright CL triangular olivine crystals
207 and mesostasis near the center. It might be a surficial section through the “palisade” of a PO
208 chondrule.

209

Table 1: Modal abundances of high-temperature components in the studied chondrites.

210

Meteorite	NWA 5958	NWA 11086	ALHA 77307	Allende
Classification	C2-ung	CM-an	CO3.0	CV3
<i>Type I chondrules</i>				
Bona fide	20.3	23.2	36.3	31.3
IOG	2.3	3.0	2.6	1.5
<i>Type II chondrules</i>				

Bona fide	1.2	1.4	3.8	1.0
IOG	0.9	1.1	1.6	0.2
<i>Refractory inclusions</i>				
AOA	1.2	0.8	1.6	5.7
CAI	1.0	0.5	0.6	3.8

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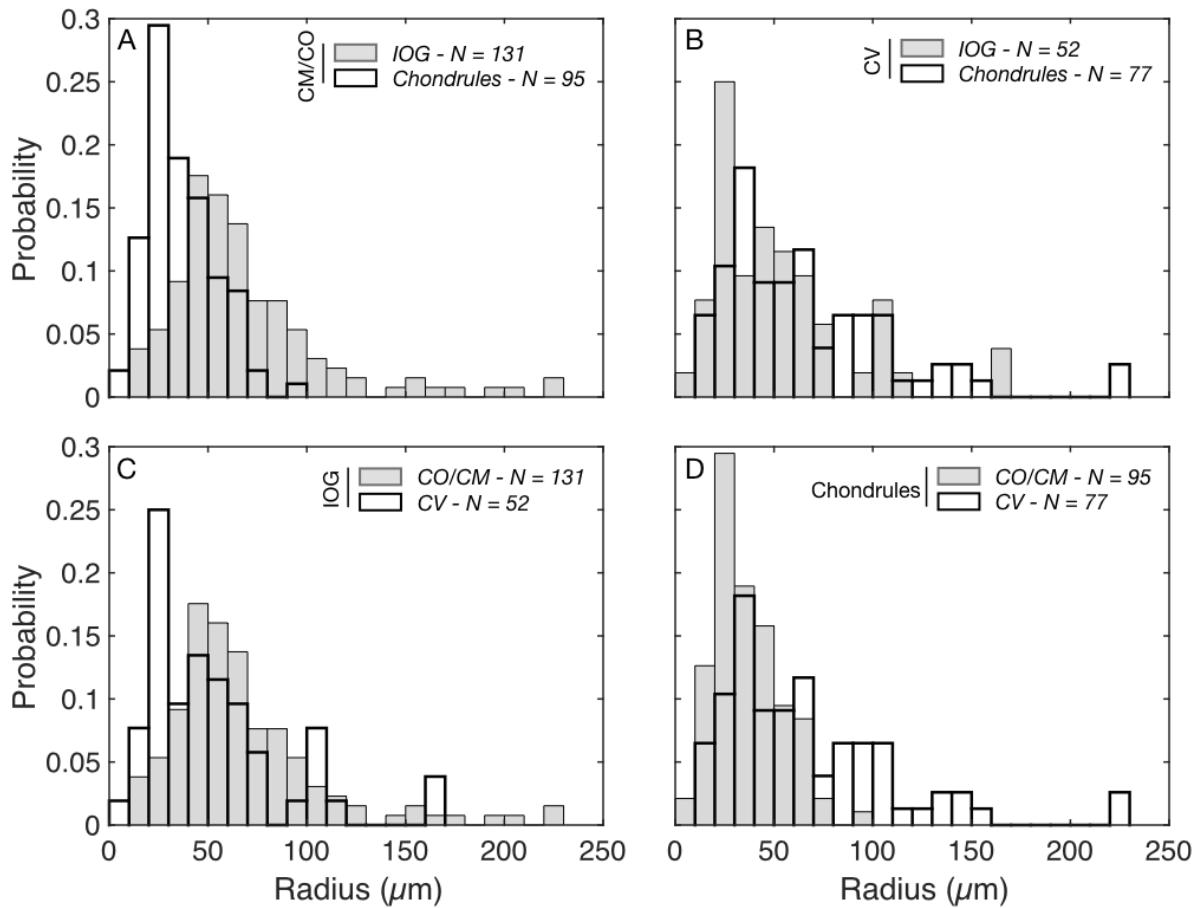
212 Point counting reveals 1.7-4.2 vol% of IOG in the studied carbonaceous chondrites
213 (Table 1). Fuchs et al. (1973) reported 8 vol% of isolated mineral grains in Murchison, about
214 two thirds of which appeared to be olivine; Browning et al. (1996) reported 6 vol% of IOG in
215 Murchison, and down to 1.5 vol% in more altered CM chondrites. The average 22 vol% of
216 “single grain and grain fragments” of Grossman and Olsen (1974) for C2 chondrites must be in
217 error as it equals the *total* high-temperature fraction normally seen in CM chondrites (e.g., Table
218 1; Howard et al. (2011). As for CO3 chondrites, McSween (1977) quoted 8 vol% for IOG. While
219 our numbers thus tend to be systematically lower than past literature estimates based on optical
220 microscopy, we note that Pack et al. (2004) never found more than 0.35 vol% of “refractory
221 forsterites” in carbonaceous chondrites. Since their refractory forsterites (recognized by CL)
222 had CaO > 0.4 wt% and such compositions represent 2/5th of our IOG analyses, and since Pack
223 et al. (2004) only counted the luminescing part of their olivine, this would indicate IOG modal
224 fractions of order 1 vol%. More recently, manual identifications on X-ray maps by Ebel et al.
225 (2016) resulted in an average of 1.16 vol% of “isolated olivine grains or aggregates in matrix”
226 in CO chondrites and 1.37 vol% in Allende. Part of the differences between studies may lie in
227 the ambiguity in discriminating IOG from chondrules. Despite this systematic uncertainty, an
228 order of magnitude of a few percent for IOG modes in carbonaceous chondrites seems overall
229 sound.

230 While most IOG are of type I, it is noteworthy that the type II/type I modal ratio is
231 significantly higher for IOG (0.1-0.6) than for *bona fide* chondrules (0.03-0.11), by a factor of
232 4 to 7. This is qualitatively in line with the histograms of McSween (1977) which indicate
233 values of 2 and 0.8 for this ratio in CM and CO chondrites, respectively, with that of Desnoyers
234 (1980) indicating a ratio of 1.2 for Niger (C2), although those histograms are based on
235 microprobe analyses and not areas (perhaps accounting for the systematic difference). Seven
236 out of the 12 Tagish Lake IOG of Russell et al. (2010) were of type II; same for 25 out of 101

237 CI chondrite IOG compiled by Piralla et al. (2020). This point being made, we henceforth
238 exclusively focus on type I IOG.

