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Distinct evolution of the carbonaceous and non-carbonaceous reservoirs: Insights from Ru, Mo, and W isotopes

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27 Abstract

Recent work has identified a nucleosynthetic isotope dichotomy between "carbonaceous" (CC) 28 29 and "non-carbonaceous" (NC) meteorites. Here, we report new Ru isotope data for rare iron 30 meteorite groups belonging to the NC and CC suites. We show that by studying the relative isotopic characteristics of Ru, Mo, and W in iron meteorites, it is possible to constrain the 31 32 processes leading to the distinct isotope heterogeneities in both reservoirs. In NC meteorites, internally normalized, mass-independent isotope ratios of Mo and Ru are correlated, but those of 33 Mo and W are not. In CC meteorites, Mo and W isotope ratios are correlated, but those of Mo 34 35 and Ru are not; specifically, Mo isotopic compositions are variable and those of Ru are more restricted. The contrasting behaviors of Ru and W relative to Mo in the two reservoirs likely 36 require processing of the presolar carriers under distinct redox conditions. This provides further 37 evidence that NC and CC meteorites originated from spatially separated reservoirs that evolved 38 under different prevailing conditions. 39

40 Keywords: molybdenum; ruthenium; tungsten; nucleosynthetic heterogeneity; meteorite
41 dichotomy; thermal processing

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43 1. INTRODUCTION

The current structure of the solar system, i.e., terrestrial planets in the inner solar system and gaseous giants and icy moons in the outer solar system, resulted from the formation of chemical reservoirs early in the evolution of the protoplanetary disk. In recent decades, isotopic reservoirs have also been identified by the presence of nucleosynthetic isotope anomalies in most elements in bulk meteorites, which likely reflects the heterogeneous distribution of isotopically 49 diverse presolar materials in the early protoplanetary disk (see summary in Dauphas and Schauble, 2016). These nucleosynthetic isotope anomalies are small (typically identified at the 50 parts per 10^4 - 10^6 level), mass-independent, and are representative of the unique mixtures of 51 presolar materials in the various nebular reservoirs from which planetary bodies accreted. By 52 contrast, other elements, including Os and Pt, display no nucleosynthetic heterogeneity in bulk 53 meteorites at the current level of precision (with few exceptions - Goderis et al., 2015), 54 55 indicating that the solar nebula may have been initially isotopically homogeneous, or that these 56 elements were hosted in homogeneously distributed carriers (e.g., Walker, 2012; Kruijer et al., 2013). Considering this hypothesized initial homogeneity, the origin of the heterogeneous 57 58 distribution of presolar materials and, thus, the origin of nucleosynthetic anomalies remains ambiguous. Some proposed mechanisms involve inefficient mixing of presolar materials, 59 resulting in inherited heterogeneities in the solar nebula (Clayton, 1982; Dauphas et al., 2002a), 60 61 grain type- or size-sorting (Regelous et al., 2008; Dauphas et al., 2010), or thermal processing of unstable presolar phases (Trinquier et al., 2009). 62

Another facet of nucleosynthetic heterogeneity is the recent identification of a major 63 isotopic dichotomy among meteorites in several elements, including Ti, Cr, Mo, W, Ru, and Ni 64 (Warren, 2011; Budde et al., 2016; Kruijer et al., 2017; Poole et al., 2017; Worsham et al., 2017; 65 Bermingham et al., 2018; Nanne et al., 2019). Carbonaceous chondrites, several iron meteorite 66 groups, and some ungrouped irons and achondrites fall into the "carbonaceous" (CC) suite. The 67 "non-carbonaceous" (NC) suite is comprised of ordinary and enstatite chondrites and all other 68 69 iron meteorite groups and achondrites measured thus far. Broadly speaking, CC meteorites have 70 elevated abundances of nuclides synthesized in neutron-rich stellar environments (including rprocess Mo isotopes), relative to NC meteorites. The presence of both irons, which have older 71

estimated accretion ages, and chondrites, which have younger accretion ages, in both suites indicates that they represent different nebular reservoirs that were spatially distinct, and that the reservoirs remained separated for several Ma (Budde et al., 2016; Kruijer et al., 2017). Kruijer et al. (2017) suggested that the reservoirs were separated due to the growth of Jupiter's core. In this case, the NC and CC reservoirs represent the inner and outer solar system, suggesting that the conditions that affected presolar carriers in each reservoir may have been different.

