

Oxygen isotopic and chemical composition of chromites in micrometeorites: Evidence of ordinary chondrite precursors

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1	Title:
2 3	Oxygen isotopic and chemical composition of chromites in micrometeorites: Evidence of ordinary chondrite precursors
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29 Abstract

We identified 66 chromite grains from 42 of ~5000 micrometeorites collected from Indian 30 Ocean deep-sea sediments and the South Pole water well. To determine the chromite grains 31 precursors and their contribution to the micrometeorite flux, we combined quantitative 32 electron microprobe analyses and oxygen isotopic analyses by high-resolution secondary ion 33 mass spectrometry. Micrometeorite chromite grains show variable O isotopic compositions 34 with δ^{18} O values ranging from -0.8 to 6.0‰, δ^{17} O values from 0.3 to 3.6‰, and Δ^{17} O values 35 from -0.9 to 1.6‰, seems majority of them are similar to those of chromites from ordinary 36 chondrites. The oxygen isotopic compositions of olivine as a proxy to the chromite in 37 chromite-bearing micrometeorites where we could not measure chromite have Δ^{17} O values 38 suggesting largely related to ordinary chondritic with some having carbonaceous 39 chondrite precursors. Furthermore, the chemical compositions of chromites in 40 micrometeorites are close to those reported for ordinary chondrite chromites, but some 41 contribution from carbonaceous chondrites cannot be ruled out. Consequently, 42 carbonaceous chondrites cannot be a major contributor of chromite-bearing micrometeorites. 43 Based on their oxygen isotopic and elemental compositions, we thus conclude with no 44 ambiguity that chromite-bearing micrometeorites are largely related to fragments of 45 ordinary chondrites with small fraction from carbonaceous chondrites, unlike other 46 micrometeorites deriving largely from carbonaceous chondrites. 47

49 **1. Introduction**

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The Earth's upper atmosphere accretes nearly 30,000 tons of extraterrestrial dust 51 particles (from tens of micrometers to a few millimeters in size) every year: these particles 52 are sourced either from collisions between asteroidal bodies or trails of cometary bodies 53 during perihelion passage (Brownlee 2001). Most (~90%) of these micrometeorites are 54 ablated during atmospheric entry, and only a small percentage reach the Earth's surface 55 intact. They are collected from various environments such as deep-sea sediments (DSS), 56 57 Antarctic ice, or in the stratosphere during entry and provide information on the varied objects that existed in the early solar system (Love and Brownlee 1993; Taylor et al. 1998; 58 Peucker-Ehrenbrink and Ravizza 2000; Plane 2012; Prasad et al. 2013). 59

60

Ordinary chondrites are the most abundant meteorites, accounting for ~80% of all 61 falls (Hutchison, 2004); of those falls, L and H chondrites dominate (~45% each) and ~10% 62 63 are LL chondrites (Schmitz and Tassinari 2001). This subdivision may preferentially represent the ordinary chondrite component in the inner region of the asteroid belt, where 64 meteorites break down more easily during collisional processes (Meibom and Clark 1999). 65 However, micrometeorites are sourced to a large extent from bodies analogous to 66 carbonaceous chondrites (>70-90%) and probably sample a more diverse population of 67 68 carbonaceous-type asteroids than represented by meteorites (Love and Brownlee 1993; Beckerling and Bischoff 1995; Brownlee et al. 1997; Yada et al. 2005; Rudraswami et al. 69 2015a). In addition, a few to ~30% of micrometeorites have asteroidal precursors related to 70 ordinary chondrites, ~1-2% to iron meteorites, and fewer than a percent to HED bodies 71 (Taylor et al. 2007; Suavet et al. 2010; Rudraswami et al. 2015a, b, 2016b). 72

74 The chemical compositions of micrometeorites are modified during atmospheric entry, complicating the identification of their precursors based solely on chemical 75 composition. Since oxygen diffuses slower than cations (Ryerson et al. 1989), the oxygen 76 77 isotopic characteristics of micrometeorites should be better preserved and thus represent a powerful tool for deciphering micrometeorite origins. For instance, oxygen isotopic 78 investigations have indicated the contribution of ordinary chondrites (or precursors analogous 79 to ordinary chondrites) to be only a few percent for micrometeorites sized 100-300 µm 80 (Rudraswami et al. 2015b, 2016a), but \sim 30% for larger micrometeorites (\sim 600–1000 µm). It 81 82 should be noted, however, that this size range comprises much less than 1% of the total 83 micrometeorite flux by mass (Suavet et al. 2010).

84

85 Chromite is a refractory, high-temperature mineral that is robust to acid treatments 86 and weathering (Schmitz et al. 2001, 2003; Schmitz and Häggström 2006), and spherical to euhedral chromite grains tens of micrometers in size are largely found in ordinary chondrites. 87 88 Based on their oxygen isotopic and chemical compositions, it has been proposed that micrometeorite chromites could have largely survived parent-body collisional events, 89 90 frictional heating during atmospheric entry, and subsequent terrestrial alteration. However, chromite is rarely found in micrometeorites, and earlier studies reported only about a dozen 91 chromite grains among ~500 spherules analyzed, which were conceivably linked to L/H 92 93 chondrites (Van Ginneken et al. 2012; Prasad et al. 2015). The rarity of chromite grains characterized in micrometeorites limits the information we can obtain on their chemical and 94 isotopic characteristics, and thus the micrometeorite sources in the asteroid belt. Here we 95 determine the chemical and oxygen isotopic compositions of a suite of micrometeorite 96 chromites from DSS and Antarctic ice, and compare their Δ^{17} O values with those of ordinary 97 chondrites to assess and distinguish the nature of micrometeorite precursors. 98

