ZIRCONIUM ISOTOPIC CONSTRAINTS ON EARLY SOLAR SYSTEM EVOLUTION AND PLANETARY BUILDING BLOCKS

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Introduction: Nucleosynthetic isotope anomalies provide critical insights concerning the structure and evolution of the Solar System, the mixing of reservoirs in the early protoplanetary disk, and the accretion history of the terrestrial planets. For instance, the recognition of the 'non-carbonaceous' and 'carbonaceous' meteorite families, which are defined by a bimodal distribution in isotopic signatures of Cr, O, Ti, and other elements have been proposed to represent ancient inner and outer Solar System reservoirs, respectively [1]. Whereas their origin is not yet unequivocally known, physicochemical processes (*e.g.*, thermal processing) [2] and isotopically anomalous late infalling material [3] have been suggested as potential causes for generating such isotopic signatures of nucleosynthetic origin.

By investigating a broad set of meteoritic and planetary samples for their zirconium isotopic signatures, we here aim to shed further light on these subjects. Zirconium (Zr) has five stable isotopes (^{90}Zr, ^{91}Zr, ^{92}Zr, ^{94}Zr, and ^{96}Zr) and is a highly refractory, fluid-immobile, and lithophile element. These characteristics imply that any nucleosynthetic anomaly present in meteorites is unlikely to be hampered by secondary processes like alteration on the parent body and that the Zr isotopic compositions of a given silicate mantle is representative of the entire accretion history of its planetary body. Thus, nucleosynthetic anomalies in Zr contrast and complement information obtained from (moderately) siderophile elements, which predominantly reflect the later stages of terrestrial planet accretion [4] and may be more prone to alteration.

Methods: Zirconium was separated from sample matrices using previously established methods [5], employing TODGA and LN spec resins and resulting in purified Zr cuts and low blanks (<0.5ng Zr). Measurements were performed at the University of Münster and LLNL on Neptune *Plus* MC-ICPMS employing a Cetac Aridus II desolvating system and a Jet sampler and H skimmer cone setup. Isotopic data are internally normalized to ^{94}Zr/^{90}Zr = 0.3381 following the exponential law and provided in the μ-notation (parts per million deviations from terrestrial standards), resulting in an external reproducibility (2SD) of ±6, ±6, and ±18 for μ^{91}Zr, μ^{92}Zr, μ^{96}Zr, respectively.

Results: Our data are more precise, but in overall agreement with previously published Zr isotope data [6-7] and terrestrial rock standards are indistinguishable from the NIST SRM3169 solution standard, demonstrating the accuracy of our methods. We find that relative to this terrestrial Zr isotopic composition, all meteorites investigated here show variable excesses in μ^{96}Zr (Figure 1), whereas μ^{91}Zr and μ^{92}Zr are generally indistinguishable from terrestrial Zr isotope values. Meteorites of carbonaceous pedigree and presumed outer Solar System provenance are consistently more anomalous in μ^{96}Z compared to their non-carbonaceous counterparts.

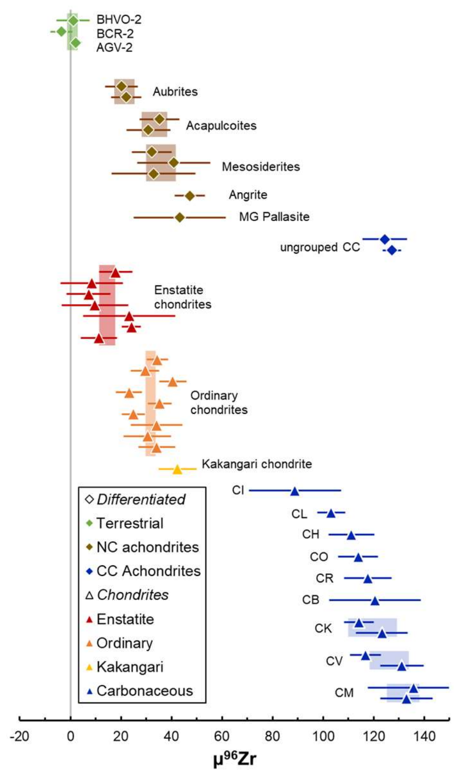

Figure 1: μ^{96}Zr isotopic compositions of planetary and meteoritic samples investigated in this study. Shaded areas indicate mean compositions for meteorite groups.

Discussion: The Zr isotopic signatures observed for bulk meteorites are consistent with variable deficits in Zr isotopes produced through the *s*-process of nucleosynthesis and overall well correlated with isotopic anomalies in elements of different nucleosynthetic origin (*e.g.*, Ca, Ti, Cr) and different cosmo-/geochemical behavior (*e.g.*, Mo, Ru). These ubiquitous deficits in *s*-process Zr demonstrate that Earth cannot have accreted from a mixture of known meteorites, and any such contribution would require addition of currently unknown meteoritic building blocks enriched in *s*-process Zr. Considering the apparent increment in μ^{96}Zr and other nucleosynthetic signatures with heliocentric distance [5], this hypothetical planetary building material would have most likely derived from the inner Solar System and suggests only miniscule contributions of outer Solar System material in the terrestrial accretion history.

[1] Warren (2011) *EPSL* 311, 93. [2] Trinquier et al., (2009) *Science* 324, 374. [3] Nanne et al. (2019) *EPSL* 511, 44. [4] Dauphas (2017) *Nature* 541, 521. [5] Render and Brennecka (2021) *EPSL* 555, 116705. [6] Akram et al. (2015) *GCA* 165, 484. [7] Elfers et al (2020) *GCA* 270, 475.