To investigate the generation of nucleosynthetic variations among nebular reservoirs, we 78 utilize the relative isotopic characteristics of Mo, Ru, and W in iron meteorites. These elements 79 are useful because they have distinct physicochemical behaviors under different nebular 80 conditions. Further, Mo, Ru, and W are created by a combination of *p*-process, *s*-process, and *r*-81 process nucleosynthesis. Therefore, these elements are ideal tracers of diverse nucleosynthetic 82 signatures in solar system materials. We report new Ru isotope data for magmatic iron meteorite 83 groups belonging to both the NC (IC and IIIE) and CC suites (IIC, IID, IIF, and IIIF), for most of 84 which no Ru isotope data have been reported before. In conjunction with Mo and W isotope data, 85 the Ru data provide new constraints on the various processes and environmental conditions that 86 led to the isotope heterogeneities within the NC and CC reservoirs. 87

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89 2. ANALYTICAL METHODS

90 **2.1. Samples**

The samples used to obtain Ru isotope data were predominately adjacent pieces of the same samples that were used to obtain Mo, W, and Pt isotope data in the study of Kruijer et al. (2017). Additional samples were incorporated into this study, for which Ru and Mo isotope data 94 (and sometimes Pt) were obtained from aliquots of the same digestion. Platinum was used as a
95 neutron fluence dosimeter to monitor for the effects of cosmic ray exposure (CRE), which can
96 modify the isotopic compositions of Ru, Mo, and W (Kruijer et al., 2013; Fischer-Gödde et al.,
97 2015; Worsham et al., 2017).

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2.2. Chemical purification procedures

A detailed description of the purification procedures is given in the supplementary 100 material (SM). Briefly, iron meteorite samples between 0.3 and 0.6 g were digested in Teflon 101 102 beakers in 6M HCl with traces of HNO₃. Ruthenium was separated from the matrix using cation exchange chromatography, and was purified via micro-distillation (Birck et al., 1997; Fischer-103 Gödde et al., 2015). After purification of Ru, Mo/Ru and Pd/Ru were always $< 1 \times 10^{-5}$. 104 Molybdenum was separated and purified using a three-stage cation and anion exchange 105 106 chromatographic procedure, including a Tru-spec column to remove Ru (Burkhardt et al., 2011). The Zr/Mo and Ru/Mo after this chemistry was typically $< 5 \times 10^{-5}$. Platinum was separated using 107 a single-stage anion exchange chromatography procedure (Method 1; Rehkämper and Halliday, 108 1997). Given that the concentrations of Mo, Ru, and Pt are high in the iron meteorites studied 109 here, the total analytical blanks were negligible (< 1 ng for Ru and Pt, < 10 ng for Mo). 110

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112 **2.3.** Mass spectrometry

Ruthenium, Mo, and Pt analyses were conducted using a *Thermo Scientific Neptune Plus* multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS) at the Institut für Planetologie, University of Münster. Ion beams were collected simultaneously using Faraday

cups for 100 cycles. Molybdenum-97 and ¹⁰⁵Pd, ⁹¹Zr and ⁹⁹Ru, and ¹⁸⁹Os and ²⁰⁰Hg were used to 116 monitor and correct for interferences on Ru, Mo, and Pt, respectively. The Ru, Mo, W, and Pt 117 isotopic compositions are reported in ε notation (parts-per-10⁴ deviations from terrestrial 118 standards). Interference corrections for ε^{100} Ru were typically < 0.1 ε , usually on the order of a 119 few ppm. For ε^{i} Mo interference corrections were < 1 ε , usually on the order of 10s of ppm. The 120 data are normalized to ⁹⁹Ru/¹⁰¹Ru, ⁹⁸Mo/⁹⁶Mo, ¹⁸⁶W/¹⁸⁴W, and ¹⁹⁸Pt/¹⁹⁵Pt. Based on previous 121 studies from our lab, and monitored during this study, the external reproducibilities (2SD) of the 122 repeated analyses of terrestrial standards for each element are ± 0.13 for ϵ^{100} Ru, ± 0.28 for ϵ^{94} Mo, 123 ± 0.08 for ϵ^{183} W, and ± 0.07 for ϵ^{196} Pt. 124

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126 **3. RESULTS**

127 **3.1 Effects of cosmic ray exposure on Ru and Mo isotopes**

The CRE effects were monitored using Pt isotope data reported here and in Kruijer et al. 128 (2017) (Table 1). These effects are dependent, in part, on the shielding depth of the sample, so 129 samples used in this study typically came from within ~ 3 cm of the piece from which the Pt 130 isotope data were obtained. The effects of CRE on Ru and Mo have been described by Fischer-131 Gödde et al. (2015), Worsham et al. (2017), and Bermingham et al (2018). For Ru, the largest 132 effects are on ϵ^{100} Ru. For Mo, in order of largest to smallest, the effects are on ϵ^{92} Mo, ϵ^{95} Mo \approx 133 ϵ^{94} Mo, and ϵ^{97} Mo. Effects on ϵ^{100} Ru and ϵ^{94} Mo are similar in magnitude (i.e., ranging up to ~ 0.5 134 ε units, but typically < 0.15 ε). 135