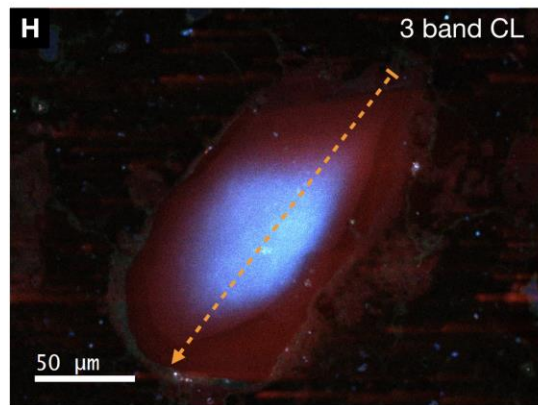
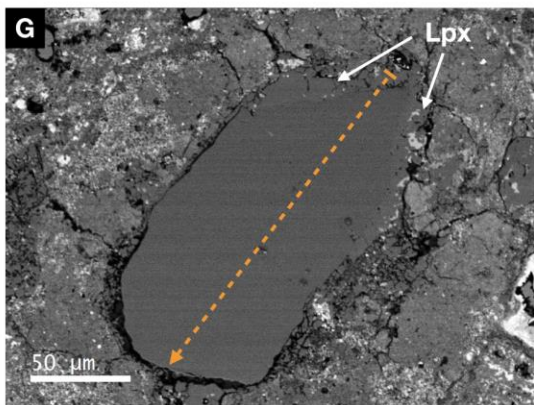
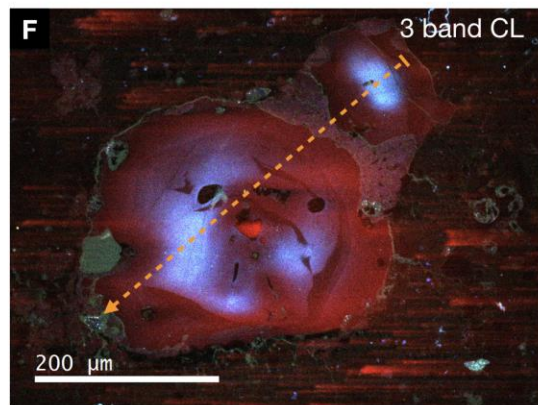
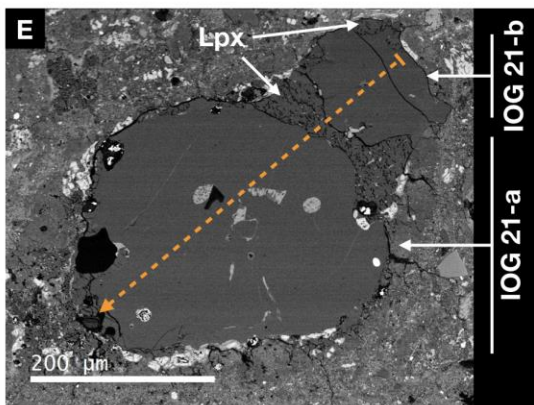
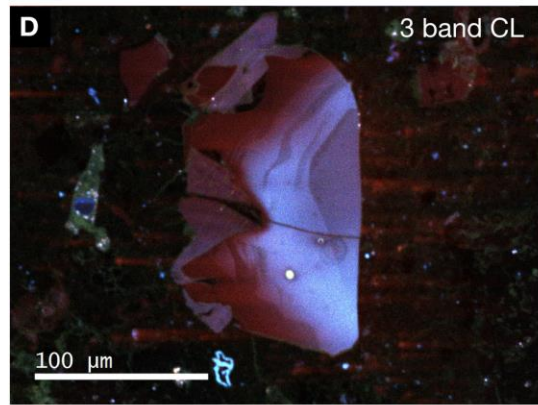
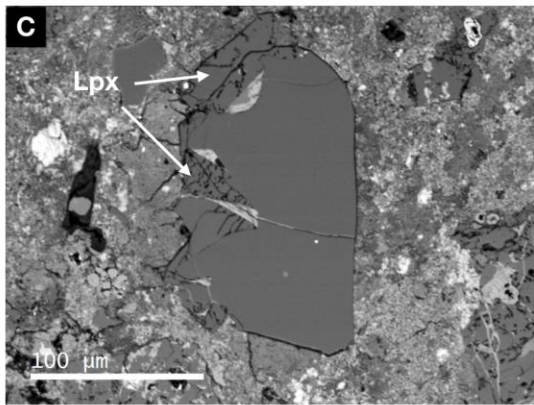
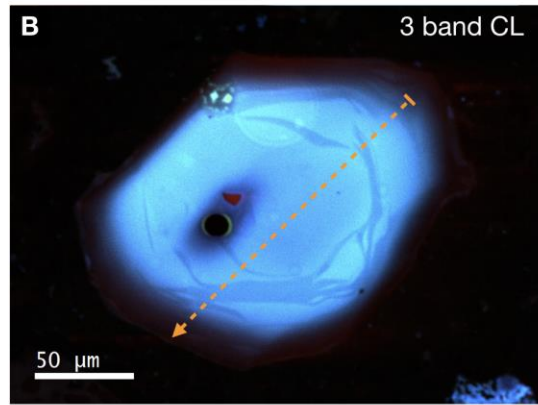
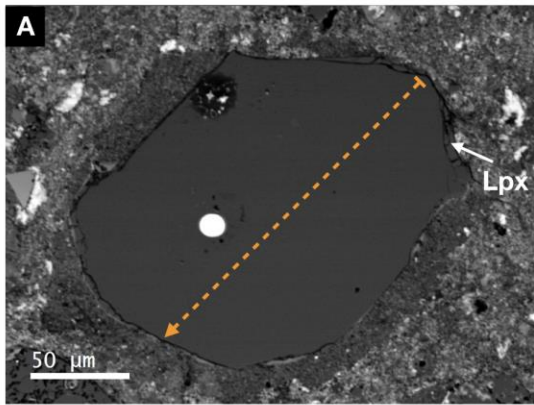
239 As noted by Olsen and Grossman (1978), the IOG tend to be big compared to chondrule
240 phenocrysts (Fig. 2, even though the first bin is cut off by our selection biases). In fact, in CV
241 chondrites, their size distribution is comparable to that of the *coarsest* chondrule phenocrysts;
242 in CM/CO chondrites, more than 90 % of the IOG studied here are even bigger than the mode
243 of the coarsest chondrule phenocrysts (near 30 μm effective radius). About half (21 out of 41¹)
244 of the IOG examined here by BSE/CL and EMP have equidimensional, often
245 euhedral/subhedral shapes, although the others are evidently fragments of larger objects (e.g.
246 Figs. 3C,D; 4G,H). The CL of the former, generally blue in the core, is concentrically zoned,
247 with intensity decreasing toward the margin which may have a redder tint. Discrete darker-CL
248 streaks, partially or entirely surrounding the center, are often superimposed on the background
249 trend, generally paralleling the grain edges. An oscillatory sector zoning is spectacularly visible
250 in NWA 11086 IO12 (Fig. 5); some may be discernible in NWA 5958-1 IO7 as well (Fig. 4F).
251 Despite their name, the IOG are not pure olivine. The olivine may enclose metal grains and
252 glass inclusions, and, when whole, is nearly always surrounded, partly or entirely, by enstatite
253 (from a few microns to tens of microns in thickness), sometimes with mesostasis (although we
254 did not find as large a mesostasis patch as in IOG N6-5 in Fig. 1e of Jacquet and Marrocchi
255 2017) and even further olivine phenocrysts. Enstatite rims, partial or total, are sometimes seen
256 even around fragments (e.g. Fig. 3C,D). We note that Jones (1992) called large olivine grains
257 surrounded by pyroxene in ALHA 77307 “macroporphyritic chondrules”, in contradistinction
258 to “bare” isolated olivine grain, but given the continuum between them, we maintain the name
259 IOG for all those objects. The continuum, in fact, extends to *bona fide* chondrules (e.g. Jacquet
260 and Marrocchi 2017), with some chondrules exhibiting disproportionately large olivine crystals
261 (e.g. Fig. 1A,B; chondrule N5-21 with three aligned coarse olivine grains in Fig. 2b of Jacquet
262 et al. 2016), not to mention chondrule-IOG or IOG-IOG compounds (see e.g. Fig. 1G, 3E,F).
263 Overall, this continuum makes the assignment of objects to IOG rather than chondrules
264 relatively subjective and must have contributed to the scatter in IOG modes in the literature
265 discussed above. We recall that we have considered here an object to be an IOG when its interior
266 was dominated by one olivine crystal.

¹ Fragments may be numerically more numerous for smaller sizes which we have not selected. Still, it may not affect the modal ratio much (here the studied “whole” IOG outweigh fragments also in this respect, with a total area of 0.74 mm² vs. 0.59 mm²).



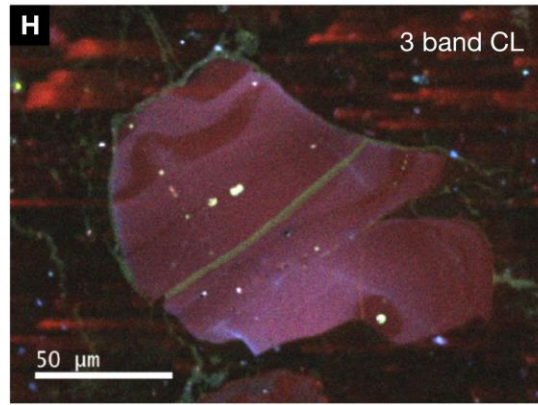
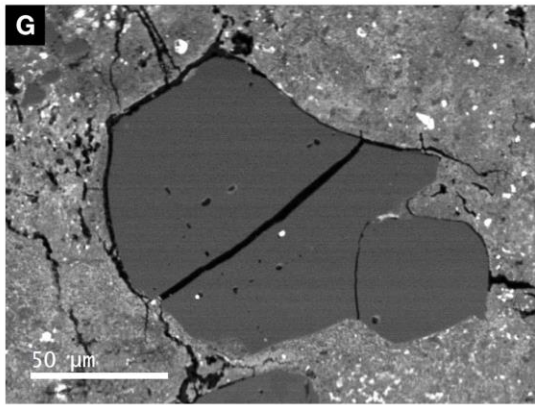
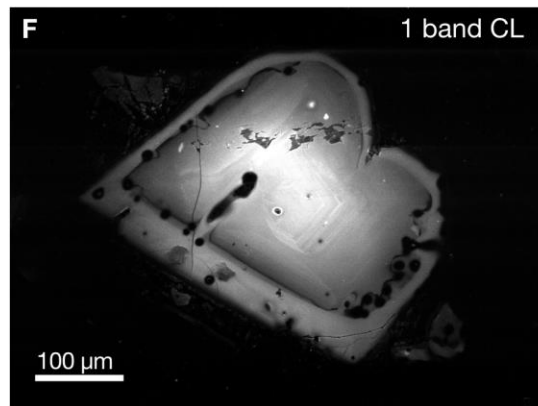
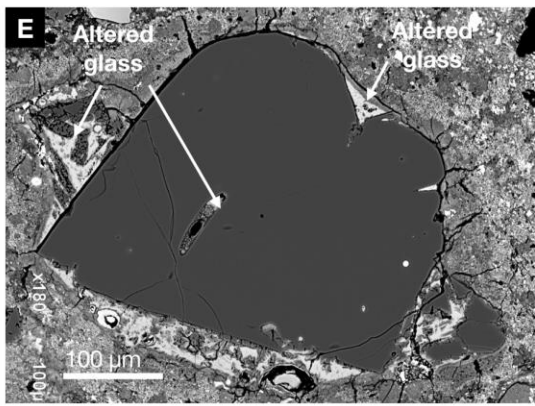
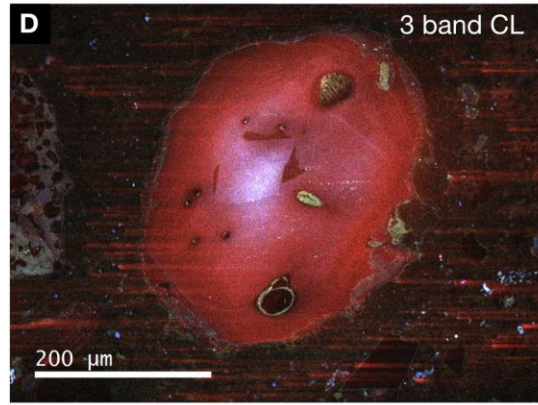
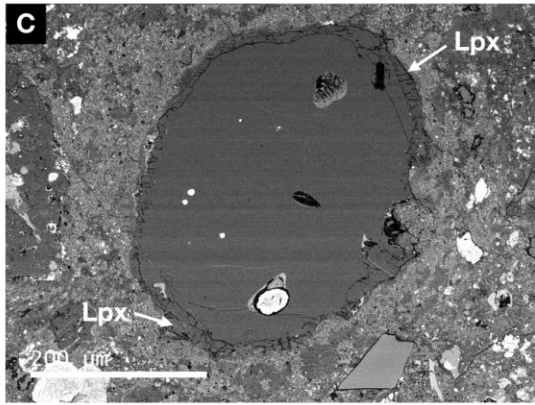
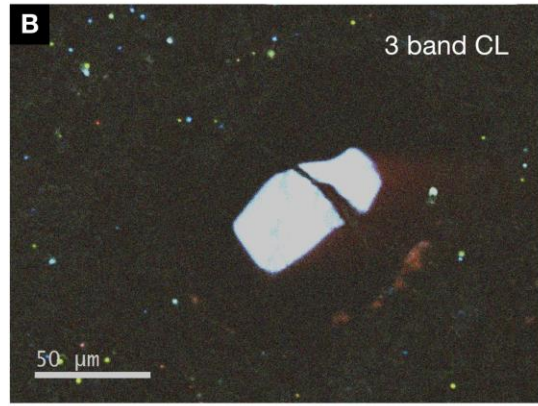
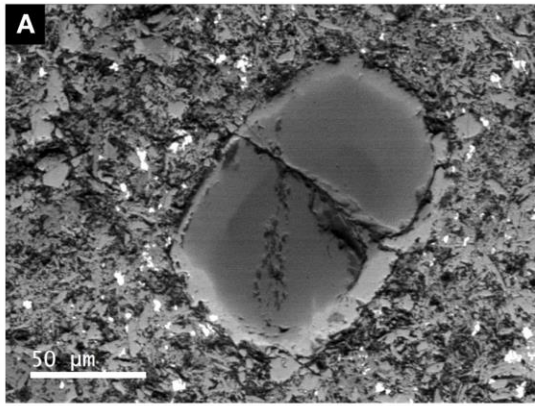
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268 **Figure 2:** Histograms of sizes of IOG and coarsest olivine grains of chondrules. A) and B) show
 269 data for CO/CM and CV chondrites, respectively, for intra-clan comparison, while C) and D)
 270 allow inter-clan comparison (for IOG and chondrules, respectively).

