ISOTOPIC EVOLUTION OF THE PROTOPLANETARY DISK AS RECORDED IN Mo ISOTOPES OF IRON METEORITES.

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Introduction: Nucleosynthetic isotope anomalies in bulk meteorites reveal a fundamental dichotomy between *non-carbonaceous* (NC) and *carbonaceous* (CC) materials [1], which represent two spatially distinct nebular reservoirs that coexisted in the circumsolar disk for several million years [2]. This prolonged spatial separation of the NC and CC reservoir is most likely related to the early formation of Jupiter between them [1,2]. Although there is little doubt that the NC and CC reservoirs were spatially separated, the extent of material exchange between them remains poorly constrained. For example, the Jupiter barrier may have resulted in a filtering effect by which the inward drift of large grains was efficiently blocked, while small dust grains could still pass through [3]. On this basis, [4] interpreted Ca isotope variations among NC meteorites to reflect a continuous compositional change of inner disk composition resulting from the inward drift of CC dust. This interpretation, however, depends on the assumed starting composition of the inner disk, and on the unknown efficiency of the Jupiter barrier over time. Thus, understanding and quantifying any compositional evolution of the NC and CC reservoirs is of considerable interest, as it would allow reconstructing the structure and temporal evolution of the solar accretion disk, and ultimately the growth history of Jupiter.

Molybdenum isotopes are key for addressing these issues. They have been instrumental for defining the dichotomy, because they can be measured in essentially all meteorite types, including iron meteorites [5]. However, thus far Mo isotopes have not revealed any systematic evolution in either the NC or CC reservoir, although both contain early (iron meteorites) and late formed bodies (chondrites). In part this might be due to the overall small Mo isotopic offset between the NC and CC meteorites, and additionally because the lack of a precise correction of cosmic ray exposure (CRE) effects on the Mo isotopic composition of iron meteorites. To address these issues we use Pt isotopes [6] to correct for CRE-effects on Mo isotopes in irons, and obtained combined Mo and Pt isotopic data for a large and comprehensive set of grouped und ungrouped iron meteorites. These data are used to assess any compositional heterogeneity within the NC and CC reservoirs that may have arisen through material exchange between both reservoirs.

Samples and methods: Several IIAB, IID, and IIIAB irons with variable CRE ages and neutron capture effects, as well as 26 ungrouped iron meteorites were selected for this study. Sample digestion and chemical separation and measurement of Mo and Pt followed our previously established methods [2].

Cosmic ray exposure effects: To correct measured Mo isotopic compositions for CRE effects using Pt isotopes, we determined empirical correlations of ε^{i}Mo vs. ε^{196}Pt for a set of IIAB, IID, and IIIAB irons with variable CRE effects (Fig. 1). This approach provides precise pre-exposure ε^{i}Mo values for each group from the intercept values at ε^{196}Pt=0. In addition, newly obtained and previously published Mo and Pt isotopic data for IC and IIIE irons [2,11] were used to calculate updated and more precise pre-exposure ε^{i}Mo values for these groups as well. The ε^{i}Mo vs. ε^{196}Pt slopes agree within uncertainty for all investigated groups, which makes it possible to use these slopes to individually correct measured Mo isotopic compositions of the ungrouped irons. Thus, with our approach it is possible to quantify and correct for CRE effects on Mo isotopes with unprecedented precision.

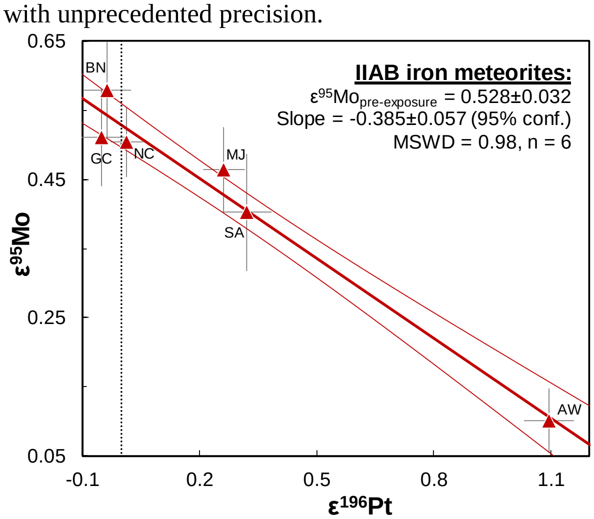

Fig. 1. Diagram of ε^{95}Mo vs. ε^{196}Pt for the IIAB iron meteorites. Similar correlations were obtained for other Mo isotopes, and for other iron groups. BN = Braunau, GC = Guadalupe y Calvo, NC = North Chile, MJ = Mount Joy, SA = Sikhote-Alin, AW = Ainsworth.

Mo isotopic variability: In a diagram of ε^{95}Mo vs. ε^{94}Mo, all irons of this study exhibit well-resolved isotope anomalies and—except Nedagolla—can be assigned to either the NC or CC reservoir (Fig. 2). The new and more precise data reveal that, even after correcting for CRE effects, there is scatter around the

NC- and CC-line. Moreover, there is a tendency for iron meteorite samples to plot below rather than above their respective NC- or CC-line, although only the IIAB irons are resolved from the NC-line. Incomplete quantification of CRE effects cannot be the cause for these variations, because samples with no CRE effects also deviate from the NC- and CC-lines.

