OPEN ACCESS

University of BRISTOL

Poole, G. M., Rehkämper, M., Goldberg, T., & Coles, B. J. (2015). Nucleosynthetic and Mass-Dependent Molybdenum Isotope Anomalies in Iron Meteorites: Constraints on Solar Nebula Heterogeneities and Parent Body Processes. Abstract from 46th Lunar and Planetary Science Conference, Texas, United States.

Publisher's PDF, also known as Version of record

Link to publication record in Explore Bristol Research
PDF-document

**University of Bristol - Explore Bristol Research**

**General rights**

## NUCLEOSYNTHETIC AND MASS-DEPENDENT MOLYBDENUM ISOTOPE ANOMALIES IN IRON METEORITES – CONSTRAINTS ON SOLAR NEBULA HETEROGENEITIES AND PARENT BODY PROCESSES.

G. M. Poole[1], M. Rehkämper[1], T. Goldberg[1] and B. J. Coles[1]. [1]Department of Earth Science and Engineering, Imperial College London, London SW7 2AZ, UK. (g.poole11@imperial.ac.uk)

**Introduction:** Nucleosynthetic isotope anomalies have long been known to exist at the grain-size level in primitive meteorites. Numerous recent studies have, furthermore, identified planetary-scale isotope anomalies for some refractory elements (e.g., Ba, Cr, Ti), whilst other elements exhibit no such effects (e.g., Hf, Os). Such isotopic anomalies are typically interpreted as representing an initial state of heterogeneity in the solar nebula. This inheritance of solar nebula signatures can be investigated further by examining mass-dependent stable isotope anomalies.

Molybdenum is ideally suited as a tracer of planetary-scale isotopic heterogeneity in the solar nebula as its seven isotopes were produced by distinct nucleosynthetic processes. However, despite its suitability, previously obtained Mo isotopic evidence has been questioned by other investigations that presented conflicting data [1-3].

Here we present Mo isotope data with improved precision and for a larger range of iron meteorites than in previous studies, to inform the debate about nucleosynthetic isotope anomalies. We have, furthermore, investigated mass-dependent Mo isotope fractionations for these samples, based on additional stable isotope analyses via the double-spike technique.

**Analytical techniques:** Iron meteorite samples of 1 to 15 g from 11 different groups were obtained from the Natural History Museum London and private collectors. The samples were sequentially leached in 0.5 M HBr, 6 M HCl and modified aqua regia (prepared with 6 M HCl), and then digested in modified aqua regia. Separation of Mo was achieved by a two-stage anion exchange procedure, which produced purified Mo fractions that typically displayed Ru/Mo and Zr/Mo ratios of $2.7 \times 10^{-5}$ or less. For measurements of stable Mo isotope compositions, the samples were mixed with a $^{100}$Mo-$^{97}$Mo double spike [4] prior to the separation chemistry.

The Mo isotopic compositions were measured by MC-ICP-MS using the Nu Plasma HR instrument at Imperial College. To determine nucleosynthetic Mo isotope anomalies, instrumental mass bias was corrected by internal normalisation to $^{98}$Mo/$^{96}$Mo = 1.453174 [5] using the exponential law. To investigate possible analytical artefacts, internal normalization to other ratios, including $^{92}$Mo/$^{98}$Mo = 0.607898 and $^{97}$Mo/$^{95}$Mo = 0.602083, were performed and found to produce internally consistent results. Normalization to

$^{98}$Mo/$^{96}$Mo resulted in larger anomalies and more distinctive anomaly patterns, in agreement with [3]. In the following, the nucleosynthetic Mo isotope anomalies are therefore described using this normalization.

The internally normalized data are reported in $\varepsilon^i$Mo notation (Fig. 1), where $\varepsilon^i$Mo = [($^i$Mo/$^{96}$Mo)$_{sample}$ / ($^i$Mo/$^{96}$Mo)$_{standard}$ - 1] × 10$^4$, calculated relative to the mean of several bracketing runs of NIST SRM 3134 Mo. External reproducibility (2σ) of the SRM measurements typically ranged from ±0.39 for $\varepsilon^{92}$Mo to ±0.14 for $\varepsilon^{97}$Mo. To determine stable Mo isotope compositions, the double spike data reduction utilized published methods [6, 7]. The sample data are reported in $\delta^{98}$Mo notation, calculated using $^{98}$Mo/$^{95}$Mo (Fig. 2) and determined relative to the mean obtained for bracketing analyses of NIST SRM 3134 Mo. Typical external reproducibility (2σ) was ±0.035‰ for $\delta^{98}$Mo.

