

1 **Oxygen isotope systematics of crystalline silicates in a giant cluster IDP:**  
2 **A genetic link to Wild 2 particles and primitive chondrite chondrules**

3 Mingming Zhang<sup>a\*</sup>, Céline Defouilloy<sup>a†</sup>, David J. Joswiak<sup>b</sup>, Donald E. Brownlee<sup>b</sup>, Daisuke  
4 Nakashima<sup>a§</sup>, Guillaume Siron<sup>a</sup>, Kouki Kitajima<sup>a</sup>, Noriko T. Kita<sup>a</sup>

5 <sup>a</sup> *WiscSIMS, Department of Geoscience, University of Wisconsin–Madison, Madison, WI 53706, USA*

6 <sup>b</sup> *Department of Astronomy, University of Washington, Seattle, WA 98195, USA*

7 <sup>\*</sup>*Corresponding address: [mzhang467@wisc.edu](mailto:mzhang467@wisc.edu)*

8 <sup>†</sup>*Present address: Ingénieur Applications at CAMECA, 29 quai des Grésillons, 92633 Gennevilliers  
9 Cedex, France*

10 <sup>§</sup>*Present address: Department of Earth and Planetary Material Sciences, Tohoku University, Miyagi 980-  
11 8578, Japan*

12 **Highlights**

13 • Five out of 20 fragments ( $\geq 5 \mu\text{m}$ ) in the giant cluster IDP U2-20GCA have chondrule-like textures.  
14 • The oxygen isotope range and Mg#– $\Delta^{17}\text{O}$  relationship of crystalline silicate fragments are closest to Wild  
15 2 particles and CR chondrite chondrules.  
16 • Four fragments likely sourced from the formation regions of ordinary, R, or CH-CB chondrite chondrules.

17 ABSTRACT

18 Anhydrous interplanetary dust particles (IDPs) collected from Earth's stratosphere are the most  
19 primitive extraterrestrial materials that likely are the remnants of major building blocks of our solar system.  
20 While they probably originated from icy outer solar system comets, this hypothesis needs to be further verified  
21 via comparison with Wild 2 (a Jupiter-family comet) particles returned by the *Stardust* mission. Besides, the  
22 origins of their components can be further constrained by comparing with materials in primitive chondrite.  
23 Here we investigate the petrology and oxygen isotope systematics of 20 fragments extracted from a giant  
24 cluster IDP U2-20GCA. Fifteen are monomineralic or polymimetic fragments composed of olivine and/or  
25 pyroxene. Others are plagioclase/glass bearing fragments similar to barred-olivine chondrules, enstatite-rich  
26 chondrules, and Al-rich chondrules (ARC). They show a range of Mg# [mol% Mg/(Mg+Fe)] in olivine and  
27 pyroxene (99-75) and MnO and Cr<sub>2</sub>O<sub>3</sub> abundances in olivine that resemble Wild 2 particles. Individual  
28 fragments have relatively <sup>16</sup>O-poor oxygen isotope ratios with  $\delta^{18}\text{O}$  and  $\delta^{17}\text{O}$  varying from  $-6.2 \pm 1.0\text{\textperthousand}$  to  $6.8$   
29  $\pm 1.9\text{\textperthousand}$  and  $-6.7 \pm 2.6\text{\textperthousand}$  to  $5.3 \pm 1.2\text{\textperthousand}$ , respectively. Most fragments show  $\Delta^{17}\text{O}$  ( $= \delta^{17}\text{O}-0.52 \times \delta^{18}\text{O}$ )  
30 increasing from  $\sim -3\text{\textperthousand}$  to  $\sim 0\text{\textperthousand}$  with decreasing Mg#, similar to those observed in Wild 2 particles and CR  
31 chondrite chondrules. Four fragments (including the ARC-like fragment) show oxygen isotope signatures of  
32 ordinary (O), R, or CH-CB chondrite chondrules. The similarities among the giant cluster IDP, Wild 2  
33 particles, and primitive chondrite chondrules demonstrate (i) a cometary origin of anhydrous IDPs; (ii) the  
34 major source of crystalline silicates in comets is likely to be CR chondrite chondrule-like materials, while  
35 minor sources include O, R, or CH-CB chondrite chondrule-like materials. The conclusions support the  
36 hypothesis that anhydrous IDPs sampled extremely heterogeneous materials formed in wide solar system  
37 regions like Wild 2 particles.

38 **Keywords:** giant cluster IDP; chondrule-like fragments; oxygen isotope ratios; Wild 2 particles; CR chondrite  
39 chondrules

41 **1. Introduction**

42 Interplanetary dust particles (IDPs, typically  $< 50 \mu\text{m}$ ) collected from the Earth's stratosphere are  
43 among the most primitive extraterrestrial materials available for laboratory investigation (e.g., Bradley, 2014).  
44 They typically have chondritic elemental compositions and can be divided into (i) chondritic smooth, hydrated  
45 IDPs, compact objects dominated by hydrous minerals like cronstedtite, tochilinite, serpentine, saponite, etc.;  
46 (ii) chondritic porous, anhydrous IDPs, highly fluffy aggregates (up to 70% porosity) composed predominantly  
47 of submicron- to micron-sized olivine, pyroxene, iron-rich sulfide, Fe-Ni metal, glass with embedded metal  
48 and sulfide (GEMS), silicate glass, and carbonaceous material (Bradley, 2014). Refractory materials such as  
49 calcium-aluminum-rich inclusions (CAIs) and amoeboid olivine aggregates (AOAs) are rare (e.g., Joswiak et  
50 al., 2017). Some anhydrous IDPs contain abundant presolar grains, solar flare tracks typically in olivine or  
51 pyroxene, and unheated or only slightly heated carbonaceous materials, suggesting that they suffered minimal  
52 mineralogical modifications during atmospheric entry (peak temperature typically  $< 100^\circ\text{C}$ ) (Matrajt et al.,  
53 2012; Bradley, 2014). The atmospheric entry velocity is inferred to be slow, consistent with an origin from  
54 asteroids or comets with low inclinations and perihelia larger than 1.2 astronomical units (Sandford and  
55 Bradley, 1989).

56 Wild 2 (a Jupiter-family comet) particles returned by the NASA *Stardust* mission in 2006 provide  
57 important insights into the mineralogy, chemistry, and isotope geochemistry of outer solar system cometary  
58 materials. The most striking findings are high-temperature objects similar to inner-disk CAIs and chondrules  
59 (Zolensky et al., 2006; Nakamura et al., 2008; Bridges et al., 2012; Ogliore et al., 2012; Gainsforth et al., 2015;  
60 Joswiak et al., 2017), strongly demonstrating large-scale transportation of solids across the full dimension of  
61 the protoplanetary disk (Brownlee, 2014). Like anhydrous IDPs, Mg# [mol% Mg/(Mg+Fe)] of olivine and  
62 pyroxene in Wild 2 particles are evenly distributed from 100 to 58, Fe vs. Mn abundances of olivine span a  
63 wide range that do not follow the trendlines defined by CO and ordinary (O) chondrite chondrules, suggesting  
64 a diversity of source regions for these particles (Zolensky et al., 2006; Berlin et al., 2011; Frank et al., 2014;  
65 Brownlee and Joswiak, 2017). Oxygen isotope ratios of Wild 2 particles have been determined by Secondary  
66 Ion Mass Spectrometry (SIMS) that show solar-like  $^{16}\text{O}$ -rich compositions ( $\delta^{18}\text{O} \sim -50\text{ ‰}$ ) and chondrule-like  
67  $^{16}\text{O}$ -poor compositions ( $\delta^{18}\text{O} \sim -6\text{ ‰}$  to  $+7\text{ ‰}$ ) (McKeegan et al., 2006; Nakamura et al., 2008; Bridges et al.,

68 2012; Nakashima et al., 2012a; Gainsforth et al., 2015; Ogliore et al., 2015; Defouilloy et al., 2017). The  
69 majority of these SIMS data were obtained from coarse (4-30  $\mu\text{m}$ ) particles using a CAMECA IMS-1280 and a  
70  $\sim 2 \mu\text{m}$  beam with a precision in  $\Delta^{17}\text{O}$  of  $\sim 2\text{\textperthousand}$ . In particular, Nakashima et al. (2012a) and Defouilloy et al.  
71 (2017) studied olivine and pyroxene bearing particles and revealed a systematic change in mass-independent  
72 fractionation of oxygen isotopes ( $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$ ) with their Mg# values similar to CR chondrite  
73 chondrules.