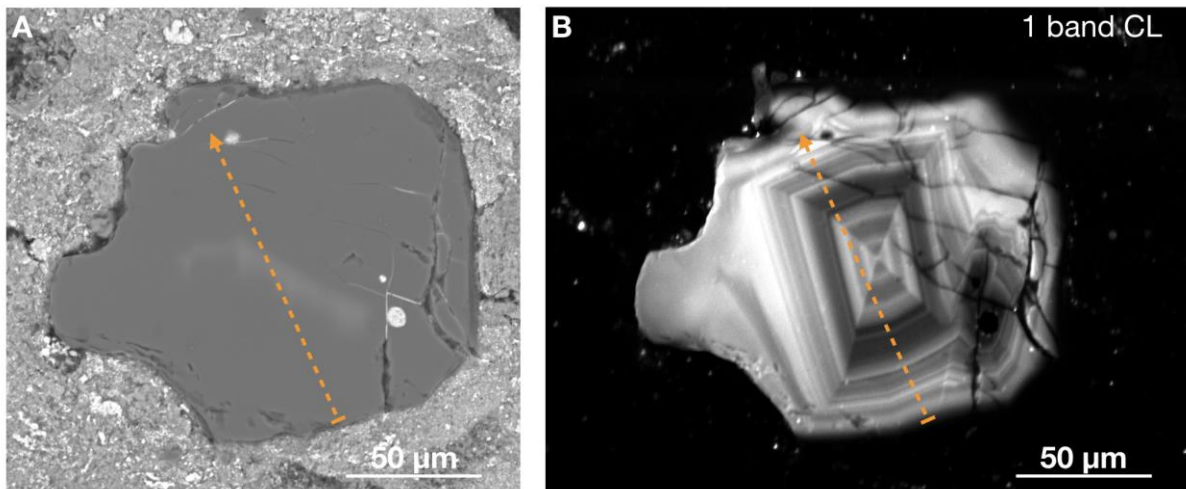


272 **Figure 3:** Back-scattered electron (left) and cathodoluminescence (right) images of
273 representative isolated olivine grains. “Lpx” indicates enstatite and orange dashed lines locate
274 the profiles shown in Fig. 6. A) and B) NWA 5958-4 IO5. This subhedral grain has a thin (<5
275 μm) enstatite layer. The olivine has bright blue CL over its interior except its outermost
276 reddish/black-CL 10 μm . Somewhat darker blue-CL streaks occur concentrically. A metal grain
277 is surrounded by a dark-CL ellipsoidal halo, perhaps due to anisotropic diffusion of Fe^{2+}
278 therefrom. C) and D) NWA 5958-4 IO13. This IOG presents a ~ 20 μm thick pyroxene +
279 (altered) mesostasis layer on the left, with an irregular boundary with the olivine. The latter
280 seems abruptly cut off on the right side. CL is zoned from right (blue) to left (reddish), and
281 would have been roughly concentric around a center located outside the present right edge. E)
282 and F) NWA 5958-4 IO21 is a compound between two IOG, tied by a pyroxene neck which
283 extends to thin layers around the peripheries of the two olivine grains. The compound must
284 have formed by collision between two IOG which had independently acquired a pyroxene
285 margin, as otherwise the olivine would have been continuous across the two lobes. The CL is
286 zoned from bright blue cores to reddish margins. The zoning of the larger IOG (NWA 5958-4
287 IO21a) is complicated by dark-CL streaks and altered mesostasis inclusions, one with spinel.
288 G) and H) NWA 5958-4 IO36. This elongated euhedral olivine grain is CL-zoned (from bright
289 blue to red) parallel to the edge and is surrounded by enstatite, thickest around the upper acute
290 angle.

291



293 **Figure 4:** Back-scattered electron (left) and cathodoluminescence (right) images of additional
 294 representative isolated olivine grains. “Lpx” indicates enstatite. A) and B) Allende IO19. This
 295 elongated subhedral grain shows a bright blue CL in its forsteritic core, mantled without
 296 transition by more ferroan (dark-CL) olivine itself surrounded by a BSE-bright more fayalitic
 297 outer layer betraying influx of Fe during metamorphism. C) and D) NWA 5958-4 IO22. This
 298 oval olivine grain shows red CL, brightest near the center, and is surrounded by a continuous
 299 10 μm -thick layer of enstatite (darker than olivine in BSE, pink in CL). Altered glass inclusions
 300 and one big altered and small, unaltered, metal grains also occur inside the olivine. E) and F)
 301 NWA 5958-1 IO21 (the CL image is here a one-band panchromatic image so as to leave the
 302 details discernible). This subhedral olivine grain has a bright-CL margin separated from a core
 303 with oscillatory zoning by a boundary strikingly parallel to the edge of the whole grain. The
 304 olivine is embedded in altered mesostasis (with pyroxene phenocrysts), with two discrete
 305 patches on the left and the right (the latter with olivine crystals), possibly attached during
 306 cooling. G) and H) NWA 5958-4 IO16. This is clearly a fragment. The darker-CL streaks in
 307 this red-CL object show no concentric arrangement.



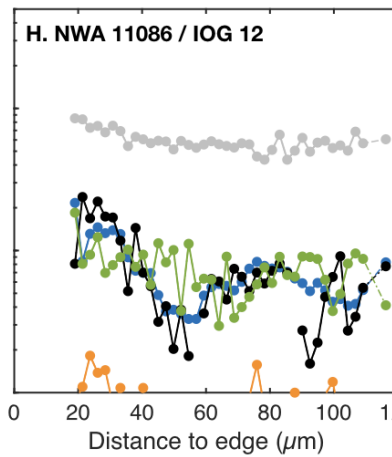
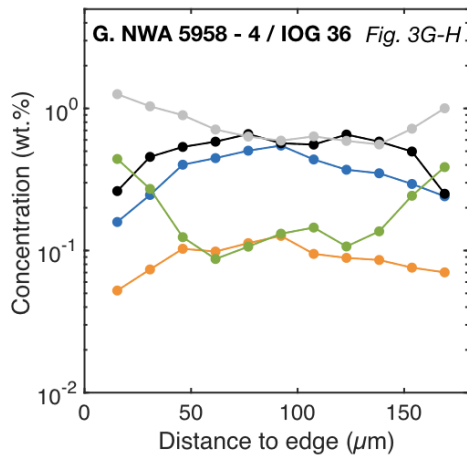
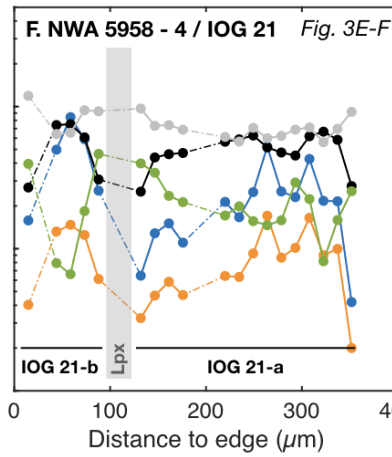
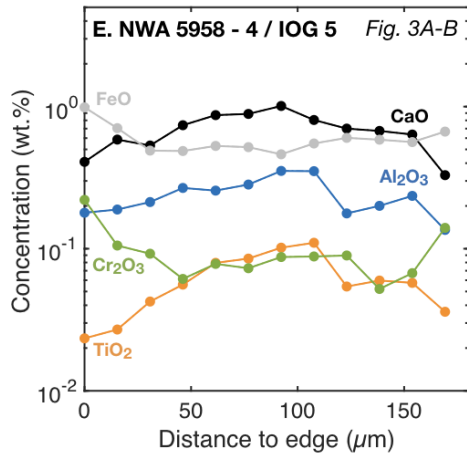
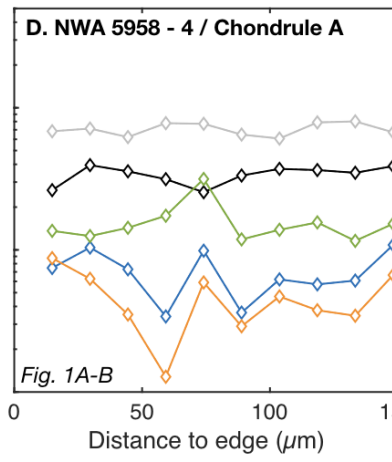
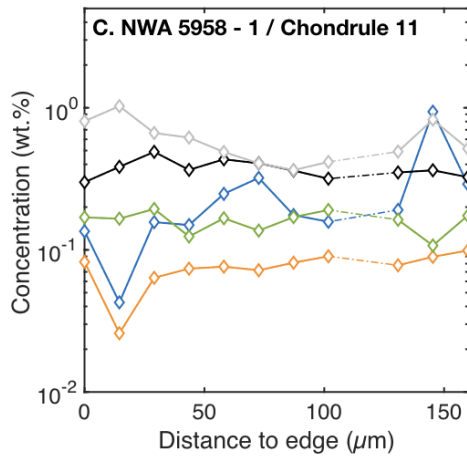
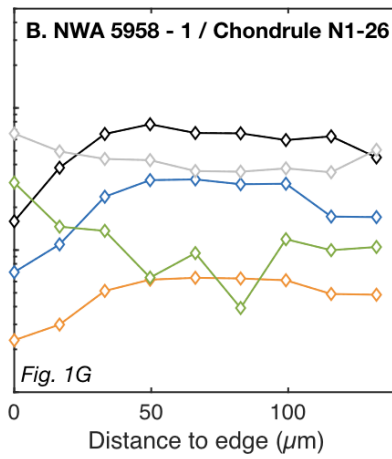
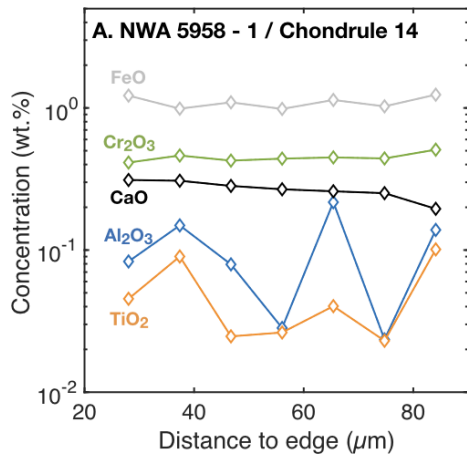
309 **Figure 5:** Back-scattered electron (A) and cathodoluminescence (B) images of isolated olivine
 310 grain NWA 11086 IO12, with dashed line indicating the location of the profile shown in Fig.
 311 6H. The CL shows spectacular oscillatory sector zoning in a central rectangular area. On the
 312 lower right, a darker-CL irregularly shaped angular patch seems to indicate an independent
 313 olivine grain (enclosing two black-CL metal grains) welded together with the previous one,
 314 with subsequent layers with oscillatory zoning swathing both. The irregular outlines may point
 315 to fragmentation around the core of the parent olivine.