100 2. Sampling method

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Micrometeorites were collected from Indian Ocean **DSS** and Antarctica. 102 Micrometeorites from Indian Ocean DSS (74–76° E, 10–13° S) were collected using a grab 103 sampler measuring 50 cm long \times 50 cm wide \times 15 cm deep with the capacity to lift 45 kg of 104 sediments from the seafloor depth of ~5000 m (Rudraswami et al. 2011, 2012; Prasad et al. 105 2013). Retrieved sediments were sieved using a ~200 µm mesh, and further dried and 106 magnetically separated. The estimated terrestrial age of cosmic spherules from DSS at 0-15 107 cm depth based on the peak tektite abundance from three sediment cores is ~0-50,000 a 108 109 (Prasad et al. 2013). Antarctic micrometeorites were collected from the South Pole water well 110 that supplies water to the Amundsen-Scott South Pole Station. The 24 m diameter well is situated at ~100 m below the surface, contains ~5000 m^3 of melt water, and extends to 16 m 111 depth below the well opening (Taylor et al. 1998, 2000). Particles from the well bottom were 112 suctioned by a sample collector and seived to various size fractions >50 µm. A binocular 113 microscope was used to pick and mount samples in epoxy for analyses. Antarctic and DSS 114 micrometeorites were mounted on 2 and 16 epoxy mounts, respectively, and polished to 115 expose their surface diametrically. The sections were cleaned using alcohol and distilled 116 water in an ultrasonic bath, then dried. Each section was carbon coated for observation under 117 118 a scanning electron microscope (SEM) to identify chromites before chemical and isotopic characterization by electron and ion microprobe, respectively. 119

120

121 **3.** Analytical techniques

122 **3.1. Electron microscopy and microprobe analyses**

SEM and microprobe analyses were performed at the National Institute of 124 Oceanography (Goa, India). A JEOL JSM-IT300LV SEM equipped with an energy 125 dispersive spectrometer (AZtecEnergy EDS Microanalysis, Oxford instruments) was used to 126 127 identify chromites in the micrometeorites and obtain back-scattered electron images. Chromite-bearing micrometeorites were further analyzed using a Cameca SX5 Electron 128 Micro Probe Analyzer equipped with four spectrometers. Microprobe analyses were 129 performed with ~15 kV accelerating voltage, ~12 nA beam current, and ~1-2 µm beam 130 diameter. The PAP model was used for matrix corrections (Pouchou and Pichoir 1991). 131 132 Multiple analyses were performed on each spherule containing different mineral phases to obtain bulk chemical compositions by calculating the exposed area of different mineral 133 phases in ImageJ or LISPIX. We note that the reported bulk compositions do not account for 134 135 density differences among phases. For scoriaceous, barred, cryptocrystalline, or glassy micrometeorites, several analyses were performed using a ~5 µm beam diameter and 136 averaged to obtain the bulk chemical composition; the beam size was restricted to 5 μ m to 137 avoid void spaces, vesicles, and pits. To improve the accuracy and statistical error on minor 138 element measurements (Ca, V, Mn, Ti, Zn), analyses were performed with long counting 139 times of 60 s on peak and 20 s on background. Replicate measurements of multiple standards 140 were within 1% error for major elements and within a few percent for minor elements. 141 Multiple analyses were performed at different spots in each chromite grain to constrain their 142 143 chemical heterogeneity.

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145 **3.2. Oxygen isotopic analyses**

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147 Oxygen isotopic analyses of micrometeorite olivine (here representing the 148 micrometeorite composition) and chromite phases were performed using a Cameca 1270 E7

at CRPG-CNRS (Nancy, France) using a Cs^+ beam with ~1 nA primary current and ~5 μ m 149 beam spot on the sample surface. We used a mass resolving power of ~2500 for 16 O and 18 O 150 and ~7000 for ¹⁷O to avoid ¹⁶OH⁻ interference in the ¹⁷O peak. Secondary ¹⁶O and ¹⁸O ions 151 were measured using a faraday cup while ¹⁷O was measured using the electron multiplier in 152 multi-collection mode. We corrected for instrumental mass fractionation in olivine grains 153 using San Carlos olivine. Stillwater chromite (Schmitz et al., 2016) and UWCr-3 chromite 154 standards were used to check the stability and reproducibility of the isotopic data (Heck et al. 155 2010), and the Stillwater chromite was used as a running standard for unknown chromite 156 grains. Typical count rates obtained on the Stillwater chromite standard were $\sim 5 \times 10^8$ cps for 157 16 O, $\sim 2 \times 10^5$ cps for 17 O, and $\sim 1 \times 10^6$ cps for 18 O. Typical errors (2 σ), accounting for 158 measurement errors and the external reproducibility of the standard, are estimated to be 159 ~0.7‰ for δ^{18} O, ~0.6‰ for δ^{17} O, and ~0.8‰ for Δ^{17} O (= δ^{17} O – 0.52 × δ^{18} O, representing 160 deviation from the terrestrial fractional line). All analytical spots were checked under SEM, 161 and any that fell on cracks or grain boundaries were excluded. 162

163

164 **4. Results**

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We examined ~3500 DSS micrometeorites and ~1500 Antarctic micrometeorites 166 to identify chromite-bearing micrometeorites for further understanding the chemical 167 and isotopic characterisitics of these particles. We found 46 chromites in 30 DSS 168 micrometeorites and 20 chromites in 12 Antarctic micrometeorites; the chemical 169 compositions and sizes of these chromites are reported in Table 1. Chromite grains varied 170 from round to euhedral (Figs. 1 and 2). Most of the chromite-bearing micrometeorites 171 displayed barred, cryptocrystalline, glass, or porphyritic type textures (Figs. 1 and 2). Only 172 two chromite grains were observed in partially melted scoriaceous particles, and their small 173

sizes made their isotopic characterization challenging. Some chromites may have been altered, as their Si, Mg, Al, and Fe contents were slightly modified due to heating during atmospheric entry, but nonetheless seem to largely retain their elemental compositions. Fig. 3 shows the **sizes** (maximum length) of chromite grains relative to **their major axes** of their host micrometeorites as well as the distribution of chromite grain sizes. Most chromites are **between ~5 and 100 µm** in their maximum dimension, though some altered chromites are smaller, between a few and <10 µm (Fig. 2).