74 Oxygen isotope ratios of anhydrous IDPs have been determined mostly by NanoSIMS on nm-scale  
75 tiny minerals or their mixtures that represent diverse components in the early solar system, including extremely  
76  $^{16}\text{O}$ -rich presolar grains ( $\delta^{18}\text{O}$  down to  $-160\text{\textperthousand}$ ), solar-like  $^{16}\text{O}$ -rich or chondritic-like  $^{16}\text{O}$ -poor objects ( $\delta^{18}\text{O} =$   
77  $-30\text{\textperthousand}$  to  $+19\text{\textperthousand}$ ), and significantly  $^{16}\text{O}$ -poor objects ( $\delta^{18}\text{O} = +80\text{\textperthousand}$  to  $+200\text{\textperthousand}$ ) that likely formed by isotope  
78 self-shielding in the outer solar system (e.g., Busemann et al., 2009; Starkey et al., 2014). Compared to Wild 2  
79 particles, high-precision oxygen isotope data of anhydrous IDPs are limited due to the lack of suitable large  
80 fragments ( $> 2 \mu\text{m}$ ) for higher precision SIMS analyses. Crystalline silicates in three anhydrous IDPs show  
81 chondrule-like  $^{16}\text{O}$ -poor isotope ratios, with average  $\delta^{18}\text{O}$  ranging from  $-2.9 \pm 0.7\text{\textperthousand}$  to  $1.0 \pm 2\text{\textperthousand}$  (Aléon et al.,  
82 2009; Nakashima et al., 2012b). Thus, new high-precision oxygen isotope data of anhydrous IDPs are needed  
83 for better comparison with Wild 2 particles and primitive chondrite components in order to verify the cometary  
84 origin of anhydrous IDPs and elucidate the sources of their crystalline silicates.

85 A giant cluster IDP U2-20GCA, collected by impact onto a silicone oil-coated substrate flag “U2-20”  
86 in the stratosphere, was utilized in this study. It is an extremely porous and fragile object composed of a  $\sim 350$   
87  $\mu\text{m}$  core surrounded by a  $\sim 1 \text{ mm}$  low-density debris halo, where the core is made up of a dense monolayer of  
88 thousands of dark and transparent fragments (up to  $42 \mu\text{m}$ ) (Joswiak et al., 2017). CAIs, AOAs, and “Kool  
89 grains” (kosmochloric Ca-rich pyroxene + Fe-rich olivine  $\pm$  Cr-rich spinel  $\pm$  aluminosilicate glass or albitic  
90 feldspar) have been found in the giant cluster IDP in addition to other common anhydrous IDP components  
91 (Joswiak et al., 2009; Matrajt et al., 2012; Joswiak et al., 2017). Large ( $> 5 \mu\text{m}$ ) crystalline silicate fragments  
92 were extracted from the giant cluster IDP and high-precision oxygen isotope ratios were determined. However,  
93 the fine-grained fraction of the giant cluster IDP has not been studied here, and thus, the sources inferred  
94 from oxygen isotope ratios of large fragments may not represent the whole giant cluster IDP.

95 **2. Analytical procedures**

96 *2.1. Sample preparation*

97       Twenty large crystalline silicate fragments from the giant cluster IDP were extracted under an optical  
98 microscope with a glass needle and washed with hexane to remove surface silicone oil (Joswiak et al., 2017).  
99       The following sample preparation procedures are similar to those for Wild 2 particles previously described by  
100 Joswiak et al. (2012), Nakashima et al. (2012a), and Defouilloy et al. (2017). The fragments were first  
101 embedded into acrylic or epoxy cylinders for ultramicrotomy. Typically, 70-100 nm thick microtome sections  
102 were made using a 45° diamond knife on a Leica Ultracut S ultramicrotome. The sections were then  
103 transferred onto 200 mesh Cu or Au transmission electron microscope (TEM) grids coated with 10 nm thick  
104 carbon films for TEM examination. The remaining portion of each fragment known as a “potted butt” was  
105 removed as a 100 µm cube and pressed into a 1.4 mm diameter indium metal located at the center of an 8 mm  
106 or 25 mm aluminum disk for SIMS analysis. A polished San Carlos olivine (SC-OI) grain was pressed within  
107 500 µm of the sample fragments to serve as a SIMS running standard. The topography of sample surroundings  
108 was checked using a ZYGO NewView white light profilometer at the University of Wisconsin-Madison (UW-  
109 Madison), demonstrating that height differences are typically < 3 µm.

110 *2.2. Electron microscopy*

111       The microtome sections on TEM grids were placed in a double-tilt beryllium sample holder and  
112 studied using a field emission scanning transmission electron microscope (Tecnai TF20 STEM) at the  
113 University of Washington. The STEM is equipped with bright-field and dark-field CCD cameras, a secondary  
114 electron (SE) detector, and a high-angle annular dark-field scanning TEM detector. The analytical procedures  
115 are similar to those described by Joswiak et al. (2009) and Joswiak et al. (2012). Secondary electron and/or  
116 backscattered electron (BSE) images of the potted butts were obtained using a field emission scanning electron  
117 microscope (FE-SEM; JEOL JSM 7000F) at the University of Washington or a conventional tungsten-sourced  
118 SEM (Hitachi S3400) at the UW-Madison. These images were taken under low electron doses with minimal  
119 beam exposure time to minimize electron damage and shrinkage of acrylic resins that could affect sample  
120 topography. In order to improve the accuracy of aiming during SIMS analysis, the candidate positions were

121 marked by removing a  $1\times 1$   $\mu\text{m}$  square of surface carbon (20-30 nm thick) using a focused ion beam SEM  
122 (FIB-SEM; Zeiss Auriga) at the UW-Madison. The carbon coatings were efficiently removed without  
123 damaging the fragments after 90 s sputtering using a focused  $\text{Ga}^+$  ion beam with an acceleration voltage of 30  
124 keV and a beam current of 5 pA (Nakashima et al., 2012a; Defouilloy et al., 2017).

125 *2.3. Oxygen-three isotope analysis*

126 Oxygen isotope ratios of 20 fragments from the giant cluster IDP were determined using the  
127 CAMECA IMS 1280 at the WiscSIMS laboratory of UW-Madison. Measurements were obtained in five  
128 separate sessions. The analytical conditions and procedures for all sessions were generally similar and have  
129 been described in detail in Nakashima et al. (2012a) and Defouilloy et al. (2017). The  $\text{Cs}^+$  primary beam was  
130 focused to  $\sim 2\times 1.5 \mu\text{m}^2$  with an intensity of 2.5-3 pA. The analysis positions were located via FIB marks,  
131 which appear as bright spots (no carbon coating) on the  $^{16}\text{O}^-$  ion image generated by rastering a  $10\times 10 \mu\text{m}^2$   
132 square. Centering FIB marks to the ion image was achieved by first adjusting the sample stage by  $\geq 1 \mu\text{m}$  steps  
133 and then, except for the Nov. 2011 session, shifting the beam position by  $\geq 0.1 \mu\text{m}$  steps using the  
134 “NanoDeflector” system developed in the WiscSIMS laboratory (Defouilloy et al., 2017). Secondary ions  $^{16}\text{O}^-$ ,  
135  $^{17}\text{O}^-$ , and  $^{18}\text{O}^-$  were detected simultaneously using a Faraday cup ( $^{16}\text{O}^-$ ) with  $10^{11} \Omega$  resistance (except for the  
136 Oct. 2017 session which used  $10^{10} \Omega$ ) and two electron multipliers ( $^{17}\text{O}^-$  and  $^{18}\text{O}^-$ ). The count rate of  $^{16}\text{O}^-$  was  
137  $1\text{-}3\times 10^6$  cps (counts per second). The mass resolving power (MRP) was set to  $\sim 6000$  for axis detector ( $^{17}\text{O}$ )  
138 and the contribution of tailing  $^{16}\text{O}^1\text{H}^-$  ions on the  $^{17}\text{O}^-$  signals was negligible (typically  $< 1\%$  correction). Each  
139 analysis required  $\sim 25$  min (including 20 cycles of acquisition of 60 s each), giving average internal precisions  
140 (2SE) of 1.2 ‰, 2 ‰, and 2‰ for  $\delta^{18}\text{O}$ ,  $\delta^{17}\text{O}$ , and  $\Delta^{17}\text{O}$ , respectively. Depending on the number and size of  
141 fragments on a single mount, blocks of 1-7 analyses of sample fragments were bracketed by 6-8 analyses on  
142 the nearby SC-O1 standard. The external reproducibility (2SD) on the SC-O1 standard was typically 1.5‰, 2‰,  
143 and 1.7‰ for  $\delta^{18}\text{O}$ ,  $\delta^{17}\text{O}$ , and  $\Delta^{17}\text{O}$ , respectively; therefore, very similar to the 2SE of individual analysis. For  
144 fragments that show indistinguishable oxygen isotope ratios among multiple analyses, mean values were  
145 calculated. The uncertainties of mean values are propagated from (i) the maxima of the 2SD of multiple  
146 sample analyses versus the 2SD of bracketing standard analyses; (ii) 2SE (standard error of the mean) of