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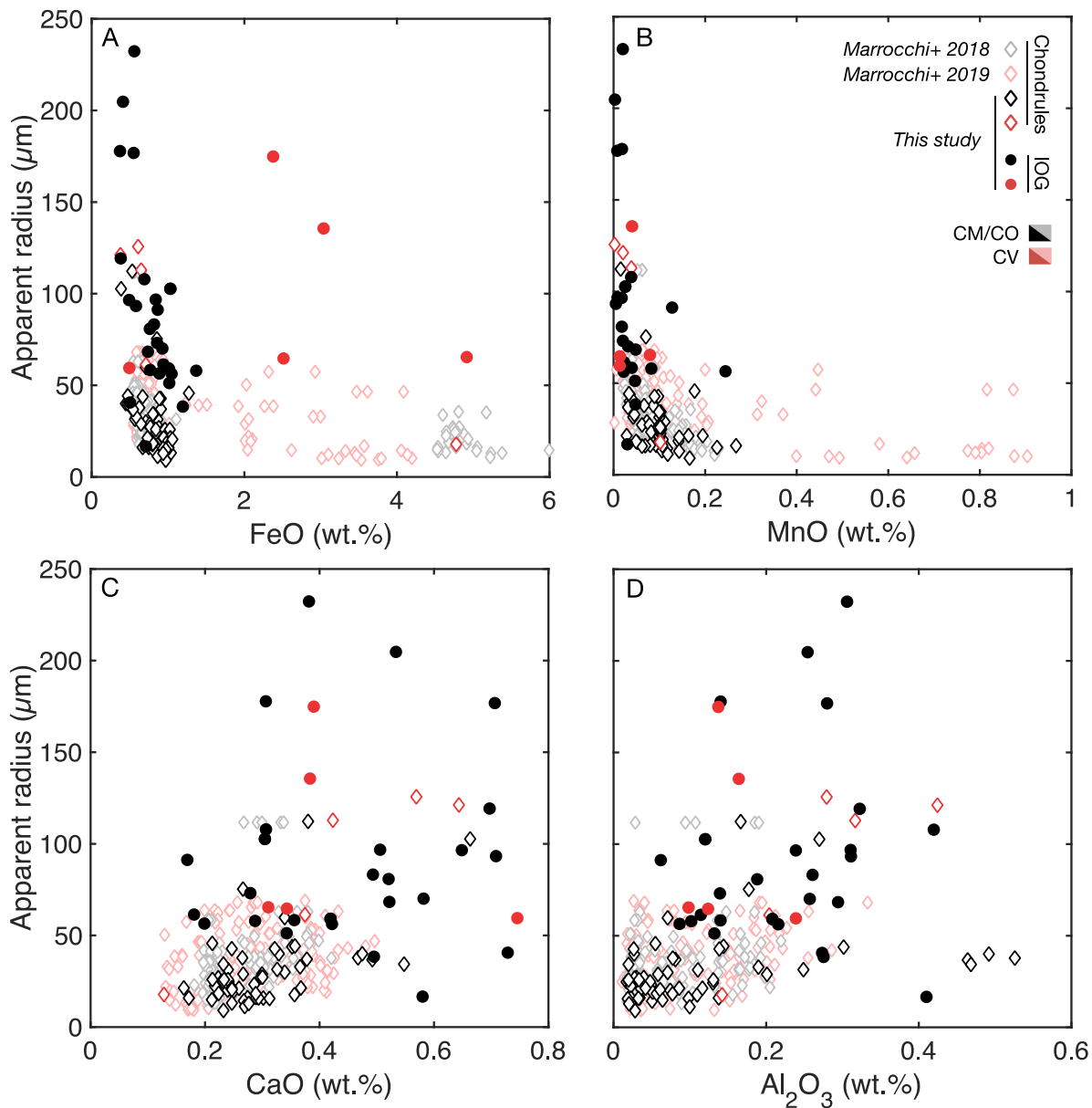
319 Individual profile location and analyses are shown in the Electronic Annex and some
320 representative ones are illustrated in Fig. 6. The CL zoning in IOG olivine is reflected by EMPA
321 traverses, with Ca, Al, Ti decreasing from core to rim, whereas Fe, Mn, Cr symmetrically
322 increase outward. Thus the zoning is basically of decreasing “refractoriness” toward the margin
323 (Fig. 6; Jones 1992; Jacquet and Marrocchi 2017). This is seen in the coarsest chondrule olivine
324 grains as well (more asymmetrically for the palisadic grains, with 0.2-0.5 wt% CaO; Marrocchi
325 et al. 2018) although many have flatter minor element profiles (Fig. 6A-D). In general,
326 excluding obvious fragments, flat profiles are associated with uniformly low CaO (<0.4 wt%)
327 abundances. While Al and Ti roughly parallel Ca, they may suffer abrupt drops or peaks (e.g.
328 Fig. 6A,D) which correspond to oscillations seen in CL (whose lengthscales may be shorter
329 than the profile steps) or occasional relict grains (mostly for chondrules; Marrocchi et al. 2018).
330 This is consistent with the contention of Libourel and Portail (2018) that CL intensity is largely
331 controlled by Al (in the absence of sufficient amounts of the CL quencher Fe). Only in NWA
332 11086 IO12 (Fig. 6H) does Ca show oscillations unattenuated relative to Al and Ti.

333 In the biplots of this paper (viz. Fig. 7, 8), the IOG will be represented by their apparent core
334 composition. CaO spans 0.16-0.89 wt% in IOG and 0.05-0.66 wt% in chondrule coarsest
335 olivine (with averages of 0.44 vs. 0.30 wt%); Al₂O₃ spans 0.06-0.42 wt% in IOG and chondrule
336 coarsest olivine 0.02-0.53 wt% (with averages of 0.20 vs. 0.12 wt%). Crystal size seems to
337 correlate negatively with Fe, Mn but the positive correlation with Ca and Al noted by Jacquet
338 and Marrocchi (2017) is very rough (Fig. 7). This may be to some extent a 2D sectioning artifact
339 as more equatorial sectioning of the grains (providing the widest areas) would pass closest to
340 the actual refractory core. Excluding Allende, whose olivine FeO contents up to 5 wt% are
341 obviously secondary, the IOG have low FeO contents (0.35-1.4 wt%) anticorrelated with CaO,
342 similar to chondrule phenocrysts in this compositional range (Fig. 8).



344 **Figure 6:** Minor element profiles in chondrule olivine (A-D) and IOG (E-H, with profile
 345 positions in the indicated figures). Recall that NWA 5958-4 IOG21 is a compound object where
 346 the profile transects two successive (formerly isolated) olivine grains (Fig. 3E,F).

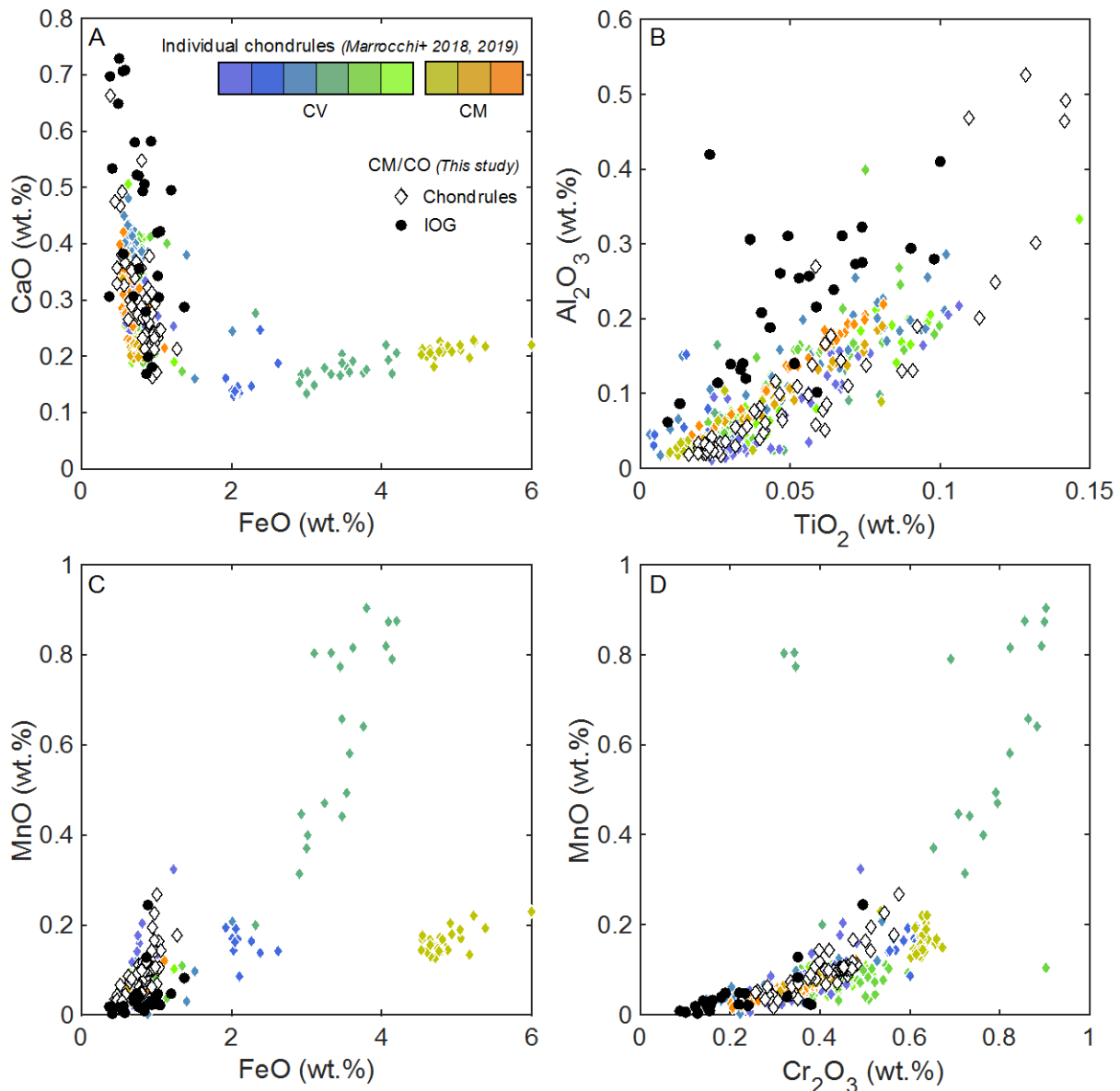
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349 **Figure 7:** Olivine size vs. minor element content (A: FeO; B: MnO; C: CaO; D: Al₂O₃) in IOG
 350 and chondrule coarsest olivine. Data for all analyzed chondrule phenocrysts in Marrocchi et al.
 351 (2018, 2019) are also shown.

