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## 27 Abstract

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 $\Delta^{17}O < -4$ ‰ 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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In their seminal description of Murchison, Fuchs et al. (1973) reported isolated olivine 56 grains (henceforth IOG), one of which earned the honor of their frontispiece. With the 57 condensation models in full swing since the fall of Allende (e.g., Grossman 1972; Marvin et al. 58 1970), it was but natural to interpret these grains, soon found in other carbonaceous chondrite 59 clans, as nebular condensation products, for which Olsen and Grossman (1974, 1978) adduced 60 61 morphological evidence such as large size, euhedral shapes and patterned crystal surfaces. Yet chemical analogies of the IOG with chondrule olivine, as well as the presence of glass 62 inclusions chemically comparable to chondrule mesostases, suggested early that they were 63 chondrule fragments, possibly liberated after alteration of the mesostasis (e.g. McSween (; 64 Richardson and McSween 1978; Desnoyers (; Nagahara and Kushiro (; Jones (1992. 65

Still, Steele (1986) drew attention to a subset of IOG, both in carbonaceous and ordinary 66 67 chondrites, with particularly forsteritic and refractory mineral chemistries and bright blue cathodoluminescence, which led him to entertain again a condensate origin (e.g. Steele (, Steele 68 69 , for which the SIMS (Secondary Ion Mass Spectrometer) measurements of Weinbruch et al. (1993) found supporting evidence in their <sup>16</sup>O enrichment (that is, in the direction of refractory 70 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 IOG largely overlap with chondrules in carbonaceous chondrites (e.g., Jones et al. (; Russell 73 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 accordance with near-equilibrium igneous partitioning (e.g. Jacquet et al. (; Jacquet and 76 77 Marrocchi (; Pack et al. (2005. While the balance of evidence thus remains in favor of a genetic link between IOG and chondrules, some systematic differences, at least among the forsteritic 78 79 ones, with the general population of chondrules remain to be understood so as to decide whether they sample the same heating events. For instance, the relative <sup>16</sup>O enrichment of Tagish Lake 80 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 incompatible element enrichment of IOG in Northwest Africa (NWA) 5958 (C2-ung) to longer 83 84 thermal processing. The IOG would be obviously important (olivine-rich) pieces of the puzzle of chondrule origin, if only their exact formation context *vis-à-vis* mainstream chondrules could
be ascertained.

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

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### 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 31811m4 (or 3181-4) of Allende (all three from the Muséum national d'Histoire naturelle, Paris, France), the thin section ALH 77307,96 of Allan Hills 109 (ALH) 77307 (from the NASA Antarctic Search for Meteorites program) and a thick section of 110 NWA 11086 prepared at CRPG. Allende is an oxidized Allende-like CV sub-group classified 111 as a CV3.6 by Bonal et al. (2006). ALH 77307 is a CO3.0 chondrite (Grossman and Brearley 112 2005; Bonal et al. 2007; Busemann et al. 2007). Opaque assemblages, chondrule mesostases, 113 olivine textures and compositions of chondrules show no indication of alteration and diffusional 114 exchange. NWA 5958 corresponds to a C2-ung CM-like chondrite with type II chondrule 115 olivine Cr content and opaque petrography indicating minimal thermal metamorphism (< 116