147 instrumental bias calculated from 6-8 bracket standard analyses; (iii) external reproducibility for instrumental  
148 bias correction across a 1 mm center area of an 8 mm disk (0.5‰ in  $\delta^{18}\text{O}$  0.25‰ in  $\delta^{17}\text{O}$ ) (Nakashima et al.,  
149 2012a; Defouilloy et al., 2017). If only one analysis was made for a fragment, we propagated uncertainties  
150 from (ii) and (iii) to the final value. At the beginning of or during each session, multiple standards of olivine  
151 (Fo<sub>100</sub>, Fo<sub>82</sub>, Fo<sub>73</sub>, Fo<sub>60</sub>), orthopyroxene (En<sub>99</sub>, En<sub>96.3</sub>, En<sub>85</sub>, En<sub>70</sub>), diopside (pure CaMgSi<sub>2</sub>O<sub>6</sub>), and/or glass  
152 (SiO<sub>2</sub>: 63.7 wt% and 76 wt%) were analyzed and bracketed by SC-OI analyses (Supplementary S1 Table S1).  
153 Instrumental biases of olivine, pyroxene, and mesostasis in the sample fragments were corrected as a function  
154 of Fo, En, Wo, and SiO<sub>2</sub> contents (Supplementary S1 Table S2). After each session, the locations and  
155 morphology of SIMS pits were examined under similar imaging conditions on the conventional SEM and FIB-  
156 SEM at the UW-Madison (Supplementary S2).

### 157 3. Results

#### 158 3.1. Petrology

159 The 20 fragments were extracted from the largest grain size fraction of the giant cluster IDP. Their  
160 longest dimensions range from 5  $\mu\text{m}$  to 35  $\mu\text{m}$ , eight larger than 10  $\mu\text{m}$  in both length and width. They are  
161 mostly monomineralic (6/20) or polymineralic (9/20) fragments consisting of olivine and/or pyroxene (Table 1;  
162 Figs. 1a, 1b, 1c). Sulfide is rare. Four fragments have chondrule-like mineral assemblages: (i) LT24 is  
163 composed mainly of enstatite with minor amounts of feldspar (An<sub>57</sub>), augite, and crystalline silica (Fig. 1d); (ii)  
164 LT11 consists of enstatite, feldspathic glass/feldspar, and minor kamacite, schreibersite [(Fe, Ni)<sub>3</sub>P], and  
165 pyrrhotite (Fig. 1e); and (iii) LT17 and LT23 have barred-olivine textures composed of olivine bars, chromite,  
166 and mesostasis (Figs. 1f, 1g), of which LT17 has two sets of olivine bars that are almost perpendicular. LT410  
167 is composed mainly of forsterite with minor anorthite and Al-diopside (Fig. 1h), with a mineral assemblage  
168 similar to AOAs and Al-rich chondrules (ARC).

169 Olivine in 13 fragments shows a compositional range from Fo<sub>75</sub> to Fo<sub>99</sub>; eleven are iron-rich (Fo<sub>75-86</sub>)  
170 and two are forsteritic (Fo<sub>99</sub>) (Table 1). Iron-rich olivines contain minor abundances of MnO (0.20-0.54 wt%)  
171 and Al<sub>2</sub>O<sub>3</sub> (0-0.47 wt%) but nearly free of Cr<sub>2</sub>O<sub>3</sub> ( $\leq$  0.12 wt%). In contrast, forsteritic olivines are richer in  
172 Cr<sub>2</sub>O<sub>3</sub> (0.67-0.74 wt%) but poorer in MnO (0.11-0.18 wt%) and free of Al<sub>2</sub>O<sub>3</sub>. CaO content in olivine varies

173 from 0.14 wt% to 0.88 wt% and is independent of their Mg# values. Pyroxene in 13 fragments is enstatite  
174 (6/13), pigeonite (6/13), or augite (1/13). Enstatite is richer in MgO (En<sub>90-98</sub>Wo<sub>≤3.2</sub>) but poorer in Al<sub>2</sub>O<sub>3</sub> (≤ 2.15  
175 wt%) compared to pigeonite (En<sub>72-75</sub>Wo<sub>5-9</sub>; Al<sub>2</sub>O<sub>3</sub>=1.82-5.17 wt%) and augite (En<sub>73</sub>Wo<sub>18</sub>; Al<sub>2</sub>O<sub>3</sub>=3.04 wt%).  
176 Enstatite and augite have higher abundances of Cr<sub>2</sub>O<sub>3</sub> (0.84-1.18 wt% vs. ≤ 0.61 wt%) and MnO (0.15-1.33 wt%  
177 vs. ≤ 0.45 wt%) than pigeonite. Pigeonite generally intergrows with iron-rich olivine (Fig. 1c) and shows  
178 similar Mg# values, except LT14 (Fo<sub>78</sub> vs. En<sub>97</sub>). TiO<sub>2</sub> and Na<sub>2</sub>O contents in pyroxene are low, i.e., ≤ 0.77 wt%  
179 and ≤ 0.25 wt%, respectively. Mesostasis in the two BO chondrule-like fragments (LT17 and LT23) is SiO<sub>2</sub>-  
180 rich (>70 wt%) and contains < 11 wt% MgO, < 7 wt% FeO, < 13 wt% Al<sub>2</sub>O<sub>3</sub>, and < 1 wt% Na<sub>2</sub>O.

181 *3.2. Oxygen isotopes*

182 A total of 73 analyses were performed on olivine (34/73) and pyroxene (39/73) of the 20 fragments in  
183 the giant cluster IDP (Supplementary S1 Table S2). Two analysis points on fragment LT7 (LT7#1, #2) have  
184 significantly higher OH<sup>-</sup> intensities (up to 3.7×10<sup>5</sup> cps) than other points on the same fragment (typically ~  
185 1×10<sup>3</sup> cps), likely indicating a severe overlap with the surrounding epoxy resin and therefore were rejected.  
186 The oxygen isotope ratios among multiple analyses on a monomineralic fragment or olivine and pyroxene of a  
187 polyminalic fragment are homogeneous, with their 2SD typically smaller than or very similar to those of the  
188 running standard SC-OI. Thus, only mean oxygen isotope ratios for each fragment were used in the following  
189 illustrations and discussions. The 20 fragments have mean δ<sup>18</sup>O, δ<sup>17</sup>O, and Δ<sup>17</sup>O ranging from -6.1 ± 0.9‰ to  
190 5.7 ± 1.1‰, -6.4 ± 2.1‰ to 5.2 ± 1.2‰, and -3.2 ± 2.0‰ to 3.4 ± 1.5‰, respectively (Table 2). The four  
191 chondrule-like fragments (LT11, LT17, LT23, and LT24) have oxygen isotope ratios similar to other  
192 monomineralic/polyminalic fragments, with mean δ<sup>18</sup>O, δ<sup>17</sup>O, and Δ<sup>17</sup>O varying from -0.1 ± 1.2‰ to 3.0 ±  
193 1.1‰, -2.4 ± 1.4‰ to 0.7 ± 0.9‰, and -2.4 ± 1.6‰ to -0.8 ± 1.0‰, respectively. The AOA/ARC-like  
194 fragment LT410 has δ<sup>18</sup>O, δ<sup>17</sup>O, and Δ<sup>17</sup>O of -2.1 ± 1.3‰, -2.5 ± 1.7‰, and -1.4 ± 1.4‰, respectively. Δ<sup>17</sup>O  
195 values of these fragments show a negative correlation with their Mg# values, i.e., Mg# > 97 fragments have  
196 Δ<sup>17</sup>O of -3.2 ± 2.0‰ to -0.9 ± 1.8‰, Mg# = 80-97 fragments have Δ<sup>17</sup>O of -3.1 ± 2.2‰ to 1.9 ± 1.4‰, and  
197 Mg# = 70-80 fragments have Δ<sup>17</sup>O of -0.8 ± 1.0‰ to 3.4 ± 1.5‰. In the diagram of δ<sup>18</sup>O vs. δ<sup>17</sup>O (Fig. 2),  
198 oxygen isotope ratios of most fragments are scattered on the primitive chondrule mineral (PCM) line

