

TESTING THE HOMOGENEITY OF THE SOLAR SYSTEM FOR IRON (54, 56, 57, and 58) AND TUNGSTEN (182, 183, 184, and 186) ISOTOPE ABUNDANCES. N. Dauphas^{1,2}, N. Foley², M. Wadhwa², A.M. Davis¹, C. Göpel³, J.-L. Birck³, P.E. Janney², and R. Gallino⁴, ¹Enrico Fermi Institute, The University of Chicago, 5640 South Ellis Avenue, Chicago, IL 60637, USA (dauphas@uchicago.edu), ²Department of Geology, The Field Museum, 1400 South Lake Shore Drive, Chicago, IL 60605, USA, ³Laboratoire de Géochimie-Cosmochimie, IGP, 4 Place Jussieu, 75252 Paris Cedex 05, France, ⁴Dipartimento di Fisica Generale, Università di Torino, Via Pietro Giuria 1, 10125 Torino, Italy.

Introduction: Except for a few notable exceptions, large-scale isotope homogenization is the rule that prevailed in the early solar system. Departures from this rule are referred to as isotopic anomalies. These mass-independent variations have proven very useful in the past for understanding how our solar system formed and evolved.

At a small scale, large isotopic variations have been found for some neutron rich nuclides in the Fe mass region (⁵⁰Ti and ⁵⁴Cr) [1,2]. Although of a much smaller magnitude, similar effects are also present in planetary objects [3,4]. Iron possesses 4 stable isotopes, 54 (5.845 %), 56 (91.754 %), 57 (2.119 %), and 58 (0.282 %). Interestingly, large isotopic anomalies affecting the neutron-rich nuclide ⁵⁸Fe have also been found at a small scale [5], raising the possibility that iron may be isotopically heterogeneous at a planetary scale.

Due to the huge abundance contrast between ⁵⁶Fe and ⁵⁸Fe, measuring all the isotopes of Fe simultaneously is technically challenging. In most configurations, when ⁵⁸Fe ion intensity is high enough to be measured at high precision, ⁵⁶Fe saturates its collector. For this reason, most high precision investigations of iron isotope abundances have focused on the 3 most abundant isotopes, 54, 56, and 57. The results obtained on a variety of objects [6-8], all fall on the terrestrial mass fractionation line. This is not unexpected because the isotope that is the most likely to vary is also the rarest, ⁵⁸Fe. Because of analytical difficulties, all the measurements of this isotope obtained so far have been plagued by large uncertainties (~ 5 ε at 2σ) [8-10]. The conclusion of these studies is that, at this level of precision and at a large scale, the solar system must have been homogenized for ⁵⁸Fe. Our ability to judge the degree of homogenization depends ultimately on the precision that is attainable during analysis. What is qualified as homogeneous today may prove to be heterogeneous as analytical capabilities improve. Here, we report the accurate composition of all iron isotopes in terrestrial samples and meteorites, with a precision that is one order of magnitude better than what had been obtained before for ⁵⁸Fe (0.5 ε at 2σ).

With the discovery of isotope anomalies for Mo [11-13] and Ru [14], it was recognized that some heavy elements synthesized by the *s*-, *p*-, and *r*-processes had escaped the global homogenization of the solar system. Some of these measurements were questioned recently [15,16]. The Mo anomalies observed by [11,12] correlate between the various isotopes. These anomalies also correlate with isotopic anomalies in Ru [17], recently measured by [14]. Furthermore, these correlations lie along lines predicted from the theory of *s*-process nucleosynthesis. It is very unlikely that analytical artifacts affecting different elements, in different laboratories, on different instruments, would correlate with each other and would mimic so closely a process governed by stellar physics. The variations measured for Mo and Ru must be real. As suggested by [12], the most likely host phase of these exotic compositions are mainstream SiC condensed in the envelopes of low-mass AGB-stars. Molybdenum, technetium, and ruthenium are known to have condensed quantitatively in these grains. If other elements were also trapped in SiC, then we should observe collateral isotopic effects for these elements as well.

Tungsten is an appealing candidate because it is refractory and forms stable carbides, it has enough isotopes to distinguish between various nucleosynthetic sources, and some of its isotopes received large contributions from the *s*-process. The ¹⁸²Hf-¹⁸²W chronometer (*t*_{1/2}=9 Ma) has proven very useful for constraining the time scale of metal-silicate differentiation on planetary objects, including the Earth [18]. One may wonder whether, in details, the *s*-process is a troublemaker for the chronometer.

Iron: The chemistry and mass spectrometry for high precision iron isotopic analyses were performed at the Isotope Geochemistry Laboratory (IGL) at The Field Museum, equipped with a GV Instruments IsoProbe MC-ICPMS. The isotopic composition of iron must be corrected for natural and instrumental mass fractionation in order to discuss the possible presence of isotope anomalies. This was done by arbitrarily fixing the ⁵⁷Fe/⁵⁴Fe ratio to a constant value and running terrestrial standards in between sample meas-

urements. The isotopic composition of iron is expressed in ϵ units,

$$\epsilon^i\text{Fe} = \left[\left(\frac{{}^i\text{Fe}/{}^{54}\text{Fe}}{({}^i\text{Fe}/{}^{54}\text{Fe})_{\text{STD}}} - 1 \right) \times 10^4 \right].$$

A method was developed that allows the determination of all iron isotopes with unprecedented precision. The isotopic composition of iron was measured in various terrestrial and extraterrestrial samples (Fig. 1).