The measured stable and nucleosynthetic Mo isotope data were combined to determine purely mass-dependent Mo isotope fractionation, by 'removing' any mass-independent effects. For this, the raw data acquired for unspiked samples were internally normalized to $^{97}$Mo/$^{95}$Mo, as both isotopes have similar s- and r-process contributions ($^{97}$Mo: 58% s- and 42% r-; $^{95}$Mo: 55% and 45%). Therefore, variations in these components will not significantly affect the $^{97}$Mo/$^{95}$Mo ratio. This is the same procedure as used by [8].

**Results:** Following internal normalization to $^{98}$Mo/$^{96}$Mo, the Mo isotope data for all magmatic iron groups display the typical 'w-shaped' isotopic patterns associated with s-process deficits (Fig. 1).
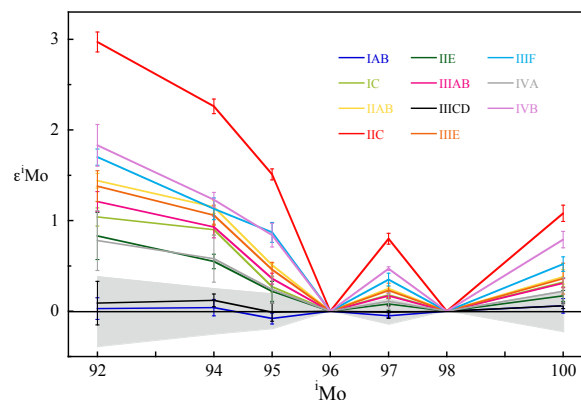


Fig. 1: Mean nucleosynthetic Mo isotope anomalies for iron meteorite groups, with 2se error bars. The grey shaded field represents 2σ external reproducibility of the SRM measurements. Normalized to $^{98}$Mo/$^{96}$Mo = 1.453174.

Whilst meteorites from the same group display consistent anomalies, the different groups show variable magnitudes of s-process deficits. In detail, IIC irons display the largest anomaly of 3.0 $\varepsilon^{92}$Mo, whilst IVAs displays the smallest effects of the magmatic irons at 0.8 $\varepsilon^{92}$Mo (Fig. 1). These results are in agreement with some published data [2, 3] but stand in contrast to others [1]. Like [3], we also find that the non-magmatic IAB and IIICD irons do not show anomalies (Fig. 1).
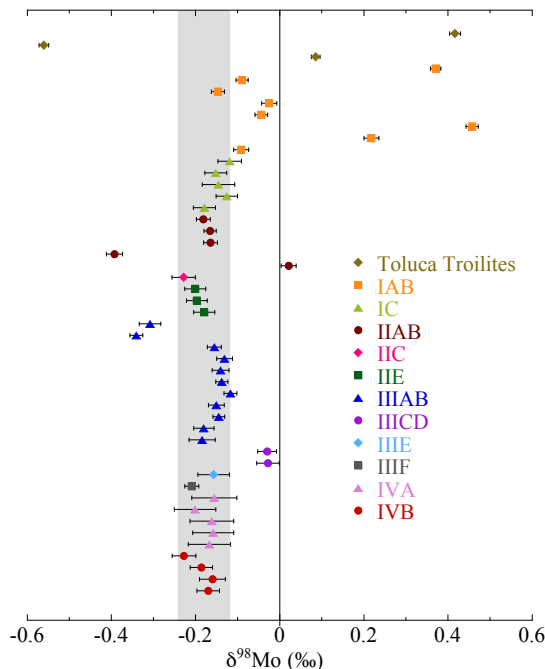


Fig. 2: $\delta^{98}$Mo = [($^{98}$Mo/$^{95}$Mo)$_{sample}$ / ($^{98}$Mo/$^{95}$Mo)$_{standard}$ - 1] × $10^3$. Data points represent mean of multiple analyses (n>5) for each meteorite sample with 2se error bars. The grey shaded field defines a 2σ uncertainty around the mean $\delta^{98}$Mo -0.18‰ of the magmatic iron meteorite groups.