199 (Ushikubo et al., 2012) and the Young & Russell (Y&R) line (Young and Russell, 1998), while three  
200 fragments (LT7, LT14, and LT400) plot above the terrestrial fractionation (TF) line.  $\delta^{18}\text{O}$  vs.  $\delta^{17}\text{O}$  diagrams  
201 for individual fragments are shown in Figs. S2-S4.

## 202 **4. Discussion**

### 203 *4.1. Petrology comparison of anhydrous IDPs with Wild 2 particles and primitive chondrite* 204 *chondrules*

205 While hydrothermal alteration has been detected rarely in anhydrous IDPs and Wild 2 particles (e.g.,  
206 Hicks et al., 2017), it is acknowledged that they are dominated by anhydrous phases/assemblages like olivine,  
207 pyroxene, Fe-Ni metals, Fe-Ni sulfides, CAIs, and AOAs (Bradley, 2014; Brownlee, 2014; Joswiak et al.,  
208 2017), which are common components in most primitive chondrites. Kool grains, a unique mineral assemblage  
209 identified in anhydrous IDPs and Wild 2 particles, are thought to be the precursor of iron-rich chondrules in  
210 ordinary chondrites, though it has not been observed in any chondrites (Joswiak et al., 2009). However,  
211 enstatite whiskers/platelets, presolar grains, GEMS, carbon, and aromatic compounds that are abundant or  
212 common in anhydrous IDPs appear to be rare or absent in Wild 2 particles (e.g., McKeegan et al., 2006; Ishii et  
213 al., 2008; Stodolna et al., 2012), which is in part due to the alteration of fine-grained and volatile materials  
214 during the collection of Wild 2 particles at a high impact velocity (~6.1 km/s) (Brownlee, 2014). Similarly,  
215 these materials could easily be destroyed during thermal metamorphism and metasomatism events on their  
216 parent bodies, resulting in extreme heterogeneity in abundances and species among different chondrites (e.g.,  
217 Nittler et al., 2019).

218 Olivine and pyroxene in anhydrous IDPs and Wild 2 particles span a similar Mg# range from 100 to  
219 58 without pronounced frequency peaks, in contrast to olivine Mg# in both chondrules and matrix in most  
220 chondrite groups that typically have peak(s) of distributions at >90 and/or 50-80 (Zolensky et al., 2006; Berlin  
221 et al., 2011; Frank et al., 2014). Unlike typical anhydrous IDPs, the Mg# of olivines and pyroxenes in the  
222 fragments of the giant cluster IDP show a bimodal distribution at >90 and 75-80. This is similar to CR  
223 chondrite chondrules that have a strong peak at >90 (Frank et al., 2014) and may have a second peak at ~71 for  
224 Type II chondrules (Berlin et al., 2011). Fe-Mn systematic of olivines in anhydrous IDPs and Wild 2 particles

225 are significantly overlapped with those of CR chondrite chondrules, but nearly cover the entire range of the  
226 chondrule data including those from CO and O chondrites; therefore, these olivines are likely from regions of  
227 CR, CO, and O chondrite chondrules instead of a single source (Brownlee and Joswiak, 2017). Fe vs. Mn in  
228 olivines in the giant cluster IDP fall in the ranges of anhydrous IDPs, Wild 2 particles, and CR chondrite  
229 chondrules. It is noteworthy that fragment LT14, whose olivine and pyroxene appear to be unequilibrated, has  
230 an elevated Mn content that plots close to the correlation line defined by O chondrite chondrules (Fig. 3). Like  
231 Wild 2 particles, iron-rich olivines in the giant cluster IDP have  $\text{Cr}_2\text{O}_3$  ( $\leq 0.12$  wt%) abundances lower than  
232 primitive CO, CR, and O chondrite chondrules (Fig. 4), which may indicate mild thermal metamorphism  
233 (petrologic type 3.05-3.15 when compared with O chondrites and  $>3.1$  when compared with CO chondrites)  
234 (Frank et al., 2014). This low  $\text{Cr}_2\text{O}_3$  content in olivine could also result from chromite precipitation before or  
235 concurrently with olivine (Gainsforth et al., 2015). In addition, pigeonite in the giant cluster IDP typically  
236 occurs as a single crystal intergrown with iron-rich olivine instead of a discrete, intermediate layer between  
237 low-Ca pyroxene and augite overgrowth like in Type II chondrules of O chondrites (Brearley and Jones, 1998),  
238 likely suggesting a carbonaceous chondrite source.

239 One of the important findings from Wild 2 particles is the identification of chondrule-like fragments  
240 (e.g., Nakamura et al., 2008; Gainsforth et al., 2015), while similar objects have rarely been reported among  
241 single anhydrous IDPs. This finding contrast with the five chondrule-like fragments observed out of 20  
242 fragments in the giant cluster IDP. LT11 and LT24 are composed mainly of enstatite with minor feldspathic  
243 glass/feldspar  $\pm$  augite  $\pm$  crystalline silica  $\pm$  kamacite  $\pm$  schreibersite  $\pm$  pyrrhotite (Figs. 1d, 1e), resembling the  
244 porphyritic pyroxene (PP) chondrules. Similar objects such as “Gen-chan,” “Lily,” and “Pyxie” have been  
245 reported in Wild 2 particles (Nakamura et al., 2008; Nakashima et al., 2012a). LT17 and LT23 have typical  
246 barred-olivine textures consisting of iron-rich olivine bars ( $\text{Fo}_{75-86}$ ),  $\text{SiO}_2$ -rich mesostasis ( $> 70$  wt%), and  
247 chromite inclusions (Figs. 1f, 1g), resembling the iron-rich BO chondrules. Similar objects have not been  
248 found in Wild 2 particles. LT410 comprises forsterite ( $\text{Fo}_{99.2}$ ), Al-diopside, and anorthite (Fig. 1h) with a mean  
249  $\Delta^{17}\text{O}$  of  $-1.4 \pm 1.4\text{\textperthousand}$ , confirming that it is an ARC-like fragment. A similar ARC-like particle (“Bidi”) has  
250 been reported in the *Stardust* track 130, composed of forsterite ( $\text{Fo}_{97}$ ), anorthite ( $\text{An}_{97}$ ), and Al-Ti-bearing  
251 clinopyroxene with a mean  $\Delta^{17}\text{O}$  of  $-2.2 \pm 2.0\text{\textperthousand}$  (Joswiak et al., 2014). In summary, the Mg# distribution, Fe-

252 Mn systematics of olivine, Cr<sub>2</sub>O<sub>3</sub> abundance in olivine, and mineral assemblages of chondrule-like fragments,  
253 in anhydrous IDPs (including the giant cluster IDP) are almost indistinguishable from Wild 2 particles.  
254 Similarities in the first two factors may indicate a genetic relationship to CR chondrite chondrules.