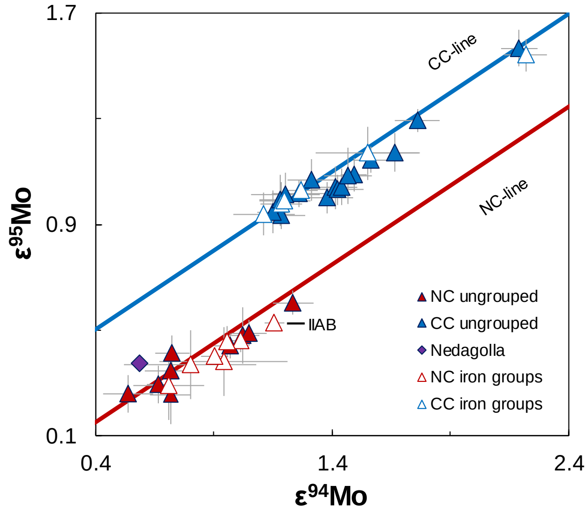

Fig. 2. Diagram of CRE-corrected ε^{95}Mo vs. ε^{94}Mo for grouped and ungrouped iron meteorites. Shaded area indicates the range of bulk meteorite literature data. NC- and CC-line from [5].

The new data indicate that although the absolute magnitude of the variability within each reservoir is small compared to the overall NC-CC difference, it is nevertheless significant. A better way to visualize the heterogeneity around the NC- and CC-lines is the Δ^{95}Mo notation (Fig. 3), which is the ppm deviation from a hypothetical *s*-process mixing line passing through the origin. As such, Δ^{95}Mo is a measure for a sample's *r*-process excess [5]. The NC and CC reservoirs have $\Delta^{95}Mo_{NC} = -9 \pm 2$ and $\Delta^{95}Mo_{CC} = +26 \pm 2$ [5]. For most irons the Δ^{95}Mo values scatter around and overlap with the respective values of the NC and CC reservoirs; however, most NC iron groups and several ungrouped CC irons tend to have slightly more negative values than the reference values of the NC or CC reservoir (Fig. 3).

Nedagolla−a unique iron meteorite: Nedagolla is the only sample that plots between the NC- and CC-line and is resolved from both of them. In contrast to the other irons, it also exhibits a radiogenic ε^{182}W value of -2.74, indicating a late melting and re-equilibration event. This non-magmatic origin is supported by its structure, petrography, and chemical composition [e.g., 9]. Hence, Nedagolla likely samples an impact-generated mixture of NC and CC material. Collisions between NC and CC bodies may have occurred after the breakdown of the dichotomy due to the growth and/or migration of the gas giant planets, which is expected to result in scattering of CC bodies into the inner Solar System [e.g., 10]. This is consistent with the elevated ε^{182}W value of Nedagolla, which corresponds to a Hf-W model age of ~8 Ma after CAI formation.

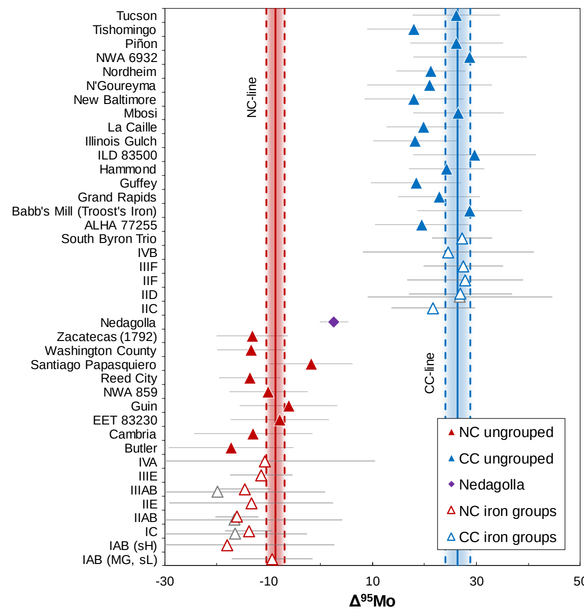

Fig. 3. Δ^{95}Mo of iron meteorites. Samples are sorted by name within each reservoir. Complimented with literature data from [5, 9 and references therein].

Implications for the NC-CC isotope dichotomy: Collectively, the new data confirm the fundamental dichotomy between NC and CC materials. The most plausible mechanism for such an efficient separation over several millions of years is the early formation of Jupiter, which acted as a barrier against exchange of material between the NC and CC reservoirs [2]. However, the scatter around the NC- and CC-lines indicates that this barrier still allowed small compositional changes within each reservoir. For instance, different areas of the disk may have received variable proportions of isotopically diverse cloud material while the Jupiter barrier was still being established and mixing between the two reservoirs not completely prohibited [12,13]. Additionally, small isotopic changes in the NC reservoir may result from the inward drift of CC dust.

References: [1] Warren P. H. (2011) *EPSL*, 311, 93-100. [2] Kruijer T. S. et al. (2017) *PNAS*, 114, 6712-6716. [3] Weber P. et al. (2018), *APJ*. 854. [4] Schiller et al. (2018) *Nature* 555 501-510 [5] Budde et al. (2019) *Nat. Astronomy* 3 736–741 [6] Kruijer et al. (2013) *EPSL* 361 162-172[7] Worsham et al. (2017) *EPSL* 467 157-166 [8] Hilton et al. (2019) *GCA* [9] Miyake and Goldstein 1974 *GCA* 38 747-755[10] Raymond and Izidoro (2017) *Icarus* 297 134-148 [11] Worsham et al. (2019) *EPSL* 521 103-112 [12] Burkhardt et al. (2019) *GCA* 261 145-170 [13] Nanne et al. (2019) *EPSL* 511 44-54