181

182 We selected 18 micrometeorites for oxygen isotopic analyses of chromite and olivine grains. In nine micrometeorites larger than 20 µm, we analyzed the oxygen isotopic 183 compositions of olivines and 17 chromites (Table 2); in the other nine, chromites were too 184 185 small to analyze, so we measured the oxygen isotopic compositions of olivines only. The $\delta^{18}O$ and $\delta^{17}O$ values of chromite grains range from –0.8 to 6.0‰ and from 0.3 to 3.6‰, 186 respectively, corresponding to Δ^{17} O values of -0.9 to 1.6‰ (Figs. 4–6). Micrometeorite 187 olivines have the same limited range of $\Delta^{17}O$ values, except for a few that fall near 188 carbonaceous chondrite. The Δ^{17} O values reported for micrometeorite olivines and 189 chromites in Figs. 5 and 6 are averaged per micrometeorite, assuming that individual 190 191 micrometeorites document a single precursor.

192

193 5. Discussion

194

195 Chromites in micrometeorites provide powerful constraints on heating during 196 atmospheric entry and micrometeorite precursors. The mere presence of chromite is 197 insufficient to distinguish between the different types of **chondrites** or metamorphic grades 198 (Bunch et al. 1967; Johnson and Prinz 1991), though chromites can be used to validate 199 analogous micrometeorite precursors. For example, Prasad et al. (2015) used chemical composition as analytical tool for the chromites in micrometeorites to show that four of 200 seven micrometeorites were related to L3-6 chondrites, and the remaining three to a 201 202 combination of L/H-type chondrites. However, such a small number of chromite grains is not statistically robust enough to conclusively identify their sources. Here, we broaden the 203 statistics from the different sampling sites (DSS and Antarctic ice), and include 204 micrometeorites of a larger size range (50-500 µm) that arrived on Earth during the past 205 ~50,000 years (Taylor et al. 1998; Prasad et al. 2015). Our detailed investigation of the 206 oxygen isotopic compositions and physical and chemical properties of chromites will 207 improve our understanding of the parent bodies of chromite-bearing micrometeorites and 208 209 their contributions to the micrometeorite flux.

210

211 5.1 Origin of chromite-bearing micrometeorites

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Chromite size has not shown any great trend with respect to micrometeorite size, 213 however, it seems to be negatively correlated to chromite abundance; i.e., the chromite 214 grains observed herein range in size from 4 to 153 μ m, but are largely dominated by ~5–50 215 µm grains from micrometeorites ~50-500 µm in size (Fig. 3). Chromites are present in 216 carbonaceous chondrites, but are typically smaller than those in ordinary chondrites (Johnson 217 218 and Prinz, 1991). In L-type chondrites, chromite sizes vary according to the different metamorphic grades (L3, ~10-50 µm; L4, ~18-150 µm; L5, ~25-160 µm; and L6, ~30-600 219 µm; Bridges et al. 2007), and the sizes of the largest chromite grains observed increase with 220 increasing petrographic grade. Based on these size distributions, our results suggest that the 221 chromite-bearing micrometeorites reported herein could derive from ordinary chondrites. 222 However, chromite size alone is not diagnostic of chondrite type or metamorphic grade, and 223

these results must be coupled with the chemical and O isotopic compositions of chromites toconvincingly relate them to specific chondrite types.

226

227 Chromite is the most abundant spinel group mineral in ordinary chondrites (up to 0.6 wt%; Keil 1962) and the least abundant in carbonaceous chondrites (<0.01 wt%; Riebe 228 2009). In addition, chromites in ordinary chondrites have more varied chemical 229 compositions than those in carbonaceous chondrites, likely due to the increased thermal 230 metamorphism of ordinary chondrite parent bodies compared to carbonaceous ones. 231 232 The concentrations of **minor oxide** such as TiO₂, MnO, Cr₂O₃, V₂O₅, or ZnO are therefore powerful tools for ascertaining the chondritic provenance of chromites, although chemical 233 overlaps are observed. Chromites in H chondrites have very restricted TiO₂ contents ranging 234 235 from ~1.5 to 3.0 wt%, LL chondrites have higher TiO₂ contents of ~2.5-4.5 wt%, and L chondrites show the widest range, from 0.5 to 3.5 wt% TiO₂ (Fig. 7). In general, the average 236 TiO₂ content of equilibrated grains increases from H to L to LL chondrites. The least 237 metamorphosed L3 chondrites have chromites with <2 wt% TiO₂, similar to the much smaller 238 chromites in carbonaceous chondrites (< 2 wt% TiO₂; Fuchs et al. 1973; Johnson and Prinz, 239 1991). Our data largely matches the range of chromite TiO₂ contents observed in ordinary 240 chondrites, and to some extent that in carbonaceous chondrites. 241

242

Furthermore, chromites in ordinary **chondrites** are enriched in MnO (0.5–0.95 wt%) relative to those in carbonaceous chondrites (0.0–0.2 wt%; Bunch et al. 1967; Wlotzka 2005). Among ordinary chondrites, chromites in H chondrites are Mn-enriched (~0.9 wt%) relative to those in L (~0.7 wt%) and LL chondrites (~0.5 wt%; Bunch et al. 1967; Wlotzka 2005). Our results show that micrometeorite chromites are relatively rich in **MnO** (0.03–0.63 wt%, averaging 0.32 ± 0.16 wt% MnO; Table 1, Fig. 8), suggesting that micrometeorites could 249 have derived from ordinary with some from carbonaceous chondrites. Most of the data in Fig. 8 fall have shown a spread in data making difficult to assert the precursor. Apart 250 from this, the diffusion of the minor element that has taken place during entry makes 251 the matter worse to identify the precursor. This is supported by (i) V_2O_3 vs. Cr_2O_3 , MnO, 252 and TiO₂ plots indicate a larger fraction of micrometeorite chromite grains plot in the area of 253 ordinary chondrite chromites with relatively small fraction in the carbonaceous chondrite 254 range (Fig. 8), and (ii) micrometeorite chromite ZnO concentrations up to 0.3 wt%, beyond 255 the range of the systematically Zn-poor carbonaceous chondrite chromites (<0.1 wt% ZnO; 256 257 Fuchs et al. 1973; Brearley and Jones 1998).

258

The oxygen isotopic compositions of micrometeorite chromites allow determination 259 260 of the contribution of chromite-bearing extraterrestrial material to the Earth's surface. The ordinary and enstatite chondrite have Δ^{17} O lying between 0.1 and 1.5‰, while the 261 carbonaceous chondrite have wider range with Δ^{17} O <0%, for e.g. CV chondrite having -5 to -262 1‰, CO and CK having -5 to -4‰, CR having -2 to -1‰, CM having -4 to -1‰, respectively 263 (Clayton et al., 1991, 1999) Our results show that micrometeorite chromites have ¹⁶O-poor 264 isotopic compositions similar to those measured in ordinary chondrites (Table 2; Clayton et 265 al. 1991; Greenwood et al. 2007). One of our micrometeorites (SP005-P219) has a chromite 266 grain with a slightly ¹⁶O-rich composition (Δ^{17} O values ranging from -0.3 to -0.9‰), and 267 thus probably derives from a carbonaceous chondrite (Table 2). Excluding this 268 micrometeorite, all other chromite grains observed seems to originate predominately 269 270 with oxygen isotopic compositions very similar to those of chromites in ordinary chondrites. It may be difficult based on the oxygen isotope studies of chromites to narrow on 271 specific population of ordinary chondrite. Nevertheless, our results indicate that chromites 272

are dominantly related to ordinary chondrites, and that the contribution from carbonaceouschondrites is small (Figs. 4–6).

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276 The O isotopic compositions of olivines in micrometeorites with chromite grains too small for ion microprobe analyses indicate that most of the micrometeorites have 277 ordinary chondrite compositions, while about six have shown $\Delta^{17}O < 0$ (Fig. 6). The 278 negative Δ^{17} O values for these chromite-bearing micrometeorites point toward 279 mainstream contribution from ordinary chondrite with some from carbonaceous 280 281 chondrites. Many of the micrometeorites have undergone alteration during atmospheric entry, as attested by their high $\delta^{17,18}$ O values that result from exchange with the 282 stratospheric oxygen isotope composition of $\delta^{18}O = 23.5\%$, and $\delta^{17}O = 11.8\%$ (Thiemens et al., 283 1995; Engrand et al. 2005; Yada et al. 2005; Suavet et al. 2010; Cordier et al. 2011; 284 Rudraswami et al. 2015a, 2016a). However, their Δ^{17} O values are similar to those reported 285 for chromite grains in ordinary chondrites, in contrast to many micrometeorites analyzed 286 287 previously that were dominated by carbonaceous chondrite compositions (Clayton et al. 1991; Engrand et al. 1999; Greenwood et al. 2007; Rudraswami et al. 2015a, 2016a). 288 Excluding the **few** dataset that has shown us Δ^{17} O in carbonaceous chondrite range, these 289 results support the conclusion that chromite grains have O-isotopic compositions similar to 290 291 ordinary chondrites. Though oxygen isotopic exchange between chromite-bearing micrometeorites and the atmosphere during entry resulted in $\delta^{17,18}$ O values shifted towards 292 atmospheric values in many olivine grains, the O isotopic compositions of chromite grains 293 indicate that they are resistive and retain their precursor, i.e., ordinary chondritic, 294 295 composition (Figs. 4-6).

296

297 5.2 Implications for asteroid belt dynamics

The chemical and mineralogical compositions of interplanetary dust particles and 299 micrometeorites show strong similarities to those of carbonaceous chondrites, unlike 300 301 meteorites that are dominated by ordinary chondrites (Brownlee et al. 1997; Rietmeijer 1998; Schmitz and Tassinari 2001). This discrepancy may be due to the dominance of ordinary 302 chondrite bodies in the inner asteroid belt, where slight planetary perturbations can send 303 larger asteroidal bodies into Earth resonant orbits, eventually reaching Earth as ordinary 304 chondrite meteorites (Meibom and Clark 1999). This is not the case with micrometeorites; 305 306 small dust particles generated by collisions can arrive in Earth resonant orbits from any region of the asteroid belt. It thus appears that the diversity of planetary bodies present in the 307 asteroid belt is better represented in the micrometeorite flux, which exceeds that of meteorites 308 309 and is therefore more statistically significant.

310

Although our study clearly indicates a genetic link between ordinary chondrites and 311 **many** chromite-bearing micrometeorites, the latter constitute only ~1% of micrometeorites 312 collected from DSS and Antarctic ice. Antarctic micrometeorites have terrestrial ages of 313 ~1000 a (Taylor et al. 1998), whereas micrometeorites in **DSS** are as old as ~50,000 a (Prasad 314 et al. 2013). In these two time spans (Taylor et al., 1998; Prasad et al., 2013), the flux of 315 316 chromite-bearing micrometeorites has remained almost constant and no major collision event 317 has been reported in the recent past. The abundance of micrometeorites related to ordinary chondrites is probably underestimated due to the non-systematic presence of chromite grains 318 in micrometeorites derived from ordinary chondrites. Nevertheless, it seems that ordinary 319 320 chondrites are rare in the asteroid belt, which is rather dominated by carbonaceous bodies as reflected in the chemical and isotopic compositions of micrometeorites (Brownlee et al. 1997; 321 Meibon and Clark 1999; Taylor et al. 2007; Genge et al. 2008; Suavet et al. 2010; 322

Rudraswami et al. 2012, 2014, 2015b, 2016b). This could be due to either (i) a population deprived of S-type asteroids in the asteroid belt or (ii) an under-representation of dust related to ordinary chondrites in Earth resonant orbits.

326

The predominance of micrometeorites derived from carbonaceous chondrites could 327 also result from the large differences in the physical properties of carbonaceous and ordinary 328 chondrites, such as tensile strength, density, and porosity. Tensile strength is a critical 329 parameter controlling the dust sizes generated during collisions; the greater the tensile 330 strength, the larger the collisional debris (Flynn et al. 1999; Consolmagno et al. 2008). 331 Furthermore, the lower porosity of ordinary compared to carbonaceous chondrites would 332 generate larger particles during asteroid breakup. Disruptive impact experiments performed 333 334 on different chondrite types revealed that carbonaceous chondrites produce more fragments in the micrometeorite mass range than ordinary chondrites (Flynn et al. 2009). These results 335 suggest that the physical properties of carbonaceous chondrites (i.e., high porosity, low 336 337 density and tensile strength) induce fragmentation into dust-sized particles during asteroidal collisions (Consolmagno et al. 2008). This is consistent with the large contribution of 338 micrometeorites from carbonaceous chondrites, whereas there is a disparity in the ordinary 339 chondrite contribution that varies from a few percent for smaller sizes ($<300 \mu m$) to $\sim30\%$ for 340 larger sizes (>600 µm) (e.g., Suavet et al. 2010; Rudraswami et al. 2015b, 2016b). The 341 porosity (10–17%) and density (3.0–3.2 g/cm³) of the higher petrographic grade L chondrites 342 (L5 and L6) are similar to those of carbonaceous chondrites such as CO3 and CV3, and the 343 increased porosity (up to 23%) and decreased density (up to 2.6 g/cm³) at lower petrographic 344 grades are being listed as similar to those reported for many primitive carbonaceous 345 chondrites (Flynn et al. 1999). H and LL chondrites, however, have porosities much lower 346 than those of L5–6 or H5–6 chondrites, even at lower petrographic (e.g., LL3) grades (Flynn 347

et al. 1999). Overall, the high porosity and low density of L-type chondrites compared to
other ordinary chondrites favor fragmentation into smaller dust particles during collisions,
although our present dataset does not allow us to conclusively relate micrometeorites to a
petrographic grade or type of ordinary chondrite.

352

353 6. Conclusions

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In-situ oxygen isotopic analyses of chromite-bearing micrometeorites revealed that 355 356 chromites in micrometeorites are indistinguishable from those in ordinary chondrites. The Δ^{17} O values of most chromite-bearing micrometeorites here and in the literature are similar to 357 those of ordinary chondrites, indicating that chromite-bearing micrometeorites are largely 358 359 derived from ordinary chondrites (~70%) with small contribution (~30%) from carbonaceous chondrites. Chromite chemical compositions support an ordinary chondrite 360 provenance despite the chemical compositions of some chromites being altered during 361 atmospheric entry, as reflected by compositions enriched in Si, Mg, Al, and Fe. The two time 362 spans analyzed herein (the last ~1000 and ~50,000 years in **DSS** and Antarctic ice, resplively) 363 suggest that the contribution of chromite-bearing micrometeorites related to ordinary 364 chondrites in the size range ~50-500 µm has remained consistent at ~1% of the total 365 micrometeorite flux during the last 50,000 years. The low overall abundance of 366 367 micrometeorites related to ordinary chondrites likely indicates a small contribution of ordinary chondrites to dust particles produced in the asteroid belt. 368

369

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529	FIGURE CAPTIONS
530	
531	Figure 1.
532	Representative back-scattered electron images of chromite-bearing micrometeorites collected
533	from Indian Ocean DSS and the South Pole water well (SPWW). Chromites are circled in
534	each image. The chemical compositions of chromites unaffected by atmospheric entry are
535	given in Table 1.
536	
537	Figure 2.
538	Enlarged back-scattered electron images of chromites in micrometeorites.
539	
540	Figure 3.
541	(a) Chromite sizes as a function of their host relict micrometeorites. (b) The size distribution
542	of chromites from micrometeorites analyzed in this study.
543	
544	Figure 4.
545	The oxygen isotopic compositions of olivines (representing micrometeorites) and chromites
546	in chromite-bearing micrometeorites. Data points represent individual analyses. Most of
547	them are depleted in ¹⁶ O and have Δ^{17} O values similar to those of ordinary chondrites.
548	The TF and Carbonaceous Chondrite Anhydrous Mineral (CCAM) lines are shown for
549	reference. The error bars of many of the data points are masked by the size of the data
550	points themselves.
551	

Figure 5.

553 Δ^{17} O versus δ^{18} O values of chromites in chromite-bearing micrometeorites. Chromite 554 compositions are averaged per micrometeorite depending on the number of oxygen 555 isotope analyses which varies from two to three chromite per micrometeorite, however, 556 there are few that only represent single analyses. Most of the data fall in the ordinary 557 chondrite range, as delimited by the range of Δ^{17} O values of LL, L, and H chondrites (green, 558 blue, and red lines, respectively).

559

560 Figure 6

561 The distribution of average Δ^{17} O values of 18 chromite-bearing micrometeorites (gray). 562 Chromites were large enough to analyze in nine of those micrometeorites (black), and the 563 values shown are averaged per micrometeorite.

564

565 **Figure 7.**

The distribution of TiO₂ contents in chromites from (a) micrometeorites studied herein and (b) different chondrite types (carbonaceous, L, LL, H). Data for chromites from ordinary chondrites are from Bunch et al. (1967), Snetsinger et al. (1967), Johnson and Prinz (1991), and Wlotzka (2005). Data for chromites from carbonaceous chondrites are from Fuchs et al. (1973) and Johnson and Prinz (1991).

571

572 **Figure 8.**

Elemental distributions of chromites from micrometeorites studied herein (red), carbonaceous
chondrites (yellow) and ordinary (L, LL, and H) chondrites (gray diamonds, inverted
triangles, and circles, respectively). Data for chromites from ordinary chondrites are from
Bunch et al. (1967), Snetsinger et al. (1967), Johnson and Prinz (1991), and Wlotzka (2005).
Data for chromites from carbonaceous chondrites are from Fuchs et al. (1973) and Johnson

578	and Prinz (1991). The rectangle and oval outline represent the spread in data of chromite
579	from ordinary and carbonaceous chondrites, respectively. In figs. a, c, d, f, the H
580	chondrite has shown narrow spread which can be recognized and is marked with
581	smaller rectangle with ordinary chondrite.
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592 Fig. 1





595 Fig. 2



598 Fig. 3









603 Fig. 5



606 Fig.

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609 Fig.7



- 613 Fig. 8

Sr. No	Cosmic spherule#	Туре	Spherule size (μm) [#]	Chromite size(µm)*	MgO	Al ₂ O ₃	SiO ₂	CaO	TiO ₂	Cr_2O_3	MnO	FeO	V_2O_3	ZnO	Total
1	AAS-38-143-P141A	Barred	150	24	11.31	1.78	0.99	0.06	2.53	57.48	0.37	22.23	0.66	0.22	97.63
2	AAS-38-167-1-P20	Barred	280	37	6.50	5.62	0.10	0.00	3.46	61.45	0.49	22.79	0.69	0.00	101.09
				22	11.62	5.67	0.09	0.01	3.30	61.43	0.33	17.54	0.71	0.00	100.70
3	AAS-38-167-1-P59	Porphyritic	210	16	6.71	10.12	0.70	0.04	0.37	54.63	0.24	25.78	0.41	0.00	99.00
4	AAS-38-167-P53	Barred	380	81	2.51	6.16	0.05	0.00	2.73	57.55	0.53	28.51	0.85	0.24	99.14
5	AAS-38-169-P60	Barred	400	52	6.91	9.70	0.29	0.04	0.35	56.85	0.24	24.82	0.43	0.00	99.63
6	AAS-38-170-P89	Porphyritic	188	8.1	8.81	20.20	1.30	0.18	1.01	45.10	0.29	21.83	0.41	0.00	99.13
7	AAS-38-177-P51	Porphyritic	228	4	8.86	23.58	0.83	0.01	0.86	36.29	0.21	28.65	0.63	0.06	99.98
8	AAS-38-180-P73	Barred	320	24	3.29	6.55	0.03	0.01	2.01	56.04	0.58	28.55	0.61	0.23	97.90
9	AAS-38-180-P107	Barred	650	23	12.30	5.69	0.18	0.01	3.14	54.61	0.28	20.07	0.81	0.15	97.24
10	AAS-38-182-P25	Porphyritic	228	14	8.69	9.96	0.15	0.00	1.01	60.39	0.56	18.58	0.56	0.01	99.89
				7	10.68	4.47	1.26	0.07	2.26	54.47	0.43	23.07	0.51	0.00	97.22
11	AAS-38-184-P28	Barred	288	153	9.36	5.46	0.07	0.03	2.69	60.67	0.41	21.78	0.67	0.19	101.33
				48	12.36	4.61	0.59	0.02	2.21	60.58	0.39	19.99	0.51	0.10	101.32
12	AAS-38-184-P9	Barred	320	6	8.69	8.45	0.53	0.00	1.26	55.14	0.17	23.42	1.03	0.00	98.69
				7	9.35	6.02	0.79	0.08	1.36	58.56	0.54	21.78	0.63	0.00	99.10
				7	10.53	5.60	0.63	0.06	2.18	57.86	0.05	20.02	0.76	0.19	97.88
				5	11.86	7.31	0.67	0.10	1.62	55.28	0.17	20.12	0.79	0.33	98.23
13	AAS-38-188-P54	Barred	375	69	6.44	5.72	0.03	0.01	3.21	54.21	0.35	29.28	0.55	0.09	99.90
14	AAS-38-193-P40	Porphyritic	250	10	16.53	6.48	0.33	0.00	2.46	61.82	0.00	11.18	0.81	0.30	99.91
				15	16.70	6.57	0.69	0.02	1.86	60.20	0.24	10.67	0.73	0.24	97.92
				5	17.68	6.97	0.46	0.01	2.12	58.97	0.10	10.67	0.72	0.20	97.91
				5	16.83	6.58	0.65	0.09	1.52	62.10	0.03	10.80	0.93	0.36	99.90

Table 1: Chemical composition (wt%) of the chromite grains from micrometeorites.

15	AAS-38-193-P43	Porphyritic	214	13	10.13	0.00	0.00	0.03	3.30	59.95	0.19	23.75	0.71	0.12	98.18
16	AAS-38-193-P45		242	6	15.91	0.10	0.40	0.01	0.05	72.81	2.97	6.46	0.13	0.01	98.84
17	AAS-38-195-P80A	Barred	257	23	7.94	5.14	1.17	0.01	0.71	52.26	0.35	28.93	0.00	0.00	96.50
18	AAS-38-203-P58	Barred	484	93	5.86	5.43	0.07	0.01	3.03	61.56	0.56	23.44	0.68	0.17	100.81
				66	4.99	4.97	0.86	0.04	2.44	59.34	0.55	25.44	0.64	0.18	99.46
				47	7.23	4.91	0.16	0.00	2.82	59.20	0.58	23.39	0.66	0.13	99.07
19	AAS-38-203-P88	Barred	205	33	10.14	4.86	0.55	0.02	2.89	59.40	0.41	21.41	0.64	0.09	100.41
				35	5.42	5.37	0.94	0.03	2.38	57.29	0.63	25.37	0.71	0.27	98.41
				11	7.15	4.99	1.08	0.04	2.14	58.65	0.43	24.77	0.69	0.19	100.13
20	AAS-38-204-P58	Cryptocrystalline	189	21	13.22	5.65	0.36	0.01	2.91	59.61	0.11	17.74	0.78	0.11	100.49
21	AAS-38-207-P15	Porphyritic	250	87	4.83	6.18	0.13	0.03	2.61	61.04	0.43	24.77	0.79	0.21	101.02
22	AAS-38-207-P117	Porphyritic	250	100	7.63	6.74	0.19	0.01	2.32	56.65	0.34	24.99	0.87	0.24	99.99
23	AAS-62-5-P77	Barred	190	15	6.54	5.47	0.23	0.04	3.31	56.11	0.33	23.67	0.95	0.29	96.95
24	AAS-62-10-P35	Barred	275	60	8.53	5.86	0.35	0.03	2.01	58.11	0.52	21.17	0.86	0.18	97.62
25	AAS-62-26-P86	Porphyritic	184	19	16.42	5.31	0.90	0.01	1.68	58.86	0.10	14.73	0.79	0.30	99.10
				22	8.09	2.92	0.44	0.16	1.38	64.87	0.51	19.71	0.73	0.13	98.93
26	AAS-62-26-P132	Porphyritic	220	83	7.21	5.40	0.15	0.01	2.94	57.42	0.32	23.40	1.06	0.32	98.24
				54	8.62	7.47	0.15	0.04	2.94	53.85	0.41	24.77	0.73	0.16	99.13
				40	8.50	6.61	0.21	0.00	2.97	54.40	0.34	22.98	0.84	0.20	97.06
27	AAS-62-32-P102	Porphyritic	173	16	2.76	8.75	0.79	0.02	1.56	46.66	0.31	37.66	1.00	0.00	99.49
28	AAS-62-32-P106	Scoriaceous	232	15	5.45	11.46	0.60	0.05	0.72	47.83	0.22	32.69	0.00	0.00	99.02
29	AAS-62-51-P4	Porphyritic	211	19	11.22	19.74	0.27	0.03	1.42	33.06	0.24	30.64	0.22	0.00	96.84
30	AAS-62-51-P52	Porphyritic	275	9	6.74	10.25	1.46	0.01	0.82	43.94	0.28	31.97	0.62	0.00	96.09
31	SP009-P126	Cryptocrystalline	112	8	7.49	7.41	0.22	0.11	2.14	55.04	0.29	24.83	0.64	0.02	98.20
				12	7.13	4.22	0.93	0.06	1.91	53.25	0.41	26.85	0.59	0.04	95.39
				20	8.47	5.81	1.98	0.09	1.24	51.81	0.38	24.69	0.64	0.00	95.11
				6	9.30	6.45	1.37	0.12	1.25	47.25	0.27	27.83	0.57	0.01	94.42
				18	9.00	9.98	0.51	0.11	1.64	54.21	0.11	23.06	0.70	0.00	99.31

32	SP005-P135	Porphyritic	119	11	8.58	17.27	0.58	0.00	0.92	45.49	0.08	25.21	0.64	0.05	98.82
				13	7.95	19.34	0.56	0.00	1.04	42.28	0.18	25.40	0.63	0.00	97.38
				8	7.82	20.47	0.41	0.00	1.08	41.41	0.18	25.97	0.64	0.00	97.97
33	SP005-P219	Glass	89	12	8.39	8.57	1.46	0.54	1.28	50.55	0.61	27.87	0.67	0.05	99.99
				10	6.75	9.45	1.10	1.00	1.62	47.36	0.66	27.73	0.45	0.00	96.13
				7	6.51	9.02	1.41	0.36	1.02	50.25	0.57	29.06	0.47	0.02	98.69
34	SP005-P325	Porphyritic	59	7	10.03	10.03	0.75	0.07	1.54	55.26	0.49	18.77	0.48	0.00	97.42
35	SP005-P530	Glass	72	7	13.05	4.97	0.10	0.03	2.83	60.86	0.36	13.56	0.61	0.00	96.37
36	SP005-P658	Porphyritic	100	17	4.29	14.51	0.34	0.04	1.00	48.20	0.26	28.87	0.79	0.00	98.29
37	SP005-P711	Porphyritic	97	25	7.86	12.93	0.22	0.13	0.44	51.21	0.26	24.42	0.50	0.00	97.97
38	SP005-P769	Porphyritic	124	31	6.39	16.05	0.59	0.00	0.58	46.21	0.09	27.31	0.61	0.07	97.91
39	SP005-P791	Porphyritic	95	34	4.61	3.31	0.21	0.02	0.97	62.48	0.48	26.38	0.61	0.00	99.07
40	SP007-P72	Relict bearing	132	7	6.80	12.14	0.84	0.13	1.59	46.40	0.12	30.45	0.90	0.01	99.36
41	SP007-P2	Unmelted	153	14	2.81	6.01	0.28	0.05	2.03	53.25	0.10	33.88	1.00	0.30	99.71
42	SP007-P29	Relict bearing	74	6	9.55	13.69	0.29	0.05	1.21	49.56	0.31	24.34	0.28	0.01	99.27

indicate spherule size as diameter

* indicate chromite size measured along the maximum length.

All measurements are in wt%

#	Name	Туре	Spot#	δ^{18} O	2σ	δ^{17} O	2σ	Δ^{17} O	2σ
1	SP005-P126	Glass	1	5.8	0.6	1.3	0.5	-1.7	0.6
			2	8.8	0.7	2.7	0.5	-1.8	0.6
			3	10.2	0.6	3.8	0.5	-1.5	0.6
			4	9.9	0.7	4.3	0.4	-0.9	0.5
			5	11.2	0.8	5.2	0.4	-0.7	0.5
			6	11.5	0.5	5.4	0.5	-0.5	0.5
2	SP005-P219	Glass	*1	4.7	0.9	1.5	0.4	-0.9	0.6
			*2	3.9	0.7	1.7	0.4	-0.3	0.6
			*3	3.7	0.6	1.2	0.4	-0.7	0.5
			4	3.7	0.8	1.3	0.5	-0.6	0.6
			5	6.4	0.9	3.1	0.3	-0.2	0.6
3	AAS-38-203-P58	Barred	*1	1.4	0.7	1.3	0.4	0.6	0.5
			*2	0.9	0.5	0.8	0.5	0.3	0.5
			*3	0.7	0.7	0.6	0.4	0.3	0.6

5

*1

*2

*3

4

5

6

*1

*2

4 AAS-38-203-P88

5 AAS-38-207-P15

Barred

Barred

12.4

12.4

6.0

1.4

5.8

7.9

8.2

9.4

2.7

-0.1

0.7

0.7

0.7

0.6

0.6

0.8

0.7

0.7

0.4

0.4

6.5

6.5

3.6

1.7

3.6

4.4

5.5

5.2

2.7

1.3

0.5

0.5

0.5

0.4

0.4

0.4

0.5

0.5

0.6

0.6

0.0

0.1

0.4

1.0

0.6

0.3

1.2

0.4

1.3

1.4

0.6

0.6

0.6

0.5

0.5

0.6

0.6

0.6

0.6

0.6

Table 2. Oxygen isotopic composition of chromites and the olivine phases in micrometeorites. All the values of the oxygen isotopic composition are reported in ‰.

			3	7.6	0.7	4.5	0.5	0.6	0.6
			4	9.9	0.6	5.4	0.5	0.3	0.6
6	AAS-38-169-P60	Barred	*1	-0.6	0.9	1.3	0.4	1.6	0.6
			2	12.7	0.8	7.4	0.5	0.8	0.7
			3	12.2	0.6	7.9	0.5	1.5	0.6
7	AAS-38-188-P54	Barred	*1	0.3	0.6	1.2	0.5	1.1	0.6
			2	13.5	0.8	7.7	0.5	0.7	0.6
			3	11.8	0.8	7.5	0.5	1.4	0.6
8	AAS-38-167-1-P20	Barred	*1	0.3	0.5	1.3	0.6	1.1	0.6
			2	14.5	0.8	8.6	0.5	1.1	0.6
			3	14.9	0.8	8.4	0.5	0.7	0.7
9	AAS-38-167-P53	Barred	*1	-0.6	0.6	0.7	0.4	1.0	0.5
			*2	-0.2	0.6	0.9	0.5	1.0	0.6
			3	14.7	0.7	8.1	0.5	0.4	0.6
			4	14.2	0.6	8.1	0.5	0.8	0.6
10	AAS-38-204-P58	Barred	*1	-0.8	0.8	0.3	0.5	0.7	0.6
			2	-1.9	0.8	-1.4	0.5	-0.4	0.7
			3	-4.1	1.1	-1.3	0.6	0.8	0.8
11	SP005-P135	Porphyritic	1	26.5	0.8	11.5	0.4	-2.3	0.6
			2	26.4	0.7	11.6	0.4	-2.1	0.5
			3	30.7	0.6	13.6	0.4	-2.3	0.5
			4	28.7	0.7	13.4	0.4	-1.6	0.6
			5	29.8	0.6	13.8	0.4	-1.7	0.5
12	SP005-P658	Porphyritic	1	22.3	0.7	8.9	0.5	-2.7	0.6
			2	27.2	1.2	11.4	0.7	-2.7	0.9
13	SP005-P711	Glass	1	26.7	0.8	11.2	0.4	-2.7	0.6
			2	26.7	0.7	11.2	0.4	-2.6	0.5
14	SP005-P769	Porphyritic	1	24.7	0.7	10.9	0.5	-1.9	0.6
15	SP005-P770	Unmelted	1	3.0	0.6	2.8	0.3	1.2	0.4

			2	2.9	0.5	2.8	0.4	1.3	0.5
16	SP007-P72	Relict bearing	1	-3.6	1.0	-1.0	0.5	0.9	0.7
			2	3.1	0.9	1.6	0.5	0.0	0.7
			3	9.2	1.0	5.0	0.6	0.2	0.8
17	SP007-P2	Unmelted	1	4.0	0.7	4.1	0.5	2.0	0.6
			2	4.4	0.9	3.9	0.5	1.6	0.7
			3	-2.4	1.0	2.2	0.6	3.5	0.8
			4	2.6	0.9	4.3	0.3	3.0	0.6
18	SP007-P29	Relict bearing	1	3.3	0.6	2.5	0.5	0.8	0.6
			2	4.1	0.7	2.4	0.5	0.2	0.6
			3	2.9	0.9	3.1	0.6	1.6	0.7

* marked are the oxygen isotope analyzed for chromite grain.