300°C) but chondrule mesostases bearing witness to significant aqueous alteration (Jacquet et 117 al., 2016; Jacquet and Marrocchi 2017). NWA 11086 has been classified as a CM-an due to <sup>16</sup>O 118 enrichment and the absence of phyllosilicates according to X-ray diffraction (Gattacceca et al. 119 120 2019), but significant aqueous alteration is indicated by presence of PCP (Poorly Characterized Phases, also known as tochilinite cronstedtite intergrowths; see e.g. Lentfort et al., this issue), 121 conversion of chondrule mesostasis to « spinach » (Fuchs et al. 1973) and paucity of Fe-Ni 122 metal, so it might be a plain, if somewhat <sup>16</sup>O-rich, CM2 chondrite. In the plots and the 123 discussion, data from these latter two chondrites and ALH 77307 will be subsumed in the 124 125 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 Géochimiques (CRPG-CNRS, Nancy, France) using 3 nA electron beam accelerated at 15 kV. 128 129 Modal abundances of high-temperature components were also estimated by manual point BSE 130 counting on JMicrovision software (N. Roduit; maps using the 131 https//jmicrovision.github.io; last accessed in December 2019) with 4000 points (randomly chosen by the software) per section (except for ALHA 77307, 2000 points). An object was 132 considered an IOG if its interior was dominated by one olivine crystal. During closer BSE 133 observation, only IOG large enough for SIMS analyses (~15 µm spot; this is also roughly the 134 scale over which the BSE image allowed assignment to IOG in the point counting) were 135 selected for further study. The effective radius of the IOG or the coarsest olivine of each studied 136 chondrule was calculated as the radius of the equal area disk, from back-scattered electron 137 (BSE) images. Cathodoluminescence (CL) imaging of chondrules was performed using (i) an 138 RGB CL-detection unit attached to a field emission gun secondary electron microscope JEOL 139 J7600F at the Service Commun de Microscopie Électronique (SCMEM, Nancy, France) and 140 (ii) a field-gun ZEISS Supra 55 VP equipped with an OPEA catholuminescence device 141 (imaging and spectral) with a high-tech parabolic mirror. Quantitative chemical compositions 142 of olivine grains were obtained using a Cameca SX Five electron microprobe (EMP) at the 143 144 Université Pierre et Marie Curie (UPMC, Camparis, Paris, France) using a 150 nA focused beam (about ~2 µm in diameter) accelerated to 15 kV. We analysed Na, Mg, Si, Al, K, Ca, Fe, 145 146 Ti, Cr, and Mn in olivine grains. The high beam current allowed detection limits for silicates of 100 ppm for Al, Ca, and Ti, 150 ppm for Mn and Si, and 200 ppm for Na, K, Cr, Fe, and Mg. 147 148 The PAP software was used for matrix corrections.

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

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

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- **3. Results**
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174	Petrography

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Type I chondrules present the usual mineralogical zoning (Fig. 1) of olivine phenocrysts dominating in the interior and enstatite laths, poikilitically enclosing olivine chadacrysts, near the outside (e.g., Friend et al. (; Libourel et al. (2006. The mesostasis is altered in the CM chondrites. When viewed in CL, interior olivine grains often display dark cores (Fig. 1) that correspond to relict grains for those chondrules investigated by Marrocchi et al. (2018, 2019), 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).



Figure 1: Back-scattered electron and cathodoluminescence images of representative 186 chondrules (left and right, respectively, unless otherwise noted). Orange dashed lines show 187 location of profiles shown in Fig. 6. A) and B) Chondrule CHA in section NWA 5958-4. The 188 189 ovoid chondrule contains euhedral olivine grains with little discernible pyroxene and relatively abundant altered mesostasis. Its lower half is occupied by a large olivine apparently embayed 190 near the central hole, and has concentric and oscillatory zoning in blue CL (as the other olivine 191 phenocrysts). Many of the oval dark-CL spots trace metal inclusions. C) and D) PO chondrule 192 CH9 in section NWA 5958-1. Dark-CL spots in interior olivine grains likely correspond to 193 194 relicts. Brightest blue CL is seen in the coarser palisadic olivine grains but CL intensity declines with some oscillations near the outer edge. A small (~50 µm diameter) PO chondrule is attached 195 196 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, bluish CL (note that left panel is somewhat more zoomed than right panel). G) Chondrule N1-199 200 26 in NWA 5958 (BSE only). This irregularly shaped chondrule is dominated by pyroxene crystals, with <20 µm olivine grains, in addition to altered mesostasis and metal grains. 201 202 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 have been replaced by pyroxene to the same extent as the remainder of the chondrule margins). 205 H) Chondrule CH1 in Allende (CL only) shows coarse, bright CL triangular olivine crystals 206 207 and mesostasis near the center. It might be a surficial section through the "palisade" of a PO chondrule. 208