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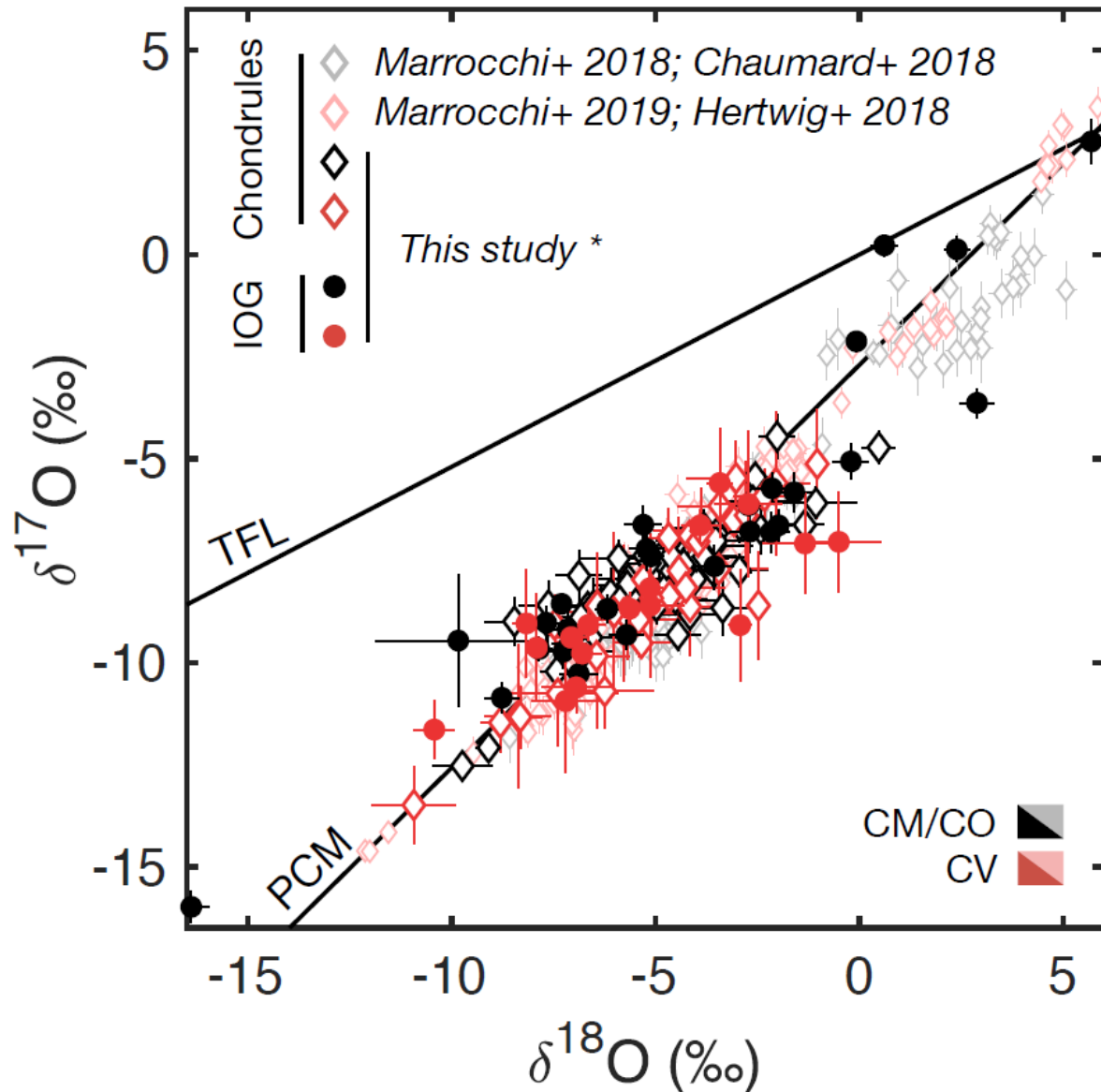
354 **Figure 8:** Minor element biplots of IOG, chondrule coarsest olivine of this study and chondrule
 355 olivine (keyed by chondrule) from Marrocchi et al. (2018, 2019). A) CaO vs. FeO. The
 356 correlation is negative for FeO below ~1.5 wt% and positive afterward. Individual chondrules
 357 plot in only one of these trends. IOG all belong to the former. B) Al₂O₃ vs. TiO₂. C) MnO vs.
 358 FeO. D) MnO vs Cr₂O₃.

359

360 The oxygen isotopic compositions of chondrule olivine and IOG plot along the Primitive
 361 Chondrule Mineral (PCM; Fig. 9) line of Ushikubo et al. (2012). When analyzed multiple times,
 362 the IOG appear generally isotopically homogeneous, with the exception of NWA 11086 IO22
 363 (with $\Delta^{17}\text{O}$ of -0.18 ± 0.60 ‰ and -5.39 ± 0.60 ‰), but the number of analyses per object is

364 usually small. $\Delta^{17}\text{O}$ ranges from -12.65 to -0.2 ‰ for chondrules and -7.46 to 0.84 ‰ for IOG.
365 The distribution (Fig. 10) is fairly similar to the chondrule host (i.e., non-relict) olivine grains
366 of Marrocchi et al. (2019), with a minor ^{16}O -poor population above the
367 ~ -4 ‰ hiatus noted by Ushikubo et al. (2012). The four IOG in this range are not more fayalitic
368 than the others, unlike the trend shown by type I chondrules in several studies (e.g. Ushikubo
369 et al. 2012; Schrader et al. 2013; Tenner et al. 2013, 2015) but similar to the Jacquet and
370 Marrocchi (2017; see their Fig. 8a) data for NWA 5958. Nevertheless, high refractory element
371 contents (e.g. CaO contents above 0.4 wt%)—corresponding to the refractory forsterites of
372 Steele (1988)—are restricted to ^{16}O -rich IOG or *bona fide* chondrules with $\Delta^{17}\text{O} < -4$ ‰ (Fig.
373 11; Libourel and Chaussidon 2011; Jacquet and Marrocchi 2017; Marrocchi et al. 2019).

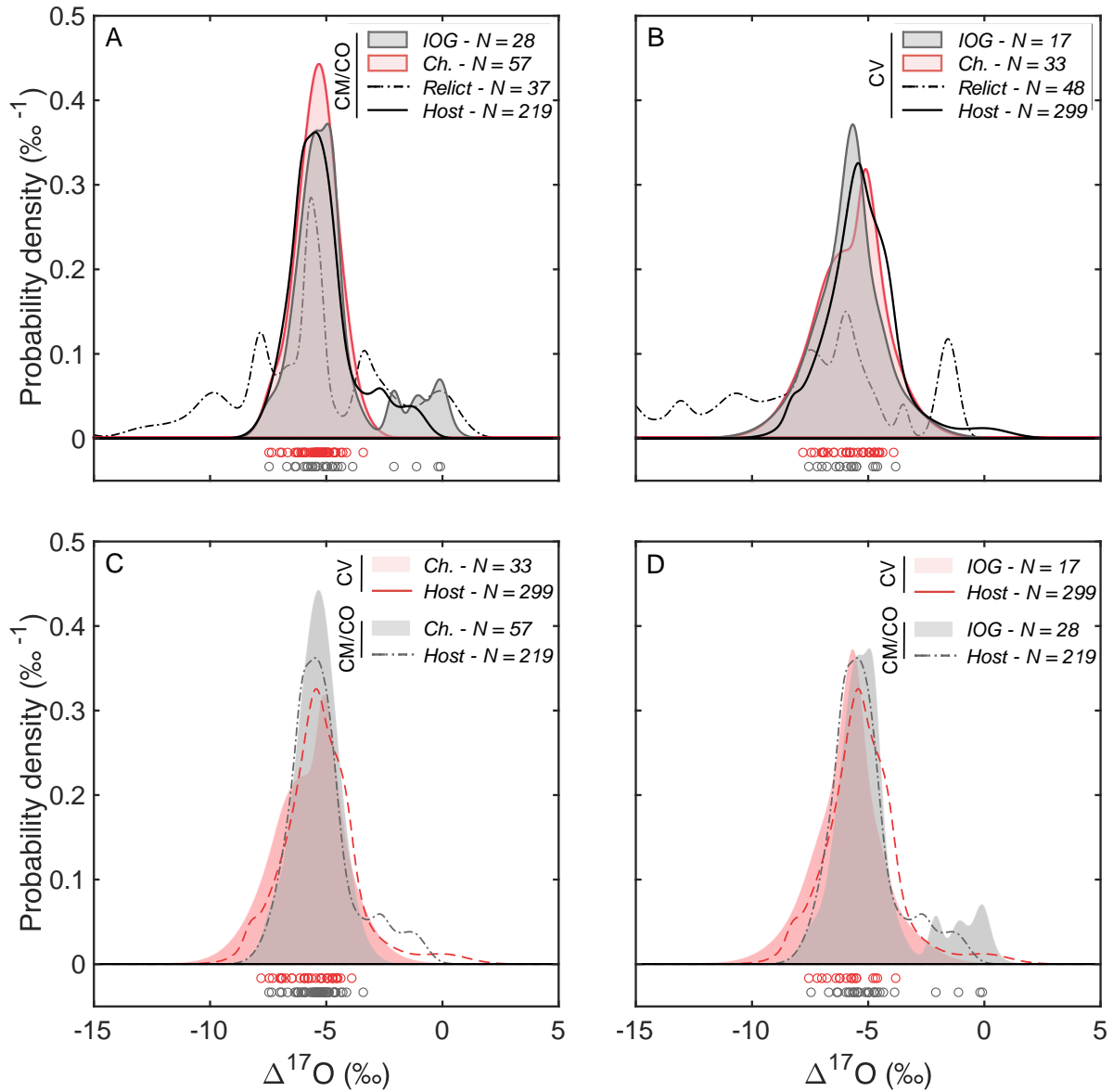
374 The isotopic, chemical and geometrical data for all analyzed objects are shown in the
375 Electronic Annex.