255 *4.2. Oxygen isotope systematic comparison of anhydrous IDPs with Wild 2 particles and*  
256 *primitive chondrite chondrules*

257 Limited high-precision oxygen isotope data of olivine and low-Ca pyroxene in anhydrous IDPs show  
258 chondrule-like <sup>16</sup>O-poor compositions, with average  $\delta^{18}\text{O}$ ,  $\delta^{17}\text{O}$ , and  $\Delta^{17}\text{O}$  ranging from  $-2.9 \pm 0.7\text{\textperthousand}$  to  $1.0 \pm$   
259  $2\text{\textperthousand}$ ,  $-5.8 \pm 0.8\text{\textperthousand}$  to  $-1.3 \pm 1.8\text{\textperthousand}$ , and  $-4.3 \pm 0.9\text{\textperthousand}$  to  $-1.8 \pm 2.2\text{\textperthousand}$ , respectively (Aléon et al., 2009;  
260 Nakashima et al., 2012b). New oxygen isotope data of 20 crystalline silicate fragments in the giant cluster IDP  
261 indicate similar chondrule-like <sup>16</sup>O-poor compositions but span a wider range, with average  $\delta^{18}\text{O}$ ,  $\delta^{17}\text{O}$ , and  
262  $\Delta^{17}\text{O}$  varying from  $-6.2 \pm 1.0\text{\textperthousand}$  to  $6.8 \pm 1.9\text{\textperthousand}$ ,  $-6.7 \pm 2.6\text{\textperthousand}$  to  $5.3 \pm 1.2\text{\textperthousand}$ , and  $-5.2 \pm 2.6\text{\textperthousand}$  to  $3.4 \pm 1.5\text{\textperthousand}$ ,  
263 respectively (Table 2). The range of oxygen isotope ratios is slightly narrower than the <sup>16</sup>O-poor silicates in  
264 Wild 2 particles ( $\delta^{18}\text{O}$ :  $-12.2 \pm 1.8\text{\textperthousand}$  to  $7.2 \pm 1.6\text{\textperthousand}$ ;  $\delta^{17}\text{O}$ :  $-13 \pm 2.0\text{\textperthousand}$  to  $5.5 \pm 2.8\text{\textperthousand}$ ;  $\Delta^{17}\text{O}$ :  $-6.9 \pm 4.2\text{\textperthousand}$  to  
265  $2.4 \pm 2.8\text{\textperthousand}$ ; Fig. 2) (Nakamura et al., 2008; Nakashima et al., 2012a; Joswiak et al., 2014; Ogliore et al., 2015;  
266 Defouilloy et al., 2017) but similar to the CR chondrite chondrules ( $\delta^{18}\text{O}$ :  $-6.4 \pm 0.4\text{\textperthousand}$  to  $7.3 \pm 0.4\text{\textperthousand}$ ;  $\delta^{17}\text{O}$ :  $-9.5 \pm 0.8\text{\textperthousand}$  to  $3.8 \pm 0.5\text{\textperthousand}$ ;  $\Delta^{17}\text{O}$ :  $-6.3 \pm 1.1\text{\textperthousand}$  to  $1.1 \pm 0.7\text{\textperthousand}$ ; Fig. 5) (Connolly and Huss, 2010; Schrader et al.,  
267 2013; Schrader et al., 2014; Tenner et al., 2015). LT11 ( $\Delta^{17}\text{O} = -0.9 \pm 1.8\text{\textperthousand}$ ) and LT24 ( $\Delta^{17}\text{O} = -2.3 \pm 1.4\text{\textperthousand}$ )  
268 in the giant cluster IDP have nearly identical oxygen isotope ratios to similar PP chondrule-like particles  
269 “Pyxie” ( $\Delta^{17}\text{O} = -1.1 \pm 0.9\text{\textperthousand}$ ) and “Gen-chan” ( $\Delta^{17}\text{O} = -2.3 \pm 1.4\text{\textperthousand}$ ) in *Stardust* tracks (Nakamura et al.,  
270 2008; Nakashima et al., 2012a). PP chondrules in CR and CV/CO/CM/Acfer 094 chondrites have negative  
271  $\Delta^{17}\text{O}$  values ( $\sim -6\text{\textperthousand}$  to  $\sim 0\text{\textperthousand}$  and  $\sim -7\text{\textperthousand}$  to  $\sim -2\text{\textperthousand}$ , respectively; Ushikubo et al., 2012; Tenner et al., 2013;  
272 Tenner et al., 2015; Chaumard et al., 2018; Hertwig et al., 2018), while similar objects in O and enstatite (E)  
273 chondrites have positive  $\Delta^{17}\text{O}$  values ( $\sim 0\text{\textperthousand}$  to  $\sim 2\text{\textperthousand}$ ; Kita et al., 2010; Weisberg et al., 2011). LT17 ( $\Delta^{17}\text{O} = -2.4 \pm 1.6\text{\textperthousand}$ ) and LT23 ( $\Delta^{17}\text{O} = -0.8 \pm 1.0\text{\textperthousand}$ ) have oxygen isotope ratios similar to iron-rich BO chondrules in  
274 CR and CO chondrites ( $\Delta^{17}\text{O}$ :  $\sim -2\text{\textperthousand}$  to  $\sim 0\text{\textperthousand}$ ; Connolly and Huss, 2010; Tenner et al., 2013; Schrader et al.,  
275 2014). Thus, the four chondrule-like fragments (LT11, LT24, LT17, and LT23) are likely related to

278 carbonaceous chondrite (except CH-CB) chondrules. In contrast, the ARC-like fragment LT410 ( $\Delta^{17}\text{O} = -1.4$   
279  $\pm 1.4\text{\textperthousand}$ ) is relatively  $^{16}\text{O}$ -poor compared to ARCs in CV/CO/Acfer 094 chondrites ( $\Delta^{17}\text{O} \sim -5\text{\textperthousand}$ ) (e.g., Zhang  
280 et al., 2020) but falls in the ranges of ARCs in O, CR, and CH chondrites ( $\Delta^{17}\text{O}: -8\text{\textperthousand}$  to  $+4\text{\textperthousand}$ ) (e.g., Krot et  
281 al., 2017), suggesting an origin similar to the latter.

282 For the mono/polymimetic fragments in the giant cluster IDP, LT7 and LT14 plot within regions of  
283 O and R chondrite chondrules, LT400 plots close to the R or CH-CB chondrite chondrules, and the others plot  
284 within the ranges of CR and CV/CO/CM/Acfer 094 chondrite chondrules (Fig. 5). This comparison suggests  
285 that most fragments have an origin of carbonaceous chondrite chondrules (except CH-CB), and few have  
286 origins of O, R, or CH-CB chondrite chondrules. The possible O chondrite origin of LT14 is consistent with  
287 the high MnO content in its iron-rich olivine plotting close to the line defined by O chondrite chondrules (Fig.  
288 3). Moreover, LT14 is in chemical disequilibrium composed of iron-rich olivine (Fo<sub>78</sub>) and magnesium-rich  
289 enstatite (En<sub>97</sub>) and suggests that olivine underwent Fe-Mg exchange during mild thermal metamorphism on  
290 the O/R-like chondrite parent body. In contrast, enstatite remains intact due to a lower Fe-Mg interdiffusion  
291 rate (Ganguly and Tazzoli, 1994).

292  $\Delta^{17}\text{O}$  of crystalline silicate fragments in the giant cluster IDP appears negatively-correlated with their  
293 Mg# values, where  $\Delta^{17}\text{O}$  of Mg# > 90 fragments typically cluster around  $-3\text{\textperthousand}$ , and their  $\Delta^{17}\text{O}$  gradually  
294 increases to  $\sim 0\text{\textperthousand}$  as Mg# decreases from 90 to 80 and finally cluster around  $0\text{\textperthousand}$  in Mg# = 75-80 fragments  
295 (Fig. 6a). As exceptions, LT14 and LT400 deviate from the Mg#- $\Delta^{17}\text{O}$  trend, consistent with their O, R, or  
296 CH-CB origins; however, LT7 and LT410 plot on this trend. For the remaining fragments, the Pearson  
297 correlation coefficient for their  $\Delta^{17}\text{O}$  and Mg# is  $-0.74$ , indicating that the two parameters are negatively  
298 correlated. The Mg#- $\Delta^{17}\text{O}$  trend of the giant cluster IDP is very similar to the  $^{16}\text{O}$ -poor Wild 2 particles, i.e.,  
299 Mg# > 97 particles have  $\Delta^{17}\text{O}$  of  $\sim -2\text{\textperthousand}$  and Mg# < 97 particles have  $\Delta^{17}\text{O}$  varying from  $-4\text{\textperthousand}$  to  $+2\text{\textperthousand}$  (Fig.  
300 6b). To quantify the similarity of Mg#- $\Delta^{17}\text{O}$  relationship between crystalline silicates in the giant cluster IDP  
301 and Wild 2 particles, a two samples Kolmogorov-Smirnov (K-S) test for two variables (Mg# and  $\Delta^{17}\text{O}$ ) was  
302 performed. This test compares the cumulative distributions of two datasets and returns a *p*-value representing a  
303 probability that the two datasets belong to the same distribution. The K-S test returns a *p*-value of 0.28 for

304 crystalline silicates in the giant cluster IDP and Wild 2 particles, indicating that they are likely from the same  
305 population.