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

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

Type II chondrules

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

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

While most IOG are of type I, it is noteworthy that the type II/type I modal ratio is significantly higher for IOG (0.1-0.6) than for *bona fide* chondrules (0.03-0.11), by a factor of 4 to 7. This is qualitatively in line with the histograms of McSween (1977) which indicate values of 2 and 0.8 for this ratio in CM and CO chondrites, respectively, with that of Desnoyers (1980) indicating a ratio of 1.2 for Niger (C2), although those histograms are based on microprobe analyses and not areas (perhaps accounting for the systematic difference). Seven out of the 12 Tagish Lake IOG of Russell et al. (2010) were of type II; same for 25 out of 101 CI chondrite IOG compiled by Piralla et al. (2020). This point being made, we henceforthexclusively focus on type I IOG.

As noted by Olsen and Grossman (1978), the IOG tend to be big compared to chondrule 239 phenocrysts (Fig. 2, even though the first bin is cut off by our selection biases). In fact, in CV 240 241 chondrites, their size distribution is comparable to that of the *coarsest* chondrule phenocrysts; in CM/CO chondrites, more than 90 % of the IOG studied here are even bigger than the mode 242 of the coarsest chondrule phenocrysts (near 30  $\mu$ m effective radius). About half (21 out of 41<sup>1</sup>) 243 of the IOG examined here by BSE/CL and EMP have equidimensional, often 244 245 euhedral/subhedral shapes, although the others are evidently fragments of larger objects (e.g. Figs. 3C,D; 4G,H). The CL of the former, generally blue in the core, is concentrically zoned, 246 247 with intensity decreasing toward the margin which may have a redder tint. Discrete darker-CL streaks, partially or entirely surrounding the center, are often superimposed on the background 248 249 trend, generally paralleling the grain edges. An oscillatory sector zoning is spectacularly visible in NWA 11086 IO12 (Fig. 5); some may be discernible in NWA 5958-1 IO7 as well (Fig. 4F). 250 Despite their name, the IOG are not pure olivine. The olivine may enclose metal grains and 251 glass inclusions, and, when whole, is nearly always surrounded, partly or entirely, by enstatite 252 (from a few microns to tens of microns in thickness), sometimes with mesostasis (although we 253 did not find as large a mesostasis patch as in IOG N6-5 in Fig. 1e of Jacquet and Marrocchi 254 2017) and even further olivine phenocrysts. Enstatite rims, partial or total, are sometimes seen 255 even around fragments (e.g. Fig. 3C,D). We note that Jones (1992) called large olivine grains 256 surrounded by pyroxene in ALHA 77307 "macroporphyritic chondrules", in contradistinction 257 258 to "bare" isolated olivine grain, but given the continuum between them, we maintain the name IOG for all those objects. The continuum, in fact, extends to bona fide chondrules (e.g. Jacquet 259 260 and Marrocchi 2017), with some chondrules exhibiting disproportionately large olivine crystals (e.g. Fig. 1A,B; chondrule N5-21 with three aligned coarse olivine grains in Fig. 2b of Jacquet 261 262 et al. 2016), not to mention chondrule-IOG or IOG-IOG compounds (see e.g. Fig. 1G, 3E,F). Overall, this continuum makes the assignment of objects to IOG rather than chondrules 263 264 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.

<sup>&</sup>lt;sup>1</sup> 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 mm<sup>2</sup> vs. 0.59 mm<sup>2</sup>).