Fig. 2 displays the stable fractionations. The $\delta^{98}$Mo values for the magmatic iron meteorites vary within a limited range, from –0.39‰ to +0.02‰. The non-magmatic irons show more variation (-0.17‰ to 0.46‰), whilst troilites (FeS) from Toluca IAB display the greatest range (-0.56‰ to +0.42‰) and exhibit the lightest and nearly the heaviest $\delta^{98}$Mo values measured here.

**Discussion:** The magnitude of the observed nucleosynthetic isotope anomalies appears to correlate with metallographic cooling rate, and hence inferred size, of the associated parent bodies (e.g., [9]). First suggested by [3], our much larger dataset for irons provides support for this observation. Such a correlation may reflect progressive homogenization of the solar nebula over the timescale of iron meteorite parent body accretion, possibly due to late injection of s-process material into the protosolar disc. Alternatively, the correlation may

imply more efficient homogenization in larger or more densely populated parent body feeding zones.

In our study of stable Mo isotope compositions, the iron meteorite groups (except IAB/IIICD) provide a mean $\delta^{98}$Mo of –0.18 ± 0.06‰ (2σ). As Mo is moderately siderophile, the $\delta^{98}$Mo value of the bulk parent body is (essentially) identical to the isotope composition measured for the core. Given the consistency of the results, our mean $\delta^{98}$Mo value for iron meteorite groups may closely approximate the solar system average. Notably, our mean is identical to a previously reported average of –0.16 ± 0.02‰ for irons and chondrites (excluding IAB, CK, CM), which was interpreted to reflect the Mo isotope composition of bulk planetary bodies in the inner solar system [8].

The large range of $\delta^{98}$Mo values observed for the Toluca troilites are most likely due to Mo isotope fractionation during metal–sulfide partitioning, whereby the observed variability may reflect both kinetic and equilibrium isotope effects. Similarly, the IAB/IIICD group shows highly variable $\delta^{98}$Mo with both isotopically heavy and light Mo. This stands in contrast to the results of [8], who found only heavy Mo isotope signatures for this group, which were interpreted to reflect evaporative loss of isotopically light Mo during energetic impacts. Based on our new data, we conclude that the large range of $\delta^{98}$Mo values observed for IAB/IIICD irons is due to the segregation and crystallization of S-rich melts on the parent body [10] and associated metal–sulfide partitioning.

We have also found small but resolvable and systematic variations of $\delta^{98}$Mo within magmatic iron groups. Here, the greatest variability is displayed by meteorites towards the Ni-rich end of the associated parent body fractional crystallization trend (e.g., [11]). Sulfur is incompatible in solid metal and thus becomes enriched in the liquid metal during fractional crystallization of cores. We therefore posit that the variations within magmatic iron meteorite groups represent processes associated with the increased liquid metal S content, most likely metal–sulfide partitioning of Mo.

**References:** [1] Becker, H. and Walker, R. J. (2003) *Nature, 425,* 152-155. [2] Dauphas, N., et al. (2002) *ApJ, 565,* 640-644. [3] Burkhardt, C., et al. (2011) *EPSL, 312,* 390-400. [4] Goldberg, T., et al. (2013) *JAAS, 28,* 724-735. [5] Lu, Q. and Masuda, A. (1994) *Int. J. Mass Spectrom. Ion. Proc., 130,* 65-72. [6] Siebert, C., et al. (2001) *GGG, 2,* art. no.-2000GC000124. [7] Rudge, J. F., et al. (2009) *Chem. Geol., 265,* 420-431. [8] Burkhardt, C., et al. (2014) *EPSL, 391,* 201-211. [9] Rasmussen, K. L., et al. (1995) *Geochimica Et Cosmochimica Acta, 59,* 3049-3059. [10] Benedix, G. K., et al. (2000) *Meteoritics & Planet. Sci., 35,* 1127-1141. [11] Wasson, J. T. (1970) *Icarus, 12,* 407-423.