306 On the other hand, the Mg#– $\Delta^{17}\text{O}$  correlation in the giant cluster IDP allows for comparing with those  
307 in primitive chondrite chondrules and then elucidating the genetic relationships (Fig. 6). While the crystalline  
308 silicate fragments in the giant cluster IDP might have formed via high-temperature annealing ( $\sim 1000 \text{ K}$ ) in the  
309 inner solar system (Nuth et al., 2000), the occurrence of igneous chondrule-like fragments suggest that it is  
310 also probable that they are pulverized dust of chondrules (Bridges et al., 2012). It is well-established that  
311 chondrules in different chondrite groups have different Mg#– $\Delta^{17}\text{O}$  trends (i) CV, CO, CM, and the ungrouped  
312 Acfer 094 chondrite chondrules are dominated by two populations, i.e., Mg# > 97 with  $\Delta^{17}\text{O}$  ranges from  $-6\text{\textperthousand}$   
313 to  $-4\text{\textperthousand}$  and Mg#  $\leq 97$  with  $\Delta^{17}\text{O} \sim -2\text{\textperthousand}$  (Ushikubo et al., 2012; Tenner et al., 2013; Chaumard et al., 2018;  
314 Hertwig et al., 2018); (ii) porphyritic chondrules in CH-CB chondrites have  $\Delta^{17}\text{O}$  decreasing from  $+4\text{\textperthousand}$  to  $-5\text{\textperthousand}$   
315 as their Mg# decreases from 99 to 96, and then varying irregularly between  $-2\text{\textperthousand}$  to  $+3\text{\textperthousand}$  when Mg# < 96  
316 (Krot et al., 2010; Nakashima et al., 2019); and (iii) O and R, E and K chondrules have nearly zero or slightly  
317 positive  $\Delta^{17}\text{O}$  values independent of their Mg# values (Kita et al., 2010; Weisberg et al., 2011; Nagashima et  
318 al., 2015; Miller et al., 2017); (iv) CR chondrite chondrules show a negative correlation between their  $\Delta^{17}\text{O}$   
319 and Mg#, that is,  $\Delta^{17}\text{O}$  gradually increases from  $-6\text{\textperthousand}$  to  $-1\text{\textperthousand}$  as Mg# decreases from 99 to 94, and the rest,  
320 with Mg# < 90, show variable  $\Delta^{17}\text{O}$  between  $-2\text{\textperthousand}$  and  $+2\text{\textperthousand}$  (Connolly and Huss, 2010; Schrader et al., 2013;  
321 Schrader et al., 2014; Tenner et al., 2015). The negative correlation between Mg# and  $\Delta^{17}\text{O}$  was explained as  
322 adding  $^{16}\text{O}$ -poor  $\text{H}_2\text{O}$  ice ( $\Delta^{17}\text{O} > 0\text{\textperthousand}$ ) to the reduced anhydrous dust ( $\Delta^{17}\text{O} = -6\text{\textperthousand}$ ) in the chondrule-forming  
323 region, which increases the dust/gas ratios and results in the formation of more oxidizing (lower Mg#)  
324 chondrules (Tenner et al., 2015). Obviously, the Mg#– $\Delta^{17}\text{O}$  trend in the giant cluster IDP is closest to the CR  
325 chondrite chondrules, where the K-S test returns a *p*-value of 0.051 (slightly higher than the commonly used  
326 threshold of 0.05) that supports they are possibly from the same population. The relatively low *p*-value is  
327 partly due to the higher abundance of iron-rich particles in the giant cluster IDP (11/20) than CR chondrite  
328 chondrules ( $\sim 2 \text{ vol\%}$ ) (Schrader et al., 2015), indicating that they formed in a more oxidizing environment  
329 with higher dust density. In contrast, the returned *p*-values for fragments in the giant cluster IDP and  
330 chondrules in other primitive chondrites are typically  $< 10^{-3}$ , rejecting a genetic relationship between them.

331     4.3. *Transportation of crystalline silicate fragments to the comet-forming region*

332         As discussed above, crystalline silicate fragments in anhydrous IDPs (including the giant cluster IDP)  
333         are similar to those in Wild 2 particles on petrography, mineral chemistry, and oxygen isotope systematics,  
334         supporting a cometary origin of anhydrous IDPs. Among the 20 fragments in the giant cluster IDP, LT7, LT14,  
335         and possibly LT410 have an origin where O or R chondrite chondrules formed, fragment LT400 has affinities  
336         to R or CH-CB chondrules, and the remaining ones are closest to CR chondrite chondrules. Since *in situ*  
337         production of chondrules in the Kuiper belt region is unlikely (Nakamura et al., 2008), we believe that the  
338         giant cluster IDP accreted diverse materials formed in a wide solar system region like Wild 2 particles  
339         (Brownlee, 2014). However, the transportation mechanism of high-temperature materials into the comet-  
340         forming region remains unclear.

341         During the early expansion of an initially quite compact disk, turbulent diffusion can rapidly transport  
342         high-temperature inner-disk particles to the 25-30 AU comet-forming region, especially near the disk midplane  
343         (Ciesla, 2007). This model explains the presence of the oldest solid materials, CAIs and AOAs, in the comets.  
344         However, as the disk evolves, radial drift dominates over disk expansion, and therefore the outward velocity  
345         and flux decreases as more and more materials are lost onto the central star. While the presence of O chondrite  
346         chondrule-like fragments, which might have crystallized  $\sim$ 2 Ma after CAI formation (e.g., Siron et al., 2021),  
347         is consistent with a late accretion time ( $\geq$  2.6 Ma after CAI formation) of comet Wild 2 (e.g., Ogliore et al.,  
348         2012; Nakashima et al., 2015), it is unknown whether turbulent diffusion can transport these late-formed  
349         fragments to the disk beyond 25 AU. Furthermore, the O/R chondrite chondrule-like fragment LT14, whose  
350         minerals are apparently in chemical disequilibrium, is thought to have undergone thermal metamorphism on  
351         the O/R-like parent body. The timing of the thermal metamorphism could have been as early as 2.5 Ma after  
352         CAI formation when the center of a  $\geq$ 100 km planetesimal was quickly raised above 800°C before reaching  
353         maximum central temperatures of 850-950°C at  $\sim$ 5 Ma (inferred peak metamorphic temperature for H6 and L6  
354         chondrites) (Blackburn et al., 2017). Another important constraint is that the outward transportation of O  
355         chondrite chondrule-like fragments may be blocked by Jupiter's core, which is thought to grow up to  $\sim$ 20 M<sub>E</sub>  
356         (M<sub>E</sub>=the mass of the Earth) within 1 Ma (Kruijer et al., 2017). However, more and more O chondrite-like

357 chondrules and O chondrite-like relict olivine grains have been identified in carbonaceous chondrites by their  
358 distinct O, Cr, and Ti isotope signatures and mineral and chemical characteristics (e.g., Hertwig et al., 2018;  
359 Schrader et al., 2020; Williams et al., 2020). Thus, chondrule-sized or smaller fragments have nevertheless  
360 been transported from the O chondrite-forming region to the carbonaceous chondrite-forming regions  $\sim$ 2-3 Ma  
361 after CAI formation. Furthermore, these fragments may have been transferred farther to the comet-forming  
362 region if they were smashed in the inner disk (Bridges et al., 2012).

363 Hughes and Armitage (2010) presented a series of 1D models to explore the effects of particle size,  
364 disk evolution, and radial flow dynamics on the evolution of particles within a gas disk. They found that  $20\text{ }\mu\text{m}$   
365 particles (poorly-coupled with the gas motions) could not exist in the disk beyond 25 AU for  $>1$  Ma under the  
366 accretion-flow dynamics. The O chondrite chondrule-like fragment LT7 with a size of  $22 \times 12\text{ }\mu\text{m}^2$  found in  
367 the giant cluster IDP possibly suggests that this particle was retained in the disk beyond 25 AU for a relatively  
368 short period. Recently, Atacama Large Millimeter/submillimeter Array (ALMA) showed multiple well-defined  
369 and nearly perfectly circular rings in a large fraction of the protoplanetary disks, likely due to dust trapping in  
370 radial pressure bumps, indicating that the gas density does not always decrease with distance from the Sun  
371 (Dullemond et al., 2018). Thus, the coarse fragments could have been trapped in comparable circular rings for  
372 a certain period before comet accretion.