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



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



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



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

Chemistry and oxygen isotopes

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

In the biplots of this paper (viz. Fig. 7, 8), the IOG will be represented by their apparent core 333 composition. CaO spans 0.16-0.89 wt% in IOG and 0.05-0.66 wt% in chondrule coarsest 334 olivine (with averages of 0.44 vs. 0.30 wt%); Al<sub>2</sub>O<sub>3</sub> spans 0.06-0.42 wt% in IOG and chondrule 335 coarsest olivine 0.02-0.53 wt% (with averages of 0.20 vs. 0.12 wt%). Crystal size seems to 336 correlate negatively with Fe, Mn but the positive correlation with Ca and Al noted by Jacquet 337 and Marrocchi (2017) is very rough (Fig. 7). This may be to some extent a 2D sectioning artifact 338 as more equatorial sectioning of the grains (providing the widest areas) would pass closest to 339 340 the actual refractory core. Excluding Allende, whose olivine FeO contents up to 5 wt% are obviously secondary, the IOG have low FeO contents (0.35-1.4 wt%) anticorrelated with CaO, 341 342 similar to chondrule phenocrysts in this compositional range (Fig. 8).



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





Figure 7: Olivine size vs minor element content (A: FeO; B: MnO; C: CaO; D: Al<sub>2</sub>O<sub>3</sub>) in IOG
and chondrule coarsest olivine. Data for all analyzed chondrule phenocrysts in Marrocchi et al.
(2018, 2019) are also shown.



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Figure 8: Minor element biplots of IOG, chondrule coarsest olivine of this study and chondrule
olivine (keyed by chondrule) from Marrocchi et al. (2018, 2019). A) CaO vs. FeO. The
correlation is negative for FeO below ~1.5 wt% and positive afterward. Individual chondrules
plot in only one of these trends. IOG all belong to the former. B) Al<sub>2</sub>O<sub>3</sub> vs. TiO<sub>2</sub>. C) MnO vs.
FeO. D) MnO vs Cr<sub>2</sub>O<sub>3</sub>.

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

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



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



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



**Figure 11**: Oxygen isotopes vs. minor elements for IOG and coarsest olivine grains, including CV data by Libourel and Chaussidon (2011), along with host olivine data from the Marrocchi et al. (2019) compilation. A)  $\Delta^{17}$ O vs. FeO. B)  $\Delta^{17}$ O vs. MnO. C)  $\Delta^{17}$ O vs. CaO. D)  $\Delta^{17}$ O vs. Al<sub>2</sub>O<sub>3</sub>.

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### 4. Discussion

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397 4.1 Genetic link between isolated olivine grains and chondrules

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399 The general overlap in  $\Delta^{17}$ 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 400 401 1992; Jones et al. 2000), that IOG are closely linked to chondrules rather than early nebular 402 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 403 404 1978). Other phases associated with olivine (metal, occasional companion olivine crystals, pyroxene) are also most analogous to chondrule petrography; with IOG CL being comparable 405 to that of the coarsest olivine in chondrules (Fig. 1). We may also recall the REE fractionation 406 measured in IOG olivine (Pack et al. 2005; Jacquet et al. 2012; Jacquet and Marrocchi 2017) 407 which is at variance with the flatter patterns in AOA olivine which formed by condensation 408 409 (Ruzicka et al. (; Jacquet and Marrocchi 2017). The oscillatory sector zoning shown by the 410 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 411 412 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; 413 414 Pack et al. 2005; Russell et al. 2010; Ushikubo et al. 2012), that IOG are genetically related to 415 chondrules.

416

#### 417 4.2 Isolated olivine grains formed hot

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419 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 420 separate olivine phenocrysts from the mesostasis. While Richardson and McSween (1978) had 421 invoked the friability of the altered mesostasis at least for CM chondrite chondrules, that 422 alteration is now widely believed to have occurred on the parent body (Brearley (2014, too late 423 for chondrule-chondrule collisions, and would not be relevant for the least altered chondrites 424 such as ALH 77307. The large size of IOG compared to chondrule phenocrysts is also 425 problematic if the former are fragments of the latter (e.g., Olsen and Grossman 1978). The 426

