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A sulfur isotope dichotomy: insights from iron meteorites

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The origin of terrestrial volatiles is a long-standing problem in cosmochemistry because these elements influenced the physical structure, chemical evolution, and habitability of our planet. Nucleosynthetic isotope anomalies currently constitute the most robust tracer of planetary ancestry, but their application to the provenance of volatiles is limited by their absence among the most important of these elements [1]. Mass-dependent isotope compositions of H and N do not provide unique constraints; modern mantle and hydrosphere compositions overlap with both non-carbonaceous (NC) and carbonaceous (CC) materials (broadly associated with inner and outer solar system reservoirs, respectively) [e.g., 2, 3], and their high volatility implies mass-dependent isotopic fractionation during accretion and differentiation. The nucleosynthetic isotope anomalies of Zn (a trace moderately volatile element) reflect the NC-CC dichotomy observed in refractory elements [4]. However, extension of this observation to constrain the provenance of terrestrial CHNOPS – the "life-essential" volatiles – hinges on the assumption that Zn is a reliable, proportional tracer of these more impactful volatile species. Sulfur (S) possesses more than two stable isotopes, and so its mass independent isotopic variation might potentially serve as a basis for directly extending the NC-CC dichotomy to these elements.

We compiled a database of quadruple S–isotope ($^{32}S/^{33}S/^{34}S/^{36}S$) measurements of chondrites, stony achondrites, and iron meteorites. The S–isotope signatures of chondrites and stony achondrites document marked heterogeneity between phases, and existing data do not reveal coherent variations between groups. However, we identify a clear dichotomy between the mass-independent S–isotope compositions of NC and CC iron meteorites. As the moderately siderophile nature of S leads to its concentration and homogenization in metallic liquids during equilibrium partitioning, we suggest that iron meteorites best record the average S compositions of early planetesimals. These data constrain the mass-fraction of NC and CC materials in the Bulk Silicate Earth and establish an upper limit for the mass-fraction of CC-derived S in the Earth's core.

[1] Kleine, et al., Space Sci. Rev. 216, 55 (2020).

[2] Alexander et al., Science 337, 721–723 (2012).

[3] Piani, et al., Science 369, 1110–1113 (2020).

[4] T. Steller et al., Icarus 386, 115171 (2022).