373 For the remaining CR-CH-CB chondrite chondrule-like fragments, the transportation distance is much  
374 shorter as parent bodies of these chondrites are considered to accrete at the disk beyond the orbits of the gas  
375 giants since they preserved significant amounts of thermally unprocessed,  $^{26}\text{Al}$ -free primordial molecular cloud  
376 matters that may only survive in the outer part of the solar system (Van Kooten et al., 2016). Desch et al. (2018)  
377 constructed a protoplanetary disk based on the abundances of refractory elements and refractory inclusions in  
378 different chondrite groups and suggested that CR chondrites' parent body accreted  $\sim$ 4-5 Ma after CAI  
379 formation at 3.84-4 AU. At that time, most or all of the gas could have been lost via accretion onto the central  
380 star or dispersed via the photoevaporation process, and therefore particle transportation was likely limited.  
381 While more evidence is necessary to validate this hypothesis, it is reasonable to infer that the comets contain  
382 predominantly CR chondrite chondrule-like materials which formed and accreted nearby.

383 **5. Conclusions**

384 High precision ( $\leq 2\text{\textperthousand}$  in  $\Delta^{17}\text{O}$ ) oxygen isotope ratios of individual crystalline silicates in 20 fragments  
385 from a giant cluster IDP U2-20GCA were determined using a SIMS IMS-1280. The fragments are mainly  
386 monomineralic (6/20) or polymimetic (9/20) and composed of olivine and/or pyroxene, with the longest  
387 dimension ranging from 5  $\mu\text{m}$  to 35  $\mu\text{m}$ . Others are chondrule-like fragments including two BO-like, two PP-  
388 like, and one ARC-like. Unlike other anhydrous IDPs and Wild 2 particles showing Mg# distributions evenly  
389 from 100 to 58, the giant cluster IDP has an Mg# distribution bimodally at  $> 90$  and 75-80. Fe vs. Mn  
390 abundances in olivine of anhydrous IDPs are uncorrelated and span a wide range similar to Wild 2 particles  
391 and CR chondrite chondrules. Low chromium concentrations ( $\leq 0.1$  wt%  $\text{Cr}_2\text{O}_3$ ) in the giant cluster IDP's iron-  
392 rich olivines may suggest that they underwent mild thermal metamorphism like Wild 2 particles. The  
393 fragments are  $^{16}\text{O}$ -poor with  $\delta^{18}\text{O}$  and  $\delta^{17}\text{O}$  varying from  $-6.2 \pm 1.0\text{\textperthousand}$  to  $6.8 \pm 1.9\text{\textperthousand}$  and  $-6.7 \pm 2.6\text{\textperthousand}$  to  $5.3 \pm$   
394  $1.2\text{\textperthousand}$ , respectively. This range is similar to the CR chondrite chondrules but narrower than the Wild 2  $^{16}\text{O}$ -  
395 poor particles.  $\Delta^{17}\text{O}$  are systematically lower for fragments with higher Mg# ( $> 90$ ) than those with lower Mg#  
396 (70-80), showing a similar trend to Wild 2 particles and CR chondrite chondrule-like materials. These similarities  
397 demonstrate a cometary origin of anhydrous IDPs, and CR chondrite chondrule-like materials are likely the  
398 main source of crystalline silicates in the parent comets of anhydrous IDPs.

399 As exceptions, three fragments have resolvable positive  $\delta^{18}\text{O}$ ,  $\delta^{17}\text{O}$  values that overlap with O-R  
400 chondrite chondrules (LT7 and LT14) and CH-CB chondrite chondrules (LT400), suggesting origins similar to  
401 O, R, or CH-CB chondrite chondrules, respectively. LT14, composed of iron-rich olivine and magnesium-rich  
402 pyroxene, likely experienced thermal metamorphism on the O/R chondrite parent body.  $\Delta^{17}\text{O}$  of the ARC-like  
403 fragment LT410 ( $-1.4 \pm 1.4\text{\textperthousand}$ ) falls in the range of ARCs in O and CR-CH chondrites. The presence of O  
404 chondrite chondrule-like fragments in the giant cluster IDP, which likely crystallized 2-3 Ma after CAI  
405 formation, suggests that inner solar system materials were transported into the comet-forming region  
406 independent of Jupiter's existence.

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611

612 **Figure Captions**

613 Fig. 1. BSE (a-c, e-g) and TEM bright-field (d, h) images of eight representative crystalline silicate fragments  
614 in the giant cluster IDP. *Abbreviation:* Sul = sulfide; Ka = kamacite; Sch = schreibersite; Mt = magnetite  
615 (formed during atmospheric entry); Po = pyrrhotite; Gl = feldspathic glass; Chr = chromite; Al-Di = Al-  
616 diopside; An = anorthite; Pl = plagioclase; others used as previous. Scale bars are 5  $\mu\text{m}$ . [Color is not needed  
617 in print]

618 Fig. 2. Mean oxygen isotope ratios of crystalline silicate fragments in the giant cluster IDP. Data of crystalline  
619 silicate fragments in Wild 2 particles (Nakamura et al., 2008; Nakashima et al., 2012a; Joswiak et al., 2014;  
620 Ogliore et al., 2015; Defouilloy et al., 2017) and other anhydrous IDPs (Aléon et al., 2009; Nakashima et al.,  
621 2012b) are shown for comparison. Black arrows indicate Chondrule-like fragments and the three  
622 mono/polymineralic fragments (Lt4, LT14, and LT400) with positive oxygen isotope ratios that plot above the  
623 terrestrial fractionation (TF) line. *Abbreviation:* CCAM = carbonaceous chondrite anhydrous mineral line  
624 (Clayton et al., 1973); Y&R = Young & Russell line (Young and Russell, 1998); PCM = primitive chondrule  
625 minerals line (Ushikubo et al., 2012). [Color is not needed in print]

626 Fig.3. Fe vs. Mn (afu = atomic formula units based on four oxygen atoms) of olivine in crystalline silicate  
627 fragments of the giant cluster IDP. Data of other anhydrous IDPs (Brownlee and Joswiak, 2017), Wild 2  
628 particles (Joswiak et al., 2012; Frank et al., 2014; Brownlee and Joswiak, 2017), and CR chondrite chondrules  
629 (Schrader et al., 2015) are shown for comparison. LT14 with elevated MnO content plots close to the line  
630 defined by O chondrite (OC), while other iron-rich olivines plot between the two lines defined by O and CO  
631 chondrites (Berlin et al., 2011). [Color is not needed in print]

632 Fig.4. Fa [atom% Fe/(Fe+Mg)] vs.  $\text{Cr}_2\text{O}_3$  of olivine in crystalline silicate fragments of the giant cluster IDP.  
633 Data of Wild 2 particles and O chondrites (L/LL3.0-3.1 and LL3.2) [(Frank et al., 2014) and references  
634 therein], CO3.0-3.1 (Davidson et al., 2019), and CR (Schrader et al., 2015) chondrites are plotted for  
635 comparison. The iron-rich olivines are depleted in  $\text{Cr}_2\text{O}_3$  like Wild 2 particles, possibly suggesting mild  
636 thermal metamorphism. [Color is not needed in print]

637 Fig.5. Mean oxygen isotope ratios of crystalline silicate fragments in the giant cluster IDP as compared with  
638 chondrules in CR (Connolly and Huss, 2010; Schrader et al., 2013; Schrader et al., 2014; Tenner et al., 2015),  
639 CH-CB (Krot et al., 2010), CV (Hertwig et al., 2018), CO (Tenner et al., 2013), CM (Chaumard et al., 2018),  
640 the ungrouped Acfer 094 (Ushikubo et al., 2012), O (Kita et al., 2010), R (Miller et al., 2017), K (Nagashima  
641 et al., 2015), and E chondrites (Weisberg et al., 2011). In O-R and E-K groups, chondrules have nearly  
642 identical oxygen isotope ranges and plots in the same legends for simplicity. [Color is not needed in print]