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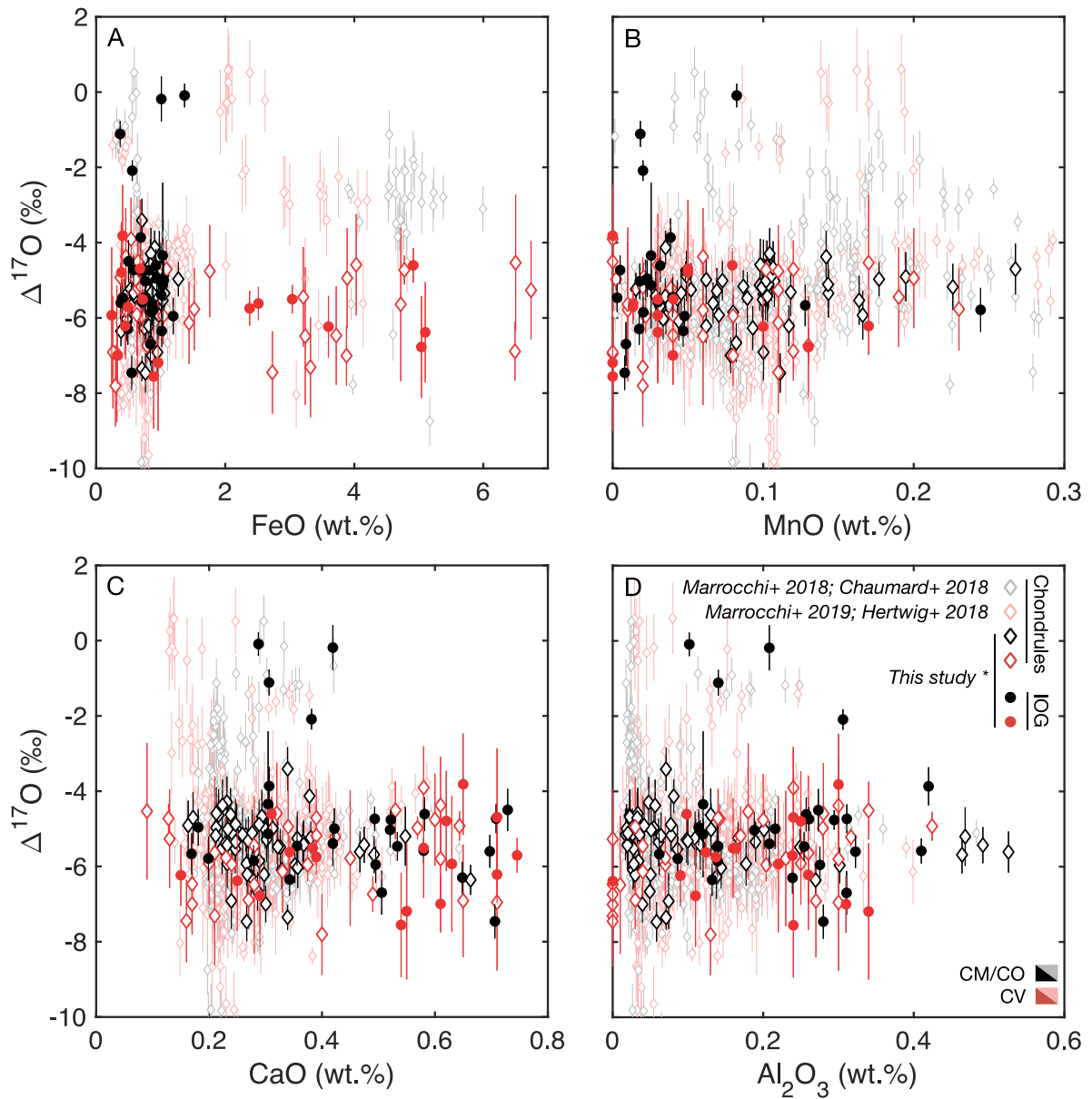
377 **Figure 9:** Three-isotope plot of olivine grains in this study, including CV data of Libourel and
 378 Chaussidon (2011), with chondrule literature data from Marrocchi et al (2018, 2019), Chaumard
 379 et al. (2018), Hertwig et al. (2018). The Primitive Chondrule Mineral (PCM) line of Ushikubo
 380 et al. (2012) is also shown,

381



382

383 **Figure 10:** Probability density function of $\Delta^{17}\text{O}$ for IOG and coarsest chondrule olivine,
 384 compared to host and relict data compiled by Marrocchi et al. (2019). A) and B) compare data
 385 within chondrite clans (CM/CO and CV respectively) and C) and D) compare chondrule and
 386 IOG data across the clans.



387

388 **Figure 11:** Oxygen isotopes vs. minor elements for IOG and coarsest olivine grains, including
 389 CV data by Libourel and Chaussidon (2011), along with host olivine data from the Marrocchi
 390 et al. (2019) compilation. A) $\Delta^{17}\text{O}$ vs. FeO. B) $\Delta^{17}\text{O}$ vs. MnO. C) $\Delta^{17}\text{O}$ vs. CaO. D) $\Delta^{17}\text{O}$ vs.
 391 Al_2O_3 .

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4. Discussion

4.1 Genetic link between isolated olivine grains and chondrules

The general overlap in $\Delta^{17}\text{O}$ (Fig. 10) between IOG and chondrule host olivine, as well as in minor elements, indicates, as in previous work (McSween 1977; Desnoyers 1980; Jones 1992; Jones et al. 2000), that IOG are closely linked to chondrules rather than early nebular condensates such as AOAs. The presence of mesostasis is also evidence for an igneous origin incompatible with gas-solid condensation (e.g. McSween 1977; Richardson and McSween 1978). Other phases associated with olivine (metal, occasional companion olivine crystals, pyroxene) are also most analogous to chondrule petrography; with IOG CL being comparable to that of the coarsest olivine in chondrules (Fig. 1). We may also recall the REE fractionation measured in IOG olivine (Pack et al. 2005; Jacquet et al. 2012; Jacquet and Marrocchi 2017) which is at variance with the flatter patterns in AOA olivine which formed by condensation (Ruzicka et al. (; Jacquet and Marrocchi 2017). The oscillatory sector zoning shown by the exceptional NWA 11086 IO12 (Fig. 5) also suggests departure from equilibrium, but the analogy with fast diffusion-controlled grown planetary igneous olivine (Welsch et al. (2014 is consistent with an igneous character. Adding to this the textural continuum between IOG and *bona fide* chondrules, we contend, in agreement with the latest literature (e.g. Jones et al. 2000; Pack et al. 2005; Russell et al. 2010; Ushikubo et al. 2012), that IOG are genetically related to chondrules.

4.2 Isolated olivine grains formed hot

It does not however follow that IOG result from the *cold* fragmentation of chondrules. Olsen and Grossman (1978) had already voiced skepticism about the feasibility to cleanly separate olivine phenocrysts from the mesostasis. While Richardson and McSween (1978) had invoked the friability of the altered mesostasis at least for CM chondrite chondrules, that alteration is now widely believed to have occurred on the parent body (Brearley (2014, too late for chondrule-chondrule collisions, and would not be relevant for the least altered chondrites such as ALH 77307. The large size of IOG compared to chondrule phenocrysts is also problematic if the former are fragments of the latter (e.g., Olsen and Grossman 1978). The

427 larger IOG/chondrule ratio for type II compared to type I objects in carbonaceous chondrites
428 would not be understandable if the chondrules were fragmented after being mixed together in
429 the general protoplanetary disk.

430 The euhedral or equidimensional morphology of many IOG and their frequent
431 concentric CL zoning patterns indicate that those feature unbroken olivine. Although, again,
432 this study focuses on type I IOG, we recall that the ferroan IOG N6-7 in NWA 5958 exhibited
433 oscillatory zoning visible in BSE (Jacquet et al. 2016; Jacquet and Marrocchi 2017). The
434 frequent presence of near-complete enstatite margins (e.g. Fig. 3G,H; Fig. 4C,D) in type I IOG
435 is comparable to those shown by whole chondrules. These are ascribed to the influx of SiO into
436 residual melts, promoting pyroxene crystallization at the expense of olivine (e.g., Libourel et al.
437 (; Chaussidon et al. (; Friend et al. (; Barosch et al. 2019). This indicates that the IOG
438 experienced such influx as independent objects. In fact, prior to enstatite precipitation, the
439 decrease of refractory incompatible elements toward IOG olivine margins reflected by the CL
440 zoning also suggests condensation of Mg and SiO into the parent melt, inducing dilution of
441 these elements, as advocated by Jacquet and Marrocchi (2017), as well as Marrocchi et al.
442 (2018, 2019) and Libourel and Portail (2018) for the palisadic olivine. More volatile Fe, Mn,
443 Cr would have recondensed contributing to their outward increase in the olivine. Perhaps in
444 some cases—for those IOG which really were bare euhedral olivine grains—the late olivine
445 condensed directly, without any intermediate liquid, which may account for the patterned
446 surfaces which Olsen and Grossman (1974) compared to terrestrial sublimate olivine. In such
447 cases, glass inclusions would of course have to date back to the time the olivine was still hosted
448 in the partially molten chondrule. Thus IOG experienced gas-melt (or gas-solid) interaction as
449 isolated objects. In that sense, condensation played a role, after all, in IOG formation.

450 Further evidence for a hot formation of IOG is afforded by compound objects, either
451 between IOG (e.g. Fig. 3E,F) or with chondrules (e.g. Fig. 1G), which require collisions in a
452 plastic state (Gooding and Keil (; Akaki and Nakamura 2005). Some *bona fide* chondrules with
453 coarse olivine phenocrysts may have engulfed them during cooling—that is, be “enveloping”
454 compound chondrules in the parlance of Wasson et al. (1995). To be sure, many IOG *are* mere
455 misshapen olivine (\pm mesostasis, pyroxene) debris, but even those may sometimes be fragments
456 of hot-formed IOGs (e.g., the IOG “half” NWA 5958-4 IO13; Fig. 3C,D), or have been
457 fragmented hot, if enstatite has surrounded them since (e.g. NWA 5958-4 IO1 in the Electronic
458 Annex). We note that Jones (1992) did envision isolated olivine grain formation from

459 fragmentation of “macroporphyrritic chondrules”, although the formation of the latter was left
460 unexplained.