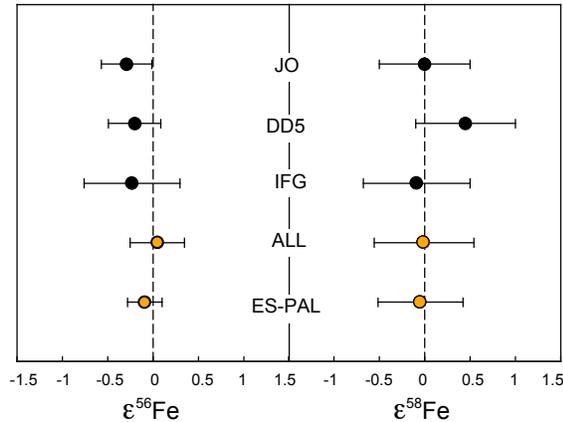


Fig. 1. Iron isotopic composition of a josephinite pebble from Josephine Creek (JO), an anhydrous lherzolite from Eifel (DD5), an Archean iron deposit from Isua (IFG), Allende CV3.2 (ALL), and Eagle Station (ES-PAL). See text for details and notations.

Josephinite (JO, Josephine Creek, USA) is the only occurrence of natural metallic nickel-iron alloy (Ni_2Fe to Ni_3Fe) on Earth. The main motivation for analyzing this mineral was to test whether Fe isotopes could be precisely measured in Ni-rich samples. Indeed ${}^{58}\text{Ni}$, which is the most abundant isotope of nickel (68.0769 %), can potentially interfere with ${}^{58}\text{Fe}$, the least abundant isotope of iron (0.282 %). If Ni had not been separated from Fe in this sample, this would have resulted in a $6 \times 10^4 \epsilon$ direct interference at mass 58. As illustrated in Fig. 1, the measured composition for josephinite is within 0.3 ϵ and 0.5 ϵ of the terrestrial composition for ${}^{56}\text{Fe}$ and ${}^{58}\text{Fe}$, respectively. An anhydrous lherzolite (DD5, Eifel massif, Germany) was analyzed to test the separation on a silicate matrix. Again, the measured composition is identical within uncertainties to that of the standard solution. Archean rocks show mass-independent variations for S isotopes [19]. The present interpretation is that, with no ozone shield at that time, the atmosphere was transparent to deep UV radiation that could photodissociate SO_2 - SO , a mechanism that is known to create mass-independent isotopic effects. Photo-oxidation of hydrated Fe^{2+} in oceans overlaid by a transparent atmosphere might have caused the precipitation of banded iron formations in the

Archean [20]. It is not known whether such a mechanism can induce mass-independent effects. IF-G is an IWG-GIT geostandard prepared from a large iron-ore deposit in the 3.8 Ga old Isua supracrustal belt (Greenland). No exotic effect has been observed in this rock. Allende (CV3.2) and Eagle Station (ES-PAL) are thought to have formed in the same region of the nebula. This region was characterized by extremely anomalous isotopic compositions compared to average solar system material. They therefore appear to be the most promising candidates to search for isotope anomalies at a planetary scale. As illustrated in Fig. 1, the iron isotopic composition is normal for these two meteorites ($\epsilon^{56}\text{Fe}=0.05 \pm 0.30$ and $\epsilon^{58}\text{Fe}=-0.01 \pm 0.55$ for Allende, $\epsilon^{56}\text{Fe}=-0.09 \pm 0.19$ and $\epsilon^{58}\text{Fe}=-0.05 \pm 0.47$ for Eagle Station). The solar nebula was probably homogenized at a planetary scale at the 0.2 ϵ and 0.5 ϵ level for ${}^{56}\text{Fe}$ and ${}^{58}\text{Fe}$, respectively.

Tungsten: Preliminary results on W have also been obtained on various meteoritic samples at the IGL at The Field Museum. These results seem to indicate the presence of a *s*-process W component in leaching experiments of primitive meteorites. This signature is released when SiC grains are digested. We are currently replicating these results at the Laboratoire de Géochimie-Cosmochimie IPGP (Paris) using N-TIMS.

References: [1] Heydegger H.R. et al. (1979) *Nature*, 278, 704. [2] Birck J.-L. and Allègre C.J. (1984) *GRL*, 11, 943. [3] Niemeier S. (1988) *GCA*, 52, 2941. [4] Shukolyukov A. and Lugmair G.W. (1998) *Science*, 282, 927. [5] Völkening J. and Papanastassiou D.A. (1989) *ApJ*, 347, L43. [6] Zhu X. K. et al. (2001) *Nature*, 412, 311. [7] Dauphas N. et al. (2003) *LPS XXXIV*, #1807. [8] Kehm K. et al. (2003) *GCA*, 67, 2879. [9] Rotaru M. (1992) *Nature*, 358, 465. [10] Podosek F.A. et al. (1997) *LPS XXVIII*, 121. [11] Dauphas N. et al. (2002) *ApJ*, 565, 640. [12] Dauphas N. et al. (2002) *ApJL*, 569, L139. [13] Yin Q. et al. (2002) *Nature*, 415, 881. [14] Chen J.H. et al. (2003) *LPS XXXIV*, #1789. [15] Becker H. and Walker R.J. (2003) *Nature*, 425, 152. [16] Lee D.-C. and Halliday A.N. (2003) *GCA*, 67, A246. [17] Dauphas N. et al. (2003) *GCA*, 67, A75. [18] Dauphas N. et al. (2002) *Geophys. Res. Lett.*, 29(6), 1084, doi: 10.1029/2001GL014237. [19] Farquhar J. and Wing B.A. (2003) *EPSL*, 213, 1. [20] Braterman P.S. et al. (1983) *Nature*, 303, 163.