larger IOG/chondrule ratio for type II compared to type I objects in carbonaceous chondrites
would not be understandable if the chondrules were fragmented after being mixed together in
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, this study focuses on type I IOG, we recall that the ferroan IOG N6-7 in NWA 5958 exhibited 432 oscillatory zoning visible in BSE (Jacquet et al. 2016; Jacquet and Marrocchi 2017). The 433 frequent presence of near-complete enstatite margins (e.g. Fig. 3G,H; Fig. 4C,D) in type I IOG 434 435 is comparable to those shown by whole chondrules. These are ascribed to the influx of SiO into residual melts, promoting pyroxene crystallization at the expense of olivine (e.g., Libourel et al. 436 437 (; Chaussidon et al. (; Friend et al. (; Barosch et al. 2019). This indicates that the IOG experienced such influx as independent objects. In fact, prior to enstatite precipitation, the 438 439 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. (2018, 2019) and Libourel and Portail (2018) for the palisadic olivine. More volatile Fe, Mn, 442 Cr would have recondensed contributing to their outward increase in the olivine. Perhaps in 443 some cases-for those IOG which really were bare euhedral olivine grains-the late olivine 444 445 condensed directly, without any intermediate liquid, which may account for the patterned surfaces which Olsen and Grossman (1974) compared to terrestrial sublimate olivine. In such 446 447 cases, glass inclusions would of course have to date back to the time the olivine was still hosted in the partially molten chondrule. Thus IOG experienced gas-melt (or gas-solid) interaction as 448 isolated objects. In that sense, condensation played a role, after all, in IOG formation. 449

450 Further evidence for a hot formation of IOG is afforded by compound objects, either between IOG (e.g. Fig. 3E,F) or with chondrules (e.g. Fig. 1G), which require collisions in a 451 452 plastic state (Gooding and Keil (; Akaki and Nakamura 2005). Some bona fide chondrules with coarse olivine phenocrysts may have engulfed them during cooling—that is, be "enveloping" 453 454 compound chondrules in the parlance of Wasson et al. (1995). To be sure, many IOG are mere misshapen olivine (± mesostasis, pyroxene) debris, but even those may sometimes be fragments 455 456 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 Annex). We note that Jones (1992) did envision isolated olivine grain formation from 458

459 fragmentation of "macroporphyritic chondrules", although the formation of the latter was left460 unexplained.

We conclude from enstatite rimming and concentric CL zoning in many IOG (Fig. 3-4) 461 that they formed as stand-alone high-temperature objects. In that sense, we deem it warranted 462 463 to also call them "chondrules" (sensu lato, e.g. Jacquet et al. 2012; Jacquet and Marrocchi 2017), whenever the distinction is not of interest, as they are part of the same continuum of objects 464 465 (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 or a forsterite-bearing CAI fragment, although no evidence (e.g. isotopic) to that effect has been 468 469 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.

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#### 472 4.3 Isolated olivine grains as chondrule splashes

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

Given the general analogy of chondrule relict grains with AOA olivine (Marrocchi et al. 480 481 2018, 2019), a first candidate would be simply AOAs. However, melting experiments of AOA analogs by Whattam et al. (2010) produced porphyritic textures. Also bulk AOA composition, 482 483 although deficient in silica relative to type I chondrules, are closer to the latter than IOG (because of their CAI-like inclusions). Perhaps, though, a fragment of the olivine portion of a 484 485 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, the abundance of AOAs overall in the studied chondrites is too low ( $\sim 1$  vol%) to account for 488 489 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 their491 AOAs, has been converted in chondrules (Marrocchi et al. 2019).

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

Separating olivine crystals from the mesostasis would be conceivable if the mesostasis 506 were *liquid*, that is during chondrule formation<sup>2</sup>. The crystals and liquid could separate (to some 507 508 extent) owing to buoyancy during sudden accelerations. Such could be caused by disruptions under strong headwind (Kato et al. (2006 or more generically by chondrule-chondrule 509 collisions, producing splashes of melt and crystals, with varying proportions thereof, the IOG 510 511 representing the olivine-dominated end of the spectrum. Some crystals may have been fragmented, either during initial splashing or subsequent collisions, and the latest ones not 512 513 rounded up by further olivine crystallization may account for the misshapen IOG (devoid of euhedral/subhedral shape and concentric CL). Collisions among hot chondrules must have been 514 515 relatively frequent as a few percent of chondrules are compound (Gooding and Keil (; Wasson et al. 1995; Akaki and Nakamura 2005; Ciesla et al. (2004. A collisional origin during chondrule 516 517 formation may explain the higher proportion of IOG among type II chondrules as the higher concentrations of solids inferred for their formation regions (e.g., Schrader et al. (; Tenner et al. 518 519 (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.