461 We conclude from enstatite rimming and concentric CL zoning in many IOG (Fig. 3-4)
462 that they formed as stand-alone high-temperature objects. In that sense, we deem it warranted
463 to also call them “chondrules”(sensu lato, e.g. Jacquet et al. 2012; Jacquet and Marrocchi 2017),
464 whenever the distinction is not of interest, as they are part of the same continuum of objects
465 (with the word “chondrule” being already applied to a diverse suite of objects: ferromagnesian,
466 aluminium-rich, chromite-rich, etc. see e.g. Connolly and Desch 2004). This does not exclude
467 the possibility that some isolated olivine grains in chondrites have different origins, e.g. as AOA
468 or a forsterite-bearing CAI fragment, although no evidence (e.g. isotopic) to that effect has been
469 found for those of this study. To avoid any confusion, we remind the reader that the expression
470 “*bona fide* chondrule” in this paper continues to refer to a non-IOG chondrule.

471

472 4.3 Isolated olivine grains as chondrule splashes

473

474 Since IOG are (by definition) olivine-dominated, they cannot be of chondritic
475 composition, if only because of the Mg/Si (atomic) ratio (2 in endmember forsterite vs. 1.02 for
476 solar abundances, Lodders (2003). This undercuts the Jacquet and Marrocchi (2017) effort to
477 explain the IOG olivine incompatible element enrichment relative to “mainstream” type I
478 chondrules in terms of varying olivine subtraction from a given parental melt. Most likely, the
479 IOG precursors oversampled olivine. We must thus attempt to identify their nature.

480 Given the general analogy of chondrule relict grains with AOA olivine (Marrocchi et al.
481 2018, 2019), a first candidate would be simply AOAs. However, melting experiments of AOA
482 analogs by Whattam et al. (2010) produced porphyritic textures. Also bulk AOA composition,
483 although deficient in silica relative to type I chondrules, are closer to the latter than IOG
484 (because of their CAI-like inclusions). Perhaps, though, a fragment of the olivine portion of a
485 compact AOA (the dominant AOA texture in CM and related chondrites; Jacquet et al. (; Krot
486 et al. (2004) would do. However, the (polycrystalline) olivine parts rarely extend beyond a few
487 tens of μm in extent (e.g., Krot et al. (2004), at variance with the large IOG sizes. Furthermore,
488 the abundance of AOAs overall in the studied chondrites is too low (~ 1 vol%) to account for
489 the abundances of magnesian IOG (2-3 vol%). Indeed, the substantial matrix fraction in

490 carbonaceous chondrites indicates that only a minority of their matter, and in particular of their
491 AOA, has been converted in chondrules (Marrocchi et al. 2019).

492 A second precursor candidate that comes to mind would be olivine phenocrysts from
493 previous generations of chondrules. Now, if the chondrule/matrix ratio of carbonaceous
494 chondrites, in particular CM-related chondrites, can be again taken as a measure of the extent
495 of chondrule formation, chondrule-to-chondrule recycling must have been limited. Assuming
496 chondrule-forming events were independent, Marrocchi et al. (2019) calculated that a fraction
497 of 12 % of chondrule material was inherited from earlier chondrules (*sensu lato*) in NWA 5958
498 (the same formula would yield 19 % for Allende and 26 % for ALH 77307). At face value, this
499 would, when multiplied by the chondrule abundance, approximate the modal abundance of
500 IOG. However, it is unlikely that the recycled chondrules were preferentially in the form of
501 free-floating olivine phenocrysts when they were remelted. This is because, again,
502 fragmentation would not necessarily liberate bare olivine phenocryst (Olsen and Grossman
503 1978), and presumably, before the purported second chondrule-forming event, such olivine
504 grains would have mixed with chondritic dust, such as rim high-temperature objects in CM
505 chondrites (Metzler et al. (1992, erasing their olivine-dominated character most of the time.

506 Separating olivine crystals from the mesostasis would be conceivable if the mesostasis
507 were *liquid*, that is during chondrule formation². The crystals and liquid could separate (to some
508 extent) owing to buoyancy during sudden accelerations. Such could be caused by disruptions
509 under strong headwind (Kato et al. (2006 or more generically by chondrule-chondrule
510 collisions, producing splashes of melt and crystals, with varying proportions thereof, the IOG
511 representing the olivine-dominated end of the spectrum. Some crystals may have been
512 fragmented, either during initial splashing or subsequent collisions, and the latest ones not
513 rounded up by further olivine crystallization may account for the misshapen IOG (devoid of
514 euhedral/subhedral shape and concentric CL). Collisions among hot chondrules must have been
515 relatively frequent as a few percent of chondrules are compound (Gooding and Keil (; Wasson
516 et al. 1995; Akaki and Nakamura 2005; Ciesla et al. (2004. A collisional origin during chondrule
517 formation may explain the higher proportion of IOG among type II chondrules as the higher
518 concentrations of solids inferred for their formation regions (e.g., Schrader et al. (; Tenner et al.
519 (2017 Tenner et al. (2015 would promote collisions, although their rate also depends on relative
520 velocities and the time in plastic state (which may have been shorter; Jacquet et al. (2015.

² In which case speaking of the olivine phenocrysts as “precursors” might be misleading as they would have formed during the same episode as the IOG.

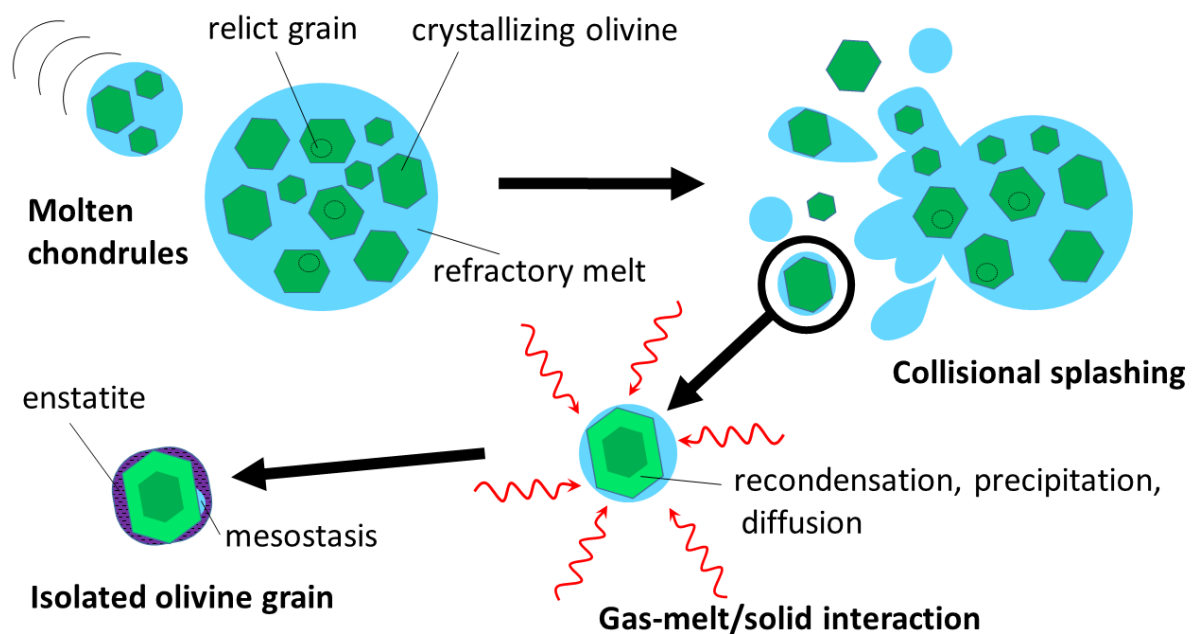
521 Incidentally, the over-representation of type II chondrules among IOG might explain the
522 dominance of ferroan olivine in Wild 2 terminal particles (13 vs. 8 (non-AOA-like) forsterites
523 in the compilation of Defouilloy, Nakashima, Joswiak, Brownlee, at variance with type I
524 chondrule dominance in carbonaceous chondrites. Indeed, if chondrule-forming regions did not
525 extend to the accretion reservoir of this comet, the latter may have received some of their
526 products by aerodynamic transport, but these would be biased toward the smaller fragments,
527 less easily decoupled from the gas (Jacquet (2014).

528 Although the chemical and isotopic properties of IOG overlap with *bona fide*
529 chondrules, their large size compared to the phenocrysts of the latter might question whether
530 they derive from the same chondrules rather than some largely lost population. However IOG
531 represent 5-13 % of type I chondrules *sensu lato* (Table 1), and if correctly interpreted as
532 chondrule splashes, are only the tip of the iceberg: one would need to add isolated pyroxene
533 grains, some cryptocrystalline chondrules (melt-dominated splashes) and larger fragments with
534 more representative silicate/melt ratios, which, if subsequently relaxed to some extent to
535 spherical shapes, would be indistinguishable from unsplashed chondrules. So the IOG source
536 chondrules have to represent a few tenths at least of the whole type I chondrule inventory in
537 carbonaceous chondrites. If we are to allow a significant population of non-IOG-related
538 chondrules, we need to assume near-complete destruction of the IOG source chondrules, and
539 very little splashing of the others, that is, very different collisional histories despite the thermal
540 history similarity suggested by petrography. It seems simpler to assume that they formed in the
541 same regions and that collisions induced about a tenth of the chondrule matter to be splashed
542 out in the form of IOG.

543 If so, how can we then explain the large size of IOG? The PO chondrules studied by
544 Marrocchi et al. (2018, 2019) and Libourel and Portail (2018) may offer a clue. Indeed, they
545 are surrounded by thick (~100 μm) palisades of olivine that asymmetrically grew toward the
546 exterior, presumably as a result of Mg and Si addition from the gas (Fig. 1C,D; Marrocchi et
547 al. 2018, 2019). Now, an isolated olivine expelled from its parent chondrule would have
548 interacted with the gas on all sides (as inferred in section 4.2), with no competing crystal around,
549 so may well have hereby attained a diameter double of that of the palisadic olivine grains, about
550 as observed (Fig. 2). Roughly speaking, the refractory (bright CL) core of the IOG might
551 correspond to the olivine crystallized in the parental chondrule and the margin (if concentric)
552 to that formed after splashing, but diffusion (for Ca) after splashing as well as recondensation
553 onto the chondrule before would make the actual boundary between these two stages uncertain.

554 Later, as mentioned in the previous subsection, the olivine would have reacted with SiO and
 555 have been replaced by enstatite to varying extent. Some objects may hence have lost their
 556 olivine-dominated nature: this may explain the largest isolated pyroxene grains seen by Jones
 557 (1999) in ALH 77307 even though some isolated pyroxene grains may be merely surficial
 558 sections of pyroxene-armoured IOG. Yet others could of course have been expelled as pyroxene
 559 grains, after pyroxene crystallization had commenced in chondrules. At any rate, Jones (1999)
 560 deemed derivation of isolated pyroxene grains from chondrules likely from textural and mineral
 561 chemical similarities with them but a dedicated study beyond her work only published in
 562 abstract form would be desirable.

