



Decoupling of chemical and isotope fractionation processes during atmospheric entry of S-type micrometeorites

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During atmospheric entry, micrometeorites experience variable degrees of (i) evaporation due to gas drag heating and (ii) mixing with atmospheric oxygen. Evaporation affects the physical properties and chemical and isotopic compositions of fully melted cosmic spherules (CSs). Oxygen isotope ratios of pristine micrometeorites are commonly used to relate these particles to their appropriate parent bodies. However, the degree of mixing with atmospheric oxygen and isotope fractionation by evaporation in CSs generally remains unclear, leading to uncertainties in their initial oxygen isotope ratios, which in turn complicates the precursor body identification. Previously, several studies have estimated the degree of evaporation based on contents of major refractory elements Ca and Al in combination with Fe/Si atomic ratios. This now commonly adopted chemical classification system has not yet been assessed with O and Fe isotope variability. As evaporation leads to both isotope and chemical fractionation, it is imperative to verify whether the predicted amounts of evaporation based on isotopic and chemical proxies converge.

Here, we measure the major and trace element compositions of 57 chondritic (mostly vitreous) CSs, along with their Fe isotope ratios. The $\delta^{56}\text{Fe}$ isotope and chemical (K, Zn, Na or CaO and Al_2O_3 concentrations) fractionation in these particles show no correlation. This can be interpreted in two ways: (i) separate processes govern chemical and isotope fractionation or (ii) the selected proxies for isotope and/or chemical fractionation are inadequate. Because the initial Fe isotope ratios of chondrites display limited variation ($0.005 \pm 0.008\text{‰}$ $\delta^{56}\text{Fe}$), Fe isotope ratios in CSs are assumed to only have changed through evaporation. At the same time, the chemical compositions of CSs show larger variability, so the CSs are thus often not chemically representative of their precursor

bodies.

As oxygen isotope ratios are commonly used to identify the precursor bodies of (micro)meteorites, triple oxygen isotope ratios are measured in 37 of the 57 CSs. Based on the relationship between $\delta^{57}\text{Fe}$ and $\delta^{18}\text{O}$, the effect of evaporation on the O isotope ratios can be corrected, which allows for a more precise precursor body reconstruction. Via this method, two ^{16}O -poor spherules with greatly varying degrees of isotope fractionation ($\sim 1.0\text{‰}$ and 29.1‰ $\delta^{56}\text{Fe}$, respectively) can be distinguished. Furthermore, it is observed that CSs that likely have an OC-like heritage all underwent the same degree of atmospheric mixing ($\sim 8\text{‰}$ $\delta^{18}\text{O}$). These findings highlight the potential of including Fe isotope measurements to the regular methodologies applied to CS studies.