<sup>&</sup>lt;sup>2</sup> 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.

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

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

If so, how can we then explain the large size of IOG? The PO chondrules studied by 543 544 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, so may well have hereby attained a diameter double of that of the palisadic olivine grains, about 549 550 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 onto the chondrule before would make the actual boundary between these two stages uncertain. 553

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

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



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

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

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

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

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

595 In type I porphyritic chondrules, moderately high fayalite contents (above ~2 mol%) seem correlated with pyroxene proportion: the three PP chondrules in CR3 chondrites studied 596 by Tenner et al. (2015) have  $Mg\# \equiv 100 \times Mg/(Mg+Fe)$  of 94.2-97.8, with no PO in this range 597 although it comprises a majority of their type I chondrules; the four PP chondrules of Lewis 598 Cliff 85332 (C3-ung, CR-related) analysed by Wasson et al. (2000) have Fs<sub>4.2-9</sub>. Outside 599 600 carbonaceous chondrites, type IAB and IB chondrules in Semarkona (LL3.0) span Fa1.8-6.7 in the study of Jones (1994). On the other hand, no Mg# below 98.4 (nor  $\Delta^{17}O > -3.55$  ‰) is 601 reported in any type I chondrule from the CV3 Kaba and NWA 8613 analysed by Hertwig et al. 602 (2018 Hertwig, although they include POP and PP chondrules; same for the Murchison (CM2) 603 chondrules studied by Chaumard et al. (2018), save for one granular olivine pyroxene (GOP) 604 chondrule with Mg# = 96 ( $\Delta^{17}$ O = -2.7 ± 0.3 ‰). The situation of the Yamato 81020 (CO3.0) 605 study by Tenner et al. (2013) is more marginal, with the 3 PP having Mg# of 97.8-99 (all <sup>16</sup>O-606

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

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

So the emerging picture is that of a CR-like chondrule population, with apparently higher solid/gas ratios and slower cooling than the others (for the type I chondrules), which contributed to non-CR reservoirs dominated by <sup>16</sup>O-rich chondrules. Such a limited mixing of chondrules beyond their formation region could be reproduced in the simulations of Goldberg et al. (2015). This limited mixing would also have involved refractory inclusions, as suggested by Al-Mg isotopic evidence (Larsen et al. (2020. While the <sup>16</sup>O-poor type I chondrules of different carbonaceous chondrite groups (CO, CV, CM) may thus come from a single source, the <sup>16</sup>Orich ones, although O isotopically similar, may still have diverse origins, as suggested by distinctive petrographic features in the different chemical groups (e.g. size, prevalence of fineor coarse-grained rims, occurrence of primary feldspar... see Jones 2012).

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**5.** Conclusion

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

The IOG typically represent a few percent of the studied carbonaceous chondrites, and are dominated by type I IOG but with an overrepresentation of type II compositions. The type I IOG present O isotopic signatures similar to *bona fide* chondrules, although they are generally coarser than chondrule phenocrysts. CaO, which may reach up to 0.9 wt% in <sup>16</sup>O-rich IOG, as well as  $Al_2O_3$  and TiO<sub>2</sub> decrease from core to rim, while FeO, MnO,  $Cr_2O_3$  increase. Electron microprobe traverses and CL imaging often reveal concentric zoning, with enstatite frequently rimming the olivine's outer edge.

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

Thus, IOG were likely derived from chondrules, as favored by most recent authors (e.g. Jones 1992; Jones et al. 2000; Pack et al. 2005; Russell et al. 2010), but were affected by significant gas-solid interactions (before and after expulsion from the parent chondrules), as inferred by 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.

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