563 The emerging scenario of IOG formation is sketched in Fig. 12.



564

565 **Figure 12:** Sketch of the proposed formation scenario for IOG. Chondrule collisions produce
 566 splashes, some dominated with one olivine phenocryst, which undergo interaction with the gas
 567 upon cooling.

568

569 4.4 Refractory forsterites and diversity among type I chondrules in carbonaceous chondrites

570

571 We have yet to understand refractory forsterites that is, those CaO-rich IOG studied by
 572 e.g. Steele (1988); Weinbruch et al. (2000), Pack et al. (2005). CaO enrichment is also seen in
 573 the above mentioned palisadic olivine grains in porphyritic olivine chondrules, which certainly

574 would not have been the least prone to expulsion upon collisions. Marrocchi et al. (2018, 2019)
575 explained the Ca, Al, Ti-enrichment in those by precipitation out of a Ca-Al-Ti-rich melt.
576 Indeed, in a reservoir with dust/gas ratio below unity, the melt upon olivine crystallization
577 should be fairly refractory because of evaporation (Ebel and Grossman (; Ruzicka et al. (2008.
578 The palisadic olivine grains would have escaped diffusional resetting of their Ca after
579 recondensation because of their size, the same would hold *a fortiori* for the large isolated
580 refractory forsterites. This incidentally constrains the diffusion length of Ca to a few tens of
581 microns, hence the timescale of days inferred by Marrocchi et al. (2018, 2019).

582 It remains to be understood why high refractory element contents are restricted to the
583 ^{16}O -rich ($\Delta^{17}\text{O} < -4 \text{ ‰}$) population of IOG or chondrules at large, as observed by Jacquet and
584 Marrocchi (2017). These authors inferred protracted cooling to allow equilibration of the
585 olivine with a late, incompatible-element-enriched melt, but their closed-system olivine
586 crystallization reasoning failed to recognize, as above, that the *initial* melt was incompatible-
587 element-rich. Ushikubo et al. (2012), Schrader et al. (2013) and Tenner et al. (2015, 2017) noted
588 that ^{16}O -poor chondrules tended to have more ferroan olivine than the ^{16}O -rich counterparts,
589 which they ascribed to greater solid/gas ratios in the chondrule-forming regions (with the solids
590 (dust and ice) being ^{6}O -depleted). ^{16}O -poor type I IOG do not, however, present such a FeO
591 enrichment, but their large size may have prevented diffusion of Fe^{2+} into their core during
592 cooling. A higher solid/gas ratio would lead to lower refractory element concentrations in
593 olivine at the onset of its crystallization and an upturn in the CaO vs. FeO trend (e.g. Ebel and
594 Grossman 2000), as observed (Fig. 8A).

595 In type I porphyritic chondrules, moderately high fayalite contents (above $\sim 2 \text{ mol}\%$)
596 seem correlated with pyroxene proportion: the three PP chondrules in CR3 chondrites studied
597 by Tenner et al. (2015) have $\text{Mg}\# \equiv 100 \times \text{Mg}/(\text{Mg}+\text{Fe})$ of 94.2-97.8, with no PO in this range
598 although it comprises a majority of their type I chondrules; the four PP chondrules of Lewis
599 Cliff 85332 (C3-ung, CR-related) analysed by Wasson et al. (2000) have $\text{Fs}_{4.2-9}$. Outside
600 carbonaceous chondrites, type IAB and IB chondrules in Semarkona (LL3.0) span $\text{Fa}_{1.8-6.7}$ in
601 the study of Jones (1994). On the other hand, no $\text{Mg}\#$ below 98.4 (nor $\Delta^{17}\text{O} > -3.55 \text{ ‰}$) is
602 reported in any type I chondrule from the CV3 Kaba and NWA 8613 analysed by Hertwig et al.
603 (2018 Hertwig , although they include POP and PP chondrules; same for the Murchison (CM2)
604 chondrules studied by Chaumard et al. (2018), save for one granular olivine pyroxene (GOP)
605 chondrule with $\text{Mg}\# = 96$ ($\Delta^{17}\text{O} = -2.7 \pm 0.3 \text{ ‰}$). The situation of the Yamato 81020 (CO3.0)
606 study by Tenner et al. (2013) is more marginal, with the 3 PP having $\text{Mg}\#$ of 97.8-99 (all ^{16}O -

607 rich), while all 3 type I chondrules with Mg# < 97 are POP and ^{16}O -poor. Of course, the estimate
608 of the true proportions of pyroxene is hampered by sectioning artifacts (Hezel and Kießwetter
609 (2010). Still, if there is indeed some link between “ferroan forsterite” and pyroxene abundance,
610 this may indicate that the chondrules in question cooled more slowly, since pyroxene should
611 appear at lower temperature than olivine (e.g. Ebel and Grossman 2000). Indeed, in Vigarano,
612 the most pyroxene-rich chondrules seem to have the lowest proportion of clinoenstatite in low-
613 Ca pyroxene, indicative of slower cooling at least around 1000 °C (Brearley et al. (; Soulié
614 (2014).

615 Thus, the ^{16}O -rich type I chondrules and the associated refractory forsterites may have
616 cooled more rapidly, under lower solid/gas ratios, than their generally more ferroan ^{16}O -poor
617 counterparts. Are these two subtypes of type I chondrules derived from different sources? Or
618 were two styles of type I chondrule formation events overlapping in individual reservoirs? The
619 ^{16}O -poor variety is more prevalent in CR chondrites than in other carbonaceous chondrites:
620 Keeping $\Delta^{17}\text{O} = -4$ ‰ as our arbitrary boundary between the two, Tenner et al. (2015) found a
621 10:35 ratio between ^{16}O -rich and ^{16}O -poor type I chondrules in CR3 chondrites; the same
622 research group obtained a ratio of 19:4 in the CO3.0 chondrite Y 81020 (Tenner et al. 2013),
623 23:2 in the CM2 chondrite Murchison (Chaumard et al. 2018) and 71:7 in CV chondrites
624 (Rudraswami et al. (; Hertwig et al. (; Hertwig et al. (2019). One could envision that ^{16}O -poor
625 type I chondrule formation was merely more frequent in the CR chondrite reservoir without
626 denying ^{16}O -poor type I chondrule formation elsewhere. However, ^{16}O -poor chondrules appear
627 systematically more ^{54}Cr -rich than their ^{16}O -rich counterparts in both CR and CV chondrites
628 (e.g., Williams et al. 2020; Schneider et al. (under review)). This indicates a different reservoir
629 of origin for the ^{16}O -poorer type I chondrule, which presumably was spatially and/or temporally
630 closer to the accretion event of CR chondrites. We note that CR chondrules exhibit initial ^{26}Al
631 abundances lower than their counterparts in CO and CV chondrules (Nagashima et al. 2014;
632 Schrader et al. 2017; Tenner et al. 2019), but possibly comparable to cometary samples (Ogliore
633 et al. 2012; Nakashima et al. 2015). The Fe/Mn ratio of olivine in type II chondrules, lower in
634 CR chondrites than in other carbonaceous chondrites (e.g., Berlin et al. 2011; Jones (; Frank et
635 al. 2014) also sets the CR chondrule population apart among carbonaceous chondrites.

636 So the emerging picture is that of a CR-like chondrule population, with apparently higher
637 solid/gas ratios and slower cooling than the others (for the type I chondrules), which contributed
638 to non-CR reservoirs dominated by ^{16}O -rich chondrules. Such a limited mixing of chondrules
639 beyond their formation region could be reproduced in the simulations of Goldberg et al. (2015).

640 This limited mixing would also have involved refractory inclusions, as suggested by Al-Mg
641 isotopic evidence (Larsen et al. (2020). While the ^{16}O -poor type I chondrules of different
642 carbonaceous chondrite groups (CO, CV, CM) may thus come from a single source, the ^{16}O -
643 rich ones, although O isotopically similar, may still have diverse origins, as suggested by
644 distinctive petrographic features in the different chemical groups (e.g. size, prevalence of fine-
645 or coarse-grained rims, occurrence of primary feldspar... see Jones 2012).

646

647

648 **5. Conclusion**

649

650 We have carried out microscopic, cathodoluminescence, chemical and O isotopic
651 measurements of type I isolated olivine grains (IOG) in the carbonaceous chondrites Allende
652 (CV3), Northwest Africa 5958 (C2-ung), Northwest Africa 11086 (CM-an), Allan Hills 77307
653 (CO3.0).

654 The IOG typically represent a few percent of the studied carbonaceous chondrites, and are
655 dominated by type I IOG but with an overrepresentation of type II compositions. The type I
656 IOG present O isotopic signatures similar to *bona fide* chondrules, although they are generally
657 coarser than chondrule phenocrysts. CaO, which may reach up to 0.9 wt% in ^{16}O -rich IOG, as
658 well as Al_2O_3 and TiO_2 decrease from core to rim, while FeO, MnO, Cr_2O_3 increase. Electron
659 microprobe traverses and CL imaging often reveal concentric zoning, with enstatite frequently
660 rimming the olivine's outer edge.

661 The general isotopic and chemical similarities, indeed the textural continuum, with *bona fide*
662 chondrules indicate that the IOG were derived from them. Their not uncommonly unbroken
663 morphology and evidence of interaction with the gas, with recondensation on all sides possibly
664 accounting for their large size, indicates that rather than being cold fragments of chondrules,
665 they were expelled from chondrules while these were molten, likely during chondrule-
666 chondrule collisions. Among IOG, the refractory forsterites retained their high-temperature
667 composition presumably established by equilibration with a refractory melt, as some palisadic
668 olivine grains in porphyritic olivine chondrules. These ^{16}O -rich type I chondrules may have
669 undergone quicker quenching than their ^{16}O -poorer (type I) counterparts, possibly derived from
670 a CR chondrite-like reservoir.

671 Thus, IOG were likely derived from chondrules, as favored by most recent authors (e.g. Jones
672 1992; Jones et al. 2000; Pack et al. 2005; Russell et al. 2010), but were affected by significant
673 gas-solid interactions (before and after expulsion from the parent chondrules), as inferred by
674 their earliest students (e.g. Fuchs et al. 1973; Olsen and Grossman 1974, 1978; Steele 1986,
675 1988, 1989), hereby reconciling the two endmembers of the isolated olivine literature.

676

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691

692

693

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