136 Most samples used in this study have minimal CRE effects. Excluding the irons with the 137 most significant effects (Arispe, Bendego, Murnpeowie, Kokstad, and Oakley), CRE results in \leq 138 0.06 ε changes in the ε^{100} Ru and ε^{94} Mo values, averaging 0.02 ε for both. Therefore, no CRE 139 correction is necessary for either Ru or Mo for most samples, and we report uncorrected Ru and 140 Mo isotope data and low-exposure averages for each iron meteorite group, incorporating only 141 irons with ε^{196} Pt ≤ 0.13 (Tables 2, 3, and SM1; Figs. 1 and 2). Although unnecessary for most 142 irons, CRE-corrected Ru data are given in Table SM2. Robust CRE corrections could not be 143 done for Mo (see SM).

The use of uncorrected data is sufficient for the aims of this study, which is primarily concerned with isotopic differences between NC and CC groups. As shown in Figures 1 and 2, these differences are larger than those expected to arise from unaccounted-for CRE effects. However, CRE effects should be corrected when Mo and Ru isotopes are used for genetic testing or where the precision and accuracy of isotopic correlations are important (Bermingham et al., 2018).

Weighted-average literature data for other meteorite groups are also shown on Figure 1.
For the major iron meteorite groups, the averages include CRE-corrected group means (FischerGödde et al., 2015; Bermingham et al., 2018) and means of low-exposure irons from each group
(Chen et al., 2010; Burkhardt et al., 2011; Poole et al., 2017) (Tables SM3 and SM4). The ¹⁸³W
data do not require CRE correction (Kruijer et al., 2017).

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156 **3.2 Ru and Mo isotope results**

157 The Ru and Mo data are reported in Tables 2 and 3. The highest precision is obtained for 158 ϵ^{100} Ru, and the other ϵ^{i} Ru values are of lower precision because they are of lower abundance 159 (⁹⁶Ru and ⁹⁸Ru) or more difficult to measure precisely and accurately due to their distance in AMU from the normalizing ratio (⁹⁶Ru and ¹⁰⁴Ru). For this reason, and because ε^{100} Ru displays the most distinctive variations, only these values are presented here and discussed, although all Ru data are reported in Table SM1. Two pairs of duplicate analyses each for ε^{100} Ru and ε^{94} Mo reproduced well (within 7 ppm of one another). The only rare iron meteorite groups with previously reported Ru data are the IC and IID groups, and the new data for those groups are in good agreement (Table SM5 – Fischer-Gödde et al., 2015; Bermingham et al., 2018).

The Mo isotope data are also in generally good agreement with previous studies (Table 166 SM6 – Burkhardt et al., 2011; Poole et al., 2017; Bermingham et al., 2018). In detail, there are 167 potentially small systematic offsets, primarily in ϵ^{92} Mo, between this study (and Kruijer et al., 168 2017) and the data reported by Poole et al. (2017) (Table SM6). Apart from the IIIE group, the 169 ϵ^{92} Mo data reported by Poole et al. (2017) are within uncertainty of the data reported here but are 170 generally higher by 0.2 to 0.3 ε for the IC, IIC, IIIE, and IIIF groups. Some of these offsets are 171 likely due to the different exposure histories of the samples used in each study, but the 172 systematic nature of the offsets would suggest this is not always the case. As the ϵ^{94} Mo and 173 ϵ^{95} Mo values show no significant offsets, these data are used in the following discussion. 174

The Mo isotope dichotomy (Fig. 1) is partially defined by excess ⁹⁵Mo, relative to ⁹⁴Mo, 175 in CC meteorites when compared to NC meteorites, resulting in two parallel trends on a plot of 176 ϵ^{94} Mo versus ϵ^{95} Mo (e.g., Budde et al., 2019). The Mo isotopic compositions indicate that the IC 177 and IIIE groups belong to the NC suite, in addition to the IAB, IIAB, IIIAB, and IVA iron 178 meteorite groups and the ordinary and enstatite chondrites (Fig.1; Kruijer et al., 2017). The IIC, 179 IID, IIF, and IIIF groups, in addition to the IVB iron group and carbonaceous chondrites, belong 180 to the CC suite (e.g., Kruijer et al., 2017). In Fig. 1, only carbonaceous chondrite metals are 181 shown, as Mo and Ru isotopic compositions obtained from separate digestions of bulk 182

unequilibrated chondrites are not directly comparable due to the potential for incompletedigestion of presolar phases.

The new ϵ^{94} Mo and ϵ^{100} Ru data for IC, IIC, IID, IIF, IIIE, and IIIF irons are shown in 185 Fig. 2, supplemented with Mo and W isotope data from Kruijer et al. (2017). Iron meteorites 186 from the NC groups IC and IIIE have Mo and Ