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## ARTICLE INFO

## ABSTRACT

Oxygen isotopic compositions allow identification of potential parent bodies of extraterrestrial materials. We measured oxygen isotope ratios of 33 large (diameter >500 μm) silicate melted micrometeorites (cosmic spherules) from Antarctica, using IR-laser fluorination coupled with mass spectrometry. It is the first time that this high-precision method is used on individual micrometeorites. The selected micrometeorites are representative of the influx of extraterrestrial materials to the Earth. Our results show that most micrometeorites are related to carbonaceous chondrites, which is consistent with previous studies. However, 20–50% of them seem to be related to CO/CV carbonaceous chondrites, whereas CM/CR carbonaceous chondrites were thought to be the main source for micrometeorites. Furthermore, ~30% of measured samples have oxygen isotope ratios lying above the terrestrial fractionation line, which relates them to ordinary chondrites or other, as yet, unsampled parent bodies.

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## 1. Introduction

Micrometeorites are extraterrestrial particles smaller than about 2 mm collected on the Earth's surface. Based on petrographic and chemical investigation of small unmelted micrometeorites (diameter ~150–250 μm), previous studies (Engrand and Maurette, 1998) have concluded that 99% of them come from carbonaceous, possibly cometary parent bodies (CM/CR related), although evidence for ordinary chondrite-related micrometeorites was recently put forward (Genge, 2008; Rochette et al., 2008). This argument has generated the belief that micrometeorites sample different objects from those sampled by macroscopic meteorites, which has been explained invoking friable parent bodies that would produce small particles upon collision rather than macroscopic fragments of rock. Subsequent ion probe oxygen isotopic analyses, mostly on melted micrometeorites (cosmic spherules: CSs) (Engrand et al., 2005; Yada et al., 2005, Taylor et al., 2005), have supported this argument as most samples had oxygen isotope ratios below the terrestrial fractionation line (TFL), except for a few samples that had ratios on the TFL within

analytical uncertainties. The typical analytical uncertainties of these ion probe analyses ($\pm 1$–2.75‰ on $\delta^{18}O$, $\pm 0.7$–1.7‰ on $\delta^{17}O$, and $\pm 0.6$–1.7‰ on $\Delta^{17}O$ for individual analyses (Engrand et al., 2005; Yada et al., 2005), including the statistical uncertainty associated with the measurement of the sample as well as the uncertainty associated with the instrumental fractionation correction derived from repeated analyses of the standard mineral), make it difficult to resolve signatures indicating a potential ordinary chondrite parent body ($\Delta^{17}O \approx 0.5$–1.5‰) from terrestrial values. Consequently, the near-TFL values were interpreted as terrestrial contamination of carbonaceous chondrite material. IR-laser fluorination coupled with mass spectrometry (IRMS) has a much better precision, by one to two orders of magnitude: the reproducibility on quartz standards at CEREGE (Aix-en-Provence, France) is $\pm 0.15$‰ for $\delta^{18}O$ ($n = 76$) (Alexandre et al., 2006), $\pm 0.11$‰ and $\pm 0.034$‰ for $\delta^{17}O$ and $\Delta^{17}O$, respectively ($n = 37$); and at the Open University (Milton Keynes, United Kingdom) the reproducibility is $\pm 0.08$‰ and $\pm 0.04$‰ for $\delta^{18}O$ and $\delta^{17}O$, respectively (measured on obsidian standard, $n = 31$), and $\pm 0.024$‰ for $\Delta^{17}O$ (measured on terrestrial whole rocks and mineral separates, $n = 47$) (Miller et al., 1999). Measurements of $\delta^{18}O$ are possible on masses as small as 0.3 mg at CEREGE (Crespin et al., 2008), and 0.2 mg at Open University (Greenwood et al., 2007), although such capability still limits analyses to the largest micrometeorites. The discovery of micrometeorites in the Transantarctic Mountains

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(Rochette et al., 2008) − in traps that collected micrometeorites by direct infall for hundreds of thousands of years (Folco et al., 2008, 2009) − made available hundreds of CSs with a sufficient mass to allow IRMS. Furthermore, this essentially unbiased collection (Suavet et al. 2009a) is representative of the influx of extraterrestrial materials to the Earth. In this study, we performed oxygen isotope measurements of 33 large (diameter >500 µm) silicate ("stony") CSs from the Transantarctic Mountains collection using IRMS at CEREGE and Open University.

## 2. Materials and methods

### 2.1. Samples

Thirty-three silicate cosmic spherules (CSs) from the Transantarctic Mountains collection were selected for this study. Their masses range from 262 µg to 877 µg after the weathered rim was leached out using diluted HCl for most of the samples. Backscattered electron images of the samples were taken at CEREGE (Aix-en-Provence, France) with a scanning electron microscope (SEM, Hitachi S-3000N) using a 30 kV accelerating voltage. SEM images (Fig. 1) allowed one to distinguish between barred olivine (BO, 23 CSs), porphyritic olivine (PO, 3 CSs) and glassy (7 CSs) textures (Genge et al., 2008). The distribution by types of the studied samples is similar to that of large collections (e.g. Taylor et al., 2000), assuming that cryptocrystalline and BO classes are merged.

### 2.2. Magnetic measurements

A magnetic characterization of the selected CSs was performed prior to the oxygen isotope measurements. Hysteresis parameters − saturation magnetization $M_s$ and saturation remanent magnetization $M_{rs}$ − were measured at CEREGE with a Princeton Measurements Corporation Vibrating Sample Magnetometer (VSM) with a peak field of 0.5 T (noise level of $\sim 10^{-9}$ A m$^2$). The remanent coercive field $B_{cr}$ was determined by DC backfield demagnetization of the saturation remanent magnetization using the VSM.

### 2.3. Oxygen isotope measurements

Measurements of $\delta^{18}O$ and $\delta^{17}O$ were carried out on 28 spherules at the Stable Isotopes Laboratory of CEREGE. Molecular oxygen was extracted using the IRMS technique (Alexandre et al., 2006; Crespin et al., 2008) and the three oxygen isotopic composition was measured with a dual-inlet mass spectrometer DeltaPlus, Thermo-Finnigan. The gas ($O_2$) was passed through a $-114$ °C slush to refreeze potential interfering gases before being sent to the mass spectrometer. In order to get sufficient 34/32 and 33/32 signals (2–3 V), the oxygen from <0.3 mg standards and all CSs samples was concentrated in the mass spectrometer in an auto-cooled 800 µl microvolume filled with silica gel and directly connected to the dual-inlet system. $\delta^{18}O$ and $\delta^{17}O$ values of the reference gas were fixed through the analysis of NBS28 ($\delta^{18}O = 9.60$‰ (Gröning, 2004), $\delta^{17}O = 4.99$‰ ). The oxygen isotope results are expressed in ‰ vs. V-SMOW. Measured $\delta^{18}O$ and $\delta^{17}O$ values of the samples were corrected on a daily basis using a 1.5 mg quartz laboratory standard "Boulangé" ($\delta^{18}O = 16.36$‰ (Alexandre et al., 2006), $\delta^{17}O = 8.507$‰, and $\Delta^{17}O = 0.006$‰). During the analyzing period, replicate analyses of 1.5 mg NBS28 ($\delta^{18}O = 9.59 \pm 0.26$‰, $\delta^{17}O = 4.98 \pm 0.16$‰, $\Delta^{17}O = -0.004 \pm 0.055$‰, $n = 11$), San Carlos olivine ($\delta^{18}O = 5.39 \pm 0.40$‰, $\delta^{17}O = 2.78 \pm 0.20$‰, $\Delta^{17}O = -0.026 \pm 0.032$‰, $n = 22$) and UWG-2 (Valley et al., 1995) ($\delta^{18}O = 5.69 \pm 0.18$‰, $\delta^{17}O = 2.96 \pm 0.11$‰, $\Delta^{17}O = -0.004 \pm 0.055$‰, $n = 8$) were made.

The relatively large scatter observed for $\delta^{18}O$ and $\delta^{17}O$ for SC olivine compared to NBS28 and UWG-2 is probably attributable to the heterogeneity of our San Carlos "standard" prepared from a number of millimetric crystals with possibly different oxygen isotope composi-
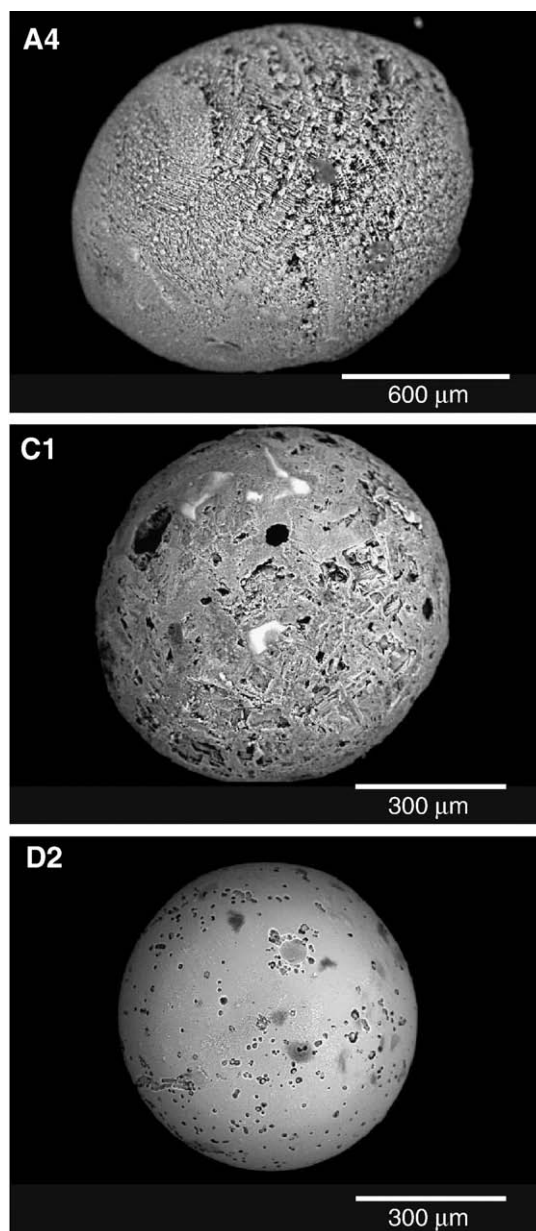


**Fig. 1.** Backscattered electron images of cosmic spherules A4 (barred olivine), C1 (porphyritic olivine) and D2 (glassy).

tion, as suggested by the scatter observed for the different San Carlos olivine standards used in Mattey and Macpherson (1993), Eiler et al. (1995), Rumble et al. (1997), Jabeen et al. (1998), Franchi et al. (1999), and Kusakabe et al. (2004).

Measurements made on microsamples (0.2–0.7 mg) of San Carlos olivine using the mass spectrometer-cooled microvolume (Fig. 2) show a systematic offset from the larger samples values for $\delta^{18}O$, $\delta^{17}O$ and $\Delta^{17}O$ ($-0.27 \pm 0.14$‰ for $\delta^{18}O$, $-0.33 \pm 0.09$‰ for $\delta^{17}O$, and $-0.189 \pm 0.052$‰ for $\Delta^{17}O$, $n = 9$). A similar offset was also observed when the microvolume was used to analyze quartz standard "Boulangé" >0.25 mg samples ($-0.1 \pm 0.1$‰ for $\Delta^{17}O$, $n = 31$), and <0.25 mg samples ($-0.16 \pm 0.05$‰ for $\Delta^{17}O$, $n = 19$). While the origin of this offset is unknown, it does not appear to be sample specific and therefore a small correction based on the systematic offsets for San Carlos olivine was made for CS samples measured at CEREGE: $+0.27$‰ for $\delta^{18}O$, $+0.33$‰ for $\delta^{17}O$, and $+0.189$‰ for $\Delta^{17}O$. The uncertainty on the value of the offset is included in the analytical uncertainties. Five more BO and PO spherules were measured at the Open University
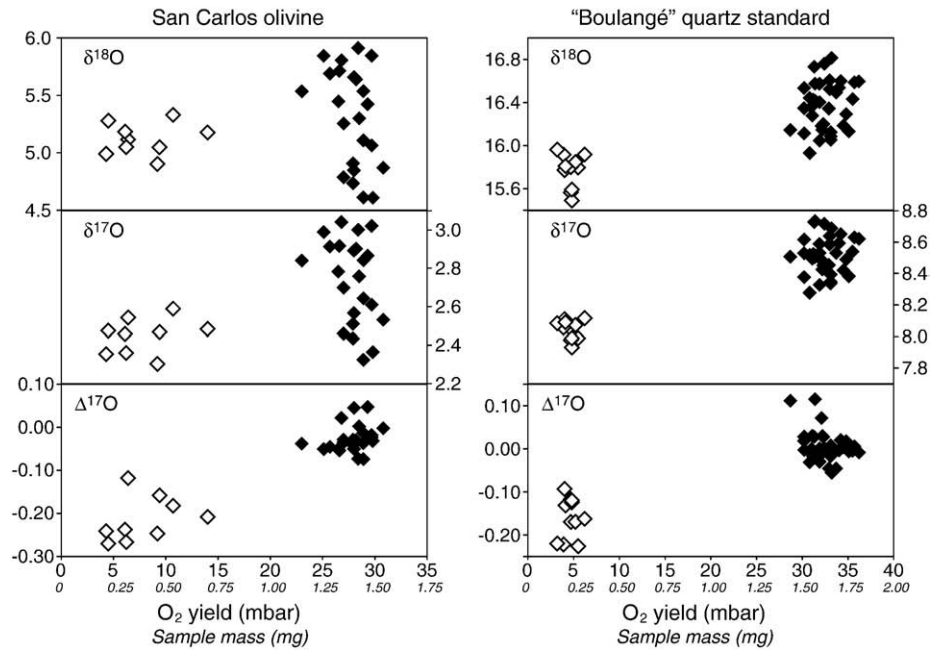
**Fig. 2.** Oxygen isotope measurements (in ‰ vs. V-SMOW) of standards. Microsamples (empty diamonds) of San Carlos olivine (0.2–0.7 mg) and "Boulangé" quartz standards (<0.3 mg) are compared with replicate analyses of 1.5 mg samples (solid diamonds). The approximate sample mass corresponding to the $O_2$ yield is indicated with the secondary labels.

using the IRMS technique (Miller et al., 1999): the oxygen three-isotopic composition was measured using a Micromass (Wythenshawe, Manchester, UK) PRISM III dual-inlet mass spectrometer. A systematic mass-dependent offset for small samples is also observed in this laboratory (Greenwood et al., 2007). The offset at the Open University appears to be due to isotopic fractionation associated with transfer of the gases in the inlet system, and therefore, again, independent of sample type, allowing a small correction relative to

the variations observed, to be applied: $-0.49‰$ for $\delta^{18}O$, $-0.23‰$ for $\delta^{17}O$.

## 3. Results

Fig. 3 features our $\delta^{18}O$ and $\delta^{17}O$ single micrometeorite IRMS data, with data obtained for previous studies by fluorination of composite samples (Clayton et al., 1986) and ion probe data measured on Antarctic
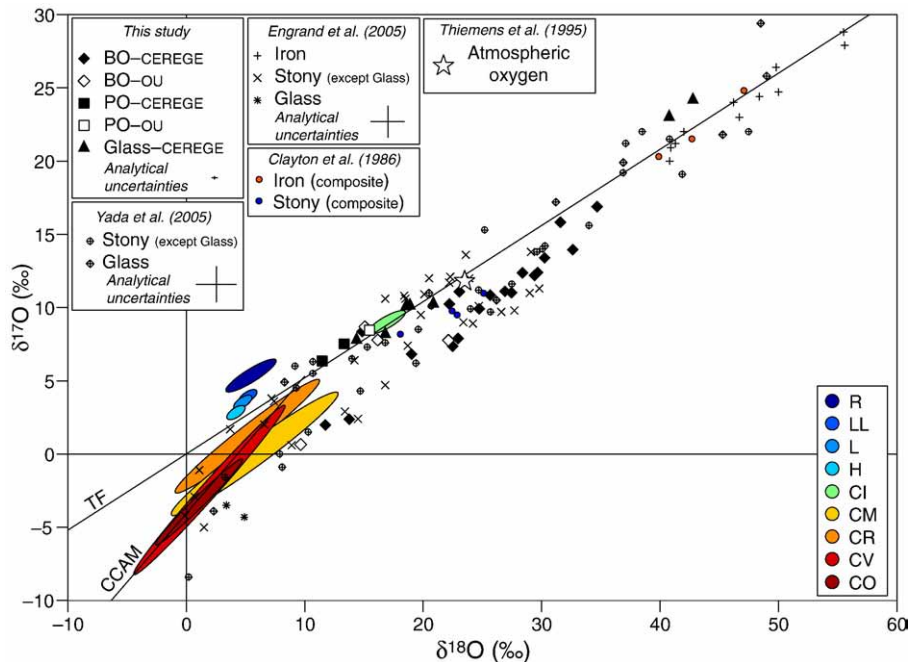


**Fig. 3.** Oxygen isotopic compositions of cosmic spherules (in ‰ vs. V-SMOW) measured: (1) at CEREGE and Open University using the IRMS technique (typical analytical uncertainties are represented: $\pm0.5‰$ for $\delta^{18}O$, $\pm0.2‰$ for $\delta^{17}O$); (2) by Clayton et al. (1986) using the conventional fluorination technique on batches of cosmic spherules; and (3) by Engrand et al. (2005) and Yada et al. (2005) using ion probe (typical analytical uncertainties for individual analyses are represented). Fields for CO, CV, CR, CM, CI, H, L, LL and R chondrites are represented (Clayton et al., 1991; Clayton and Mayeda, 1999). The solid line labeled TF is the terrestrial fractionation line (approximated $\delta^{17}O = 0.52 \times \delta^{18}O$), and the solid line labeled CCAM is the carbonaceous chondrite anhydrous minerals line (approximated $\delta^{17}O = 0.938 \times \delta^{18}O - 4.06$) (Clayton and Mayeda, 1999).

**Table 1**

| Sample name | Type | Mass (µg) | Oxygen isotope measurements | | | |
|---|---|---|---|---|---|---|
| | | | Lab. | $\delta^{18}O$ | $\delta^{17}O$ | $\Delta^{17}O$ |
| A1 | BO | 439 | C | 28.4 | 12.4 | −2.4 |
| A2 | BO | 420 | C | 22.5 | 7.4 | −4.3 |
| A3 | BO | 350 | C | 32.6 | 14.0 | −3.0 |
| A4 | BO | 276 | C | 19.0 | 6.8 | −3.1 |
| A5 | BO | 501 | C | 25.7 | 10.8 | −2.5 |
| A6 | BO | 458 | C | 30.3 | 13.4 | −2.3 |
| A7 | BO | 262 | C | 11.8 | 2.0 | −4.1 |
| A8 | BO | 282 | C | 27.5 | 11.0 | −3.3 |
| A9 | BO | 660 | OU | 22.1 | 7.8 | −3.8 |
| A10 | BO | 404 | OU | 9.7 | 0.7 | −4.4 |
| A11 | BO | 556 | C | 29.4 | 12.2 | −3.1 |
| A12 | BO | 690 | C | 22.9 | 7.9 | −4.0 |
| A13 | BO | 386 | C | 24.7 | 9.9 | −2.9 |
| A14 | BO | 528 | C | 26.9 | 11.1 | −2.9 |
| A15 | BO | 330 | C | 13.8 | 2.4 | −4.8 |
| A16 | BO | 445 | C | 29.7 | 12.4 | −3.0 |
| B1 | BO | 395 | C | 31.6 | 15.8 | −0.6 |
| B2 | BO | 590 | C | 34.7 | 16.9 | −1.2 |
| B3 | V | 444 | C | 16.8 | 8.2 | −0.6 |
| B4 | V | 559 | C | 20.9 | 10.3 | −0.6 |
| B5 | BO | 466 | OU | 16.2 | 7.8 | −0.7 |
| B6 | BO | 314 | C | 23.1 | 11.1 | −0.9 |
| B7 | BO | 353 | C | 22.2 | 10.2 | −1.3 |
| C1 | PO | 392 | C | 13.3 | 7.5 | 0.6 |
| C2 | BO | 541 | C | 14.9 | 8.4 | 0.6 |
| C3 | V | 438 | C | 18.9 | 10.1 | 0.3 |
| C4 | V | 393 | C | 18.6 | 10.1 | 0.4 |
| C5 | V | 481 | C | 14.4 | 7.8 | 0.3 |
| C6 | PO | 728 | C | 11.5 | 6.4 | 0.4 |
| C7 | BO | 636 | OU | 15.1 | 8.7 | 0.8 |
| C8 | PO | 877 | OU | 15.5 | 8.5 | 0.4 |
| D1 | V | 302 | C | 42.8 | 24.2 | 1.9 |
| D2 | V | 314 | C | 40.8 | 23.0 | 1.8 |

Type: BO=barred olivine; PO=porphyritic olivine; V=glassy. Lab.: C=CEREGE; OU=Open University. Oxygen isotope ratios are in ‰ vs. V-SMOW.

CSs (Engrand et al., 2005; Yada et al., 2005) together with bulk isotopic compositions of meteorites (Clayton et al., 1991; Clayton and Mayeda, 1999). Our data covers the spectrum of $\delta^{18}O$ and $\delta^{17}O$ values obtained by previous studies on stony CSs, with a much better precision. Furthermore, data obtained in both laboratories are in good agreement. It is possible to roughly distinguish 4 groups of isotopic signatures in our data (Table 1; see Section 4 for a discussion about this grouping).

*Group 1* (samples A1–A16, 48% of the total): most CSs with barred olivine (BO) textures (16/23) have $\Delta^{17}O \approx -3$ to $-5‰$ and $\delta^{18}O$ in the 10–30‰ range.

*Group 2* (samples B1–B7, 21% of the total): five BO CSs and 2 glass CSs have $\Delta^{17}O \approx -1‰$ and $\delta^{18}O$ in the 15–35‰ range.

*Group 3* (samples C1–C8, 24% of the total): two BO CSs, all 3 CSs with porphyritic olivine (PO) texture, and 3 glass CSs have $\Delta^{17}O \approx 0.4$ to $0.8‰$ and $\delta^{18}O$ in the 10–20‰ range.

*Group 4* (samples D1–D2, 6% of the total): 2 glass CSs have $\Delta^{17}O \approx 1.8‰$ and $\delta^{18}O \approx 41‰$, which is near to the value found for three glass CSs in a previous study (Yada et al., 2005).

## 4. Discussion

The isotopic signature of micrometeorites depends on the signature of the parent body, and of two processes: contamination by atmospheric oxygen, and mass-dependent fractionation during atmospheric entry. When micrometeoroids enter the atmosphere, they melt at an altitude of 85–90 km (in the upper mesosphere), and they are quenched in a few seconds (Love and Brownlee, 1991). When the particle is melted, oxygen is exchanged with the environment. The isotopic signature of atmospheric $O_2$ has been shown to be a constant $\delta^{18}O \approx 23.5‰$, $\delta^{17}O \approx 11.8‰$, up to an altitude of 60.9 km (Thiemens et al., 1995). Based on the observation of a downward flux of light O isotopes in the upper stratosphere and a qualitative modelling (Colegrove et al., 1965), Thiemens et al. (1995) suggested that a source of heavy O isotopes may exist in the upper mesosphere. However, no origin for such an isotopic fractionation can be specified. Therefore, we use the stratospheric isotopic composition $\delta^{18}O \approx 23.5‰$, $\delta^{17}O \approx 11.8‰$ as a minimally heavy O isotope-enriched composition to represent the oxygen micrometeorites encounter during atmospheric entry. The heating of the particles also causes loss of material by evaporation (Engrand et al., 2005) and
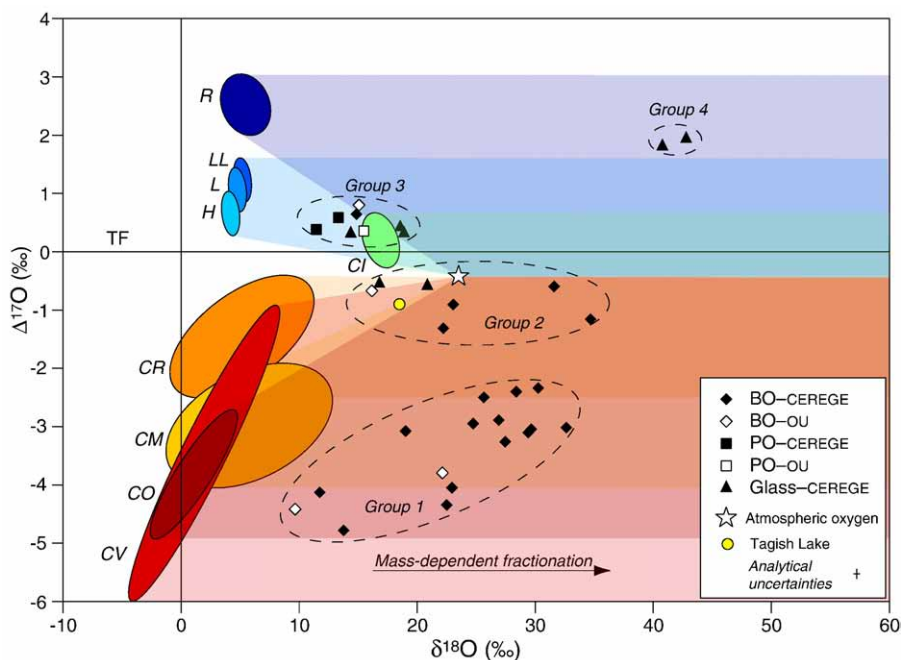


**Fig. 4.** $\Delta^{17}O$ vs. $\delta^{18}O$ values (in ‰ vs. V-SMOW) of individual cosmic spherules measured by IRMS (typical analytical uncertainties are represented: ±0.5‰ for $\delta^{18}O$, ±0.08‰ for $\Delta^{17}O$). Potential parent bodies are displayed by colored domains (Clayton et al., 1991; Clayton and Mayeda, 1999). The whole rock isotopic composition of Tagish Lake meteorite (Brown et al., 2000) is also represented. The range of possible values of $\Delta^{17}O$ and $\delta^{18}O$ for a micrometeorite from a given parent body is represented with a shaded area.

separation of iron–nickel droplets (Genge and Grady, 1998), which induce a mass-dependent fractionation of the oxygen. These processes may have various contributions to the measured isotopic signature, depending on the particle velocity, entry angle, iron–nickel content and mineralogy.

Fig. 4 represents $\Delta^{17}O$ as a function of $\delta^{18}O$ for our samples, with possible parent bodies: carbonaceous chondrites CO, CV, CM, CR, CI; ordinary chondrites H, L, LL; and R chondrites (Clayton et al., 1991; Clayton and Mayeda, 1999). Starting from a parent material, we can define a domain of possible values for the isotopic signature of micrometeorites bounded by a line parallel to the TFL along which mass-dependent fractionation occurs, and a line connecting the value of the parent material to that of atmospheric oxygen, along which pure contamination by terrestrial oxygen occurs.

Iron CSs, derived from particles made entirely of metal grains, provide a direct test of this model as they do not contain oxygen before melting and therefore the oxygen they contain now is derived entirely from the atmosphere. Their $\Delta^{17}O$ is indeed close to the atmospheric value (Clayton et al., 1986; Engrand et al., 2005), with $\delta^{18}O$ in the 40–60‰ range (Fig. 3). Therefore, the oxygen isotopic signature of CSs is expected to lie between an extraterrestrial end-member composition — that may be a known meteorite group or an unknown parent body — and an "atmospheric entry end-member" that is exemplified by the iron CSs.

CSs from *Group 1* and *Group 2* appear to be related to carbonaceous chondrites, with the CO/CV and CM/CR end-members. Although most of these CSs could originate from any type of carbonaceous chondrite, the 5 micrometeorites with $\Delta^{17}O < -4$ cannot be related to CM/CR parent bodies, and have a CO/CV chondrite-related parent body. The isotopic composition of the two glass CSs and one BO CS from *Group 2* (B5) can only be explained by a CR parent body. *Group 2* CSs could also be related to the parent body of the Tagish Lake meteorite (Brown et al., 2000). CSs from *Group 1* plot along a line of increasing $\Delta^{17}O$ with increasing $\delta^{18}O$, the slope of which reflects the relationship between mixing with atmospheric oxygen and fractionation during atmospheric entry. The differences in mixing/fractionation magnitude within this group are most probably related with the entry parameters of the particles: high-velocity particles and high-entry angle particles encounter more oxygen and are more heated, and should experience more mixing and fractionation. Fig. 5 is an attempt to quantify the degree of mass fractionation compared with mixing with atmospheric oxygen for BO CSs of *Group 1* and *Group 2* (excluding one outlier: B5): the percentage of mixing, defined between a parent body composition and atmospheric composition, is represented as a function of the maximum fractionation the particle went through. Assuming that *Group 1* CSs have a CO/CV parent body, and that *Group 2* CSs have a CM/CR parent body, we find that both groups have a similar trend, which indicates that the relative degree of mixing and fractionation is the same. Conversely, assuming that *Group 2* CSs have the same parent body as *Group 1* CSs would give very different relationships between mixing and fractionation for the two groups. Therefore, the definition of *Group 1* and *Group 2* based on the isotopic composition is also consistent once similar atmospheric entry effects and different parent bodies are assumed.

The isotopic compositions of CSs from *Group 3* are not greatly different from *Group 2* in absolute values. However, they are clearly different in terms of parent bodies. Indeed, atmospheric entry processes tend to pull the composition closer to the TFL, but it is impossible for the composition to cross this line. CSs from *Group 3* seem to have an ordinary chondrite-related parent body: although glass micrometeorites from this group are closer to the CI chondrite domain, a CI parent body is not consistent with these results once atmospheric effects are considered. Indeed, the sequence of textures with increasing heating and evaporation of Fe: PO<BO<glass (Taylor et al., 2000) is also found in the increasing $\delta^{18}O$ (fractionation) and decreasing $\Delta^{17}O$ (contamination by atmospheric oxygen) for micro-
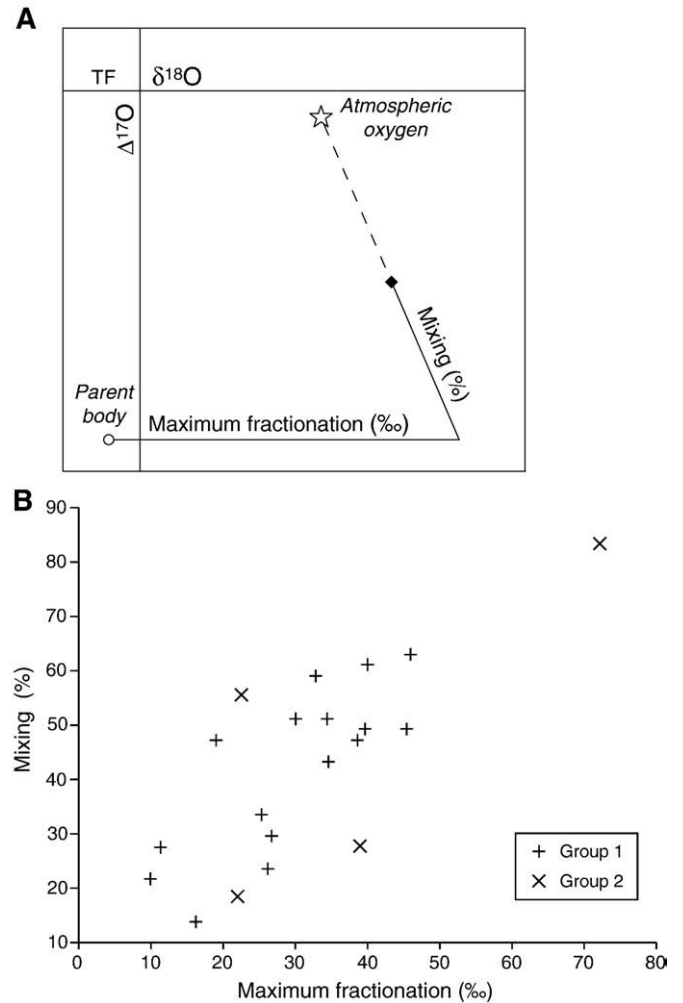
Fig. 5. Quantification of atmospheric entry effects. (A) Definition of "maximum fractionation" and "mixing." (B) Atmospheric entry effects using $\delta^{18}O = -4‰$, $\Delta^{17}O = -5.5‰$ as parent body composition for *Group 1* CSs and $\delta^{18}O = 0‰$, $\Delta^{17}O = -1.5‰$ for *Group 2*. The slopes are 0.01 ($R^2 = 0.61$) for *Group 1* and 0.009 ($R^2 = 0.56$) for *Group 2*.

meteorites of *Group 3*. This is consistent with PO CSs retaining pristine oxygen isotopic composition in relict grains (Genge et al., 2008).

Fig. 6 presents our data for *Group 4* micrometeorites with possible parent bodies. The only known parent material for these glass CSs could be the R chondrites, or the high $\Delta^{17}O$ component observed in the magnetites (Choi et al., 1998) and the mesostasis of unequilibrated ordinary chondrites (Franchi et al., 2001). Enrichments in $^{17}O$ and $^{18}O$ are known for some atmospheric components (Thiemens et al., 1995) and therefore a possible atmospheric effect in these glass spherules with very high $\delta^{18}O$ cannot be entirely ruled out. These specimens may also be related to CSs with enrichments in $^{17}O$ reported by Yada et al. (2005); the data from this study are also represented on Fig. 6. Interestingly, one cryptocrystalline spherule with a very high enrichment in $^{17}O$ is located on the crystobalite line (CRIL; Bridges et al., 1998) defined by measurements of crystobalites in the mesostases of unequilibrated ordinary chondrite Parnallee. This CS could be related to the $^{17}O$ enriched end-member hypothesized to explain this trend. This component could also yield the compositions observed in *Group 4* CSs by interaction with the atmosphere during the entry.

Magnetic measurements (Fig. 7) give more insight into the key mineral controlled by heating during atmospheric entry: magnetite. As magnetite is the only significant ferromagnetic mineral in melted micrometeorites (Suavet et al., 2009b), the value of the saturation magnetization $M_s$ is directly proportional to the amount of magnetite
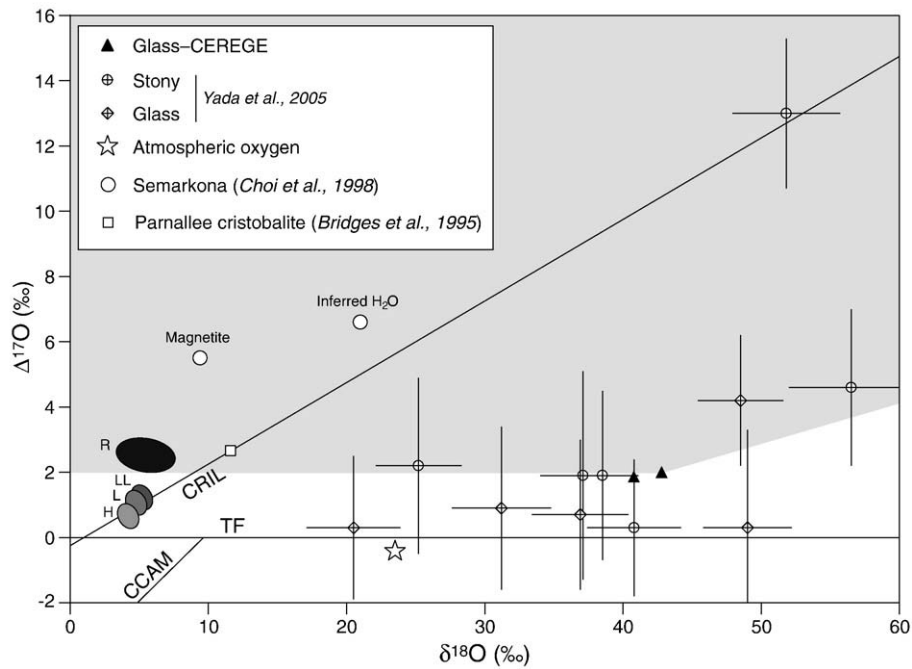
**Fig. 6.** $\Delta^{17}O$ vs. $\delta^{18}O$ values (in ‰ vs. V-SMOW) of micrometeorites from *Group 4*, with high-$\Delta^{17}O$ micrometeorites of Yada et al. (2005) and potential parent bodies: R chondrites, magnetites and inferred water of Semarkona, and crystobalite from Parnallee (only the most depleted values in $^{16}O$ are represented). The range of possible oxygen isotopic compositions for the parent bodies of *Group 4* micrometeorites is represented by the light-grey area.

in the micrometeorite (the magnetite wt% is $M_s/0.92$ (Dunlop, 2002)). A trend of higher magnetite content for CSs with higher $\delta^{18}O$ is visible in *Group 2*.

The positive correlation of magnetite amount with $\delta^{18}O$ is consistent with the processes of oxidation by high-$\delta^{18}O$ atmospheric oxygen and mass-dependent fractionation for particles that experience more heating. Magnetite amount may essentially trace the amount of oxidation of $Fe^{2+}$ in the melt of olivine composition. *Group 1* seems to have a similar trend, although two outliers have uncorrelated high $M_s$ values that may indicate presence of magnetite
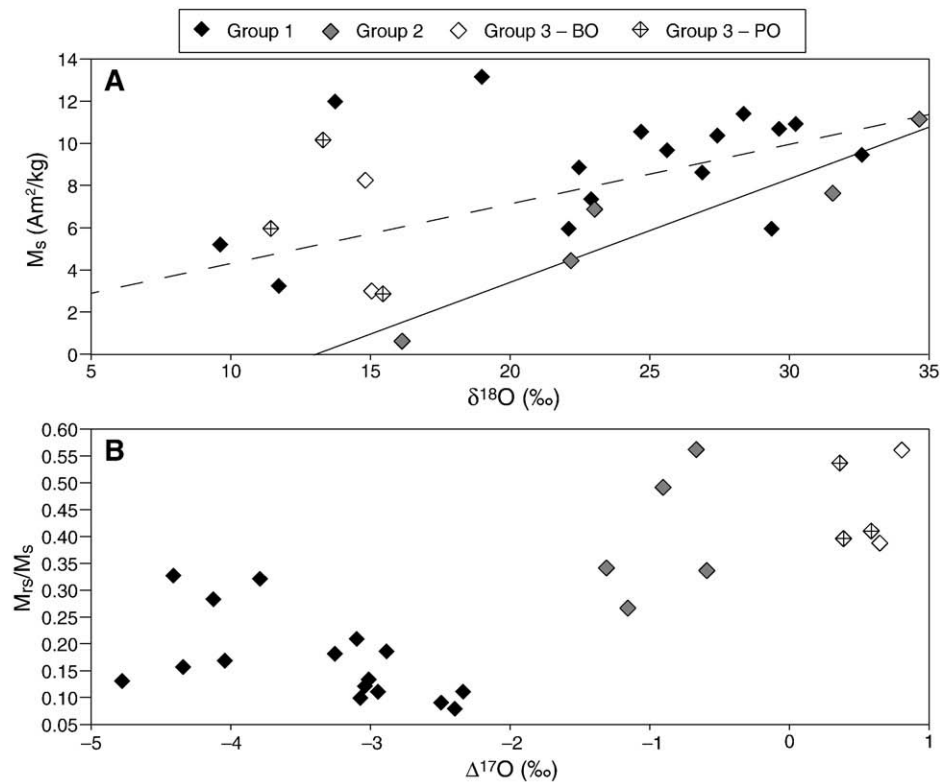


**Fig. 7.** Magnetic properties of micrometeorites from isotopic *Groups 1, 2* (barred olivine (BO) texture only) and *3* (BO and porphyritic olivine (PO) textures). (A) Saturation magnetization $M_s$ as a function of $\delta^{18}O$ (‰ vs. V-SMOW). Correlation lines are drawn for *Group 1* (dashed line, $R^2 = 0.56$ excluding two outliers) and *Group 2* (solid line, $R^2 = 0.88$). (B) Ratios of saturation remanent magnetization $M_{rs}$ and saturation magnetization $M_s$ as a function of $\Delta^{17}O$ (‰ vs. V-SMOW).

before atmospheric entry. Based on the observed correlation, the preatmospheric entry end-member composition can be predicted for $M_s = 0$ at $\delta^{18}O \approx 13‰$ for *Group 2* and $\delta^{18}O \approx -5$ for *Group 1* (if we exclude the two outliers), which is roughly in agreement with the inferred CM and CO/CV carbonaceous chondrite compositions (Clayton and Mayeda, 1999). The value of the ratio of saturation remanent magnetization $M_{rs}$ and saturation magnetization $M_s$ gives an estimate of the magnetite grain-size (Dunlop, 2002). Higher ratios correspond to smaller grain-size. Micrometeorites from *Group 1* seem to have larger magnetite grains than *Group 2* and *Group 3*. As magnetite is formed by oxidation of iron bearing phases, the magnetite grain-size differences probably reflect differences in composition of the parent minerals (especially fayalite mole percent (Fa%) in olivine), although differences in heating kinetics — as the entry velocity and angle vary for particles of different origins (Love and Brownlee, 1991) — could also cause them. Larger grain sizes are expected for the oxidation of high Fa% olivine. This is in agreement with the lower Fa% observed in H and L chondrites (*Group 3*) with respect to average carbonaceous chondrites (*Group 1*).

Our finding of 30% of micrometeorites above the TFL, while previous studies by ion probe found none (Engrand and Maurette, 1998; Matrajt et al., 2006) or 6% (Yada et al., 2005), may be related to the lower precision of these previous analyses (e.g. not allowing to distinguish the ordinary chondrite-related samples from the TFL). It could also indicate a higher proportion of ordinary chondrite-related material in our large (diameter >500 μm) CSs, that may show an intermediate proportion between meteorites (80% above TFL) and small micrometeorites (0–6% above TFL). Recent petrogeochemical investigations of coarse grained unmelted micrometeorites suggest that about 20% of all micrometeorites would be ordinary chondrite-related (Genge et al., 2008), which is in agreement with our results. It has been suggested that the high abundance of CM/CR related unmelted micrometeorites may be a result of the dust production mechanisms rather than the abundance of their parent bodies in the main asteroid belt (Genge, 2006). This idea is confirmed by impact destruction experiments (Flynn et al., 2009) on hydrous (CM) and anhydrous (ordinary chondrite) targets: there is an order of magnitude increase of dust production in the 30–300 μm size range for the hydrous target. This may explain why previous authors, who studied mainly the 150–250 μm size fraction, have concluded for an overwhelming dominance of CM parent body related micrometeorites while the large (our study) or small (IDPs) fractions may sample different populations of dust particles in Earth-crossing orbits. Our finding of CO/CV-related cosmic spherules in the >500 μm size fraction is consistent with the study of Genge et al. (2008), who explained the absence of unmelted micrometeorites with affinities to CO/CV by the higher eccentricities of their potential asteroidal sources, resulting in a higher entry velocity and a more intense heating during atmospheric entry. Our results could also reflect a variation of the composition of the flux of extraterrestrial materials to the Earth: micrometeorites from the Transantarctic Mountains sample this influx at the hundreds of thousands of years timescale, whereas other studies were made on samples accreted in the last few thousands of years.

## 5. Conclusions

The availability of the TAM micrometeorite collection, and of IRMS for high-precision oxygen isotope measurements, is a great opportunity for the characterization of the extraterrestrial dust flux entering Earth's atmosphere over the million-year time scale (Rochette et al., 2008; Folco et al., 2008, 2009). The discovery of significant numbers of micrometeorites with ordinary chondrite and CO/CV carbonaceous chondrite oxygen isotopic compositions is a strong argument against the idea that all micrometeorites derive from carbonaceous chondrite-related parent bodies that are not sampled as meteorites. It

supports the claim that ordinary chondrite- and CO/CV-related asteroids produce dust, and that interplanetary dust in the Earth-crossing orbit partly originates from these objects.

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