643 Fig. 6. Mg# vs.  $\Delta^{17}\text{O}$  of crystalline silicate fragments in the giant cluster IDP (a) compared with Wild 2  
644 particles (b) and primitive chondrite chondrules (c, d). Data are from references listed in the caption of Fig. 5.  
645 In panel (a), literature data (Aléon et al., 2009), and fragments that likely have O-R or CH-CB origins and  
646 other fragments in the giant cluster IDP, are shown in different legends. [Color is not needed in print]

647

Table 1. EDX major elements of crystalline silicate fragments in the giant cluster IDP

Fragment	Size	Petrography	Target	SiO <sub>2</sub>	MgO	FeO	CaO	Al <sub>2</sub> O <sub>3</sub>	Cr <sub>2</sub> O <sub>3</sub>	MnO	TiO <sub>2</sub>	Na <sub>2</sub> O	Fo/En	Wo
LT2	22 × 18	Polymineral	Ol	38.3	40.8	19.8	0.18	0.47	0.12	0.40	---	---	78.8	
			Pig	55.5	24.9	11.0	3.66	4.08	0.56	0.29	---	---	74.0	7.78
LT4	35 × 10	Monomineral	En	60.7	33.9	3.30	0.40	0.15	0.84	0.52	---	0.25	94.1	0.79
LT5	15 × 25	Polymineral	En	57.8	32.4	4.52	1.47	1.29	1.18	1.33	---	---	90.1	2.92
LT6	15 × 10	Polymineral	Ol	37.9	39.2	22.5	0.15	---	---	0.23	---	---	75.8	
			Pig	53.0	26.8	15.8	2.12	1.82	0.24	0.27	---	---	72.2	4.08
LT7	22 × 12	Polymineral	Ol	37.4	40.8	21.4	0.14	---	---	0.21	---	---	77.4	
			Pig	54.6	27.1	13.0	2.53	1.92	0.61	0.24	---	---	75.0	5.00
LT9	11 × 4	Monomineral	Pig	52.5	25.5	11.2	4.39	5.17	0.28	0.45	0.55	---	73.1	9.00
LT11	12 × 6	Chondrule-Frag	En	57.9	34.9	1.56	1.64	2.15	1.14	0.40	0.36	---	94.5	3.17
LT13	10 × 6	Monomineral	Ol	39.3	39.4	20.8	0.22	---	---	0.30	---	---	77.4	
LT14	5 × 5	Polymineral	Ol	38.0	40.9	20.5	---	---	0.10	0.54	---	---	78.2	
			En	57.6	38.7	2.53	---	---	1.07	0.15	---	---	96.5	0.00
LT16	15 × 12	Polymineral	Ol	37.5	41.0	21.2	---	---	---	0.25	---	---	77.7	
			Pig	54.1	27.3	12.4	2.84	3.10	---	0.33	---	---	75.3	5.60
LT17	7 × 6	BO chondrule	Ol + Mes	39.9	46.2	13.4	---	---	---	0.50	---	---	86.1	
LT18	7 × 5	Monomineral	En	58.4	37.6	0.81	0.61	0.99	0.89	0.31	0.36	---	97.7	1.13
LT19	13 × 7	Monomineral	Ol	37.5	40.6	21.3	---	0.28	---	0.25	---	---	77.4	
LT20	15 × 4	Polymineral	Aug	52.8	26.8	5.89	9.04	3.04	0.99	0.46	0.77	0.15	73.4	17.66
LT23	11 × 8	BO chondrule	Ol + Mes	40.1	36.6	21.5	0.49	---	0.38	0.94	---	---	75.3	
LT24	28 × 14	Chondrule-Frag	En	61.9	31.1	3.85	0.87	0.92	0.90	0.36	0.10	---	91.9	1.83
LT26	11 × 7	Polymineral	Ol	38.4	39.8	20.7	0.88	---	---	0.20	---	---	77.6	
			Pig	54.8	26.2	13.5	2.71	1.89	0.56	0.29	---	---	73.5	5.43
LT35	15 × 25	Polymineral	Ol	43.1	54.9	0.78	0.24	---	0.74	0.18	---	---	99.2	
LT400	5 × 3	Monomineral	Ol	39.2	39.2	20.8	0.26	0.29	---	0.26	---	---	77.2	
LT410	9 × 5	ARC-like	Ol	42.5	55.8	0.84	0.16	---	0.67	0.11	---	---	99.2	

648 Fo = mol% Mg/(Mg+Fe); En = mol% Mg/(Mg+Fe+Ca); Wo = mol% Ca/(Mg+Fe+Ca). Abbreviations: Chondrule-Frag = chondrule fragment; BO =  
 649 barred-olivine; Ol = olivine; Pig = pigeonite; En = enstatite; Mes = mesostasis; Aug = augite; ARC = Al-rich chondrule.

Table. 2 Mean oxygen isotope ratios of crystalline silicate fragments in the giant cluster IDP

IDPs	Mineral	Mg#	N	$\delta^{18}\text{O}$	$2\sigma$	$\delta^{17}\text{O}$	$2\sigma$	$\Delta^{17}\text{O}$	$2\sigma$
LT2	Ol	78.8	4	3.97	1.32	1.86	1.77	-0.20	1.61
	Pig	80.2	5	5.67	1.13	3.24	1.27	0.29	0.98
	<i>Average</i>	79.5	9	4.91	1.11	2.63	1.18	0.07	0.93
LT4	En	94.9	5	-1.30	1.02	-3.78	2.17	-3.10	2.43
LT5	En	92.8	5	-2.86	1.56	-1.83	1.60	-0.35	1.73
LT6	Ol	75.8	3	5.06	1.84	3.98	1.28	1.36	1.37
	Pig	72.3	1	4.50	2.61	3.06	1.86	0.72	2.02
	<i>Average</i>	74.0	4	4.92	1.68	3.75	1.17	1.20	1.24
LT7	Ol	77.4	1	0.69	1.08	1.08	2.36	0.72	2.02
	Pig	75.0	3	-1.22	2.61	0.31	1.69	0.94	1.97
	<i>Average</i>	76.2	4	-0.74	2.11	0.50	1.54	0.88	1.51
LT9	Pig	80.3	1	0.09	1.74	-1.61	3.08	-1.66	2.68
LT11	En	97.6	1	1.11	1.60	-0.31	1.37	-0.89	1.76
LT13	Ol	77.4	4	3.12	1.15	1.52	0.87	-0.10	0.97
LT14	Ol	78.2	1	-0.06	0.56	0.35	1.63	0.38	1.41
	En	96.0	1	0.64	0.56	2.21	1.63	1.88	1.41
	<i>Average</i>		2	0.29	0.89	1.28	1.99	1.13	1.60
LT16	Ol	77.7	2	2.74	1.49	0.67	1.70	-0.75	2.15
	Pig	75.4	2	1.06	1.49	0.08	1.67	-0.47	1.05
	<i>Average</i>	76.6	4	1.90	1.40	0.38	1.33	-0.61	1.07
LT17	Ol + Mes	86.2	3	-0.14	1.17	-2.43	1.36	-2.36	1.59
LT18	En	98.8	4	-0.10	1.20	-2.86	1.17	-2.81	1.35
LT19	Ol	77.4	2	0.09	1.74	-1.61	3.08	-1.66	2.68
LT20	Aug	89.1	2	-1.85	1.51	-3.74	2.04	-2.78	1.84
LT23	Ol + Mes	75.3	5	1.65	1.12	0.05	0.81	-0.81	1.03
LT24	En	93.6	4	1.09	0.69	-1.70	1.34	-2.27	1.35
LT26	Ol	77.6	2	3.17	1.28	2.34	1.87	0.69	1.56
	Pig	77.8	1	1.47	1.44	-0.12	2.27	-0.89	1.72
	<i>Average</i>	77.7	3	2.60	1.39	1.52	2.11	0.17	1.50
LT35	Ol	99.2	2	-6.09	0.93	-6.36	2.08	-3.20	2.00
LT400	Ol	77.2	1	3.35	0.93	5.16	1.35	3.42	1.63
LT410	Ol	99.2	4	-1.82	1.30	-2.29	1.65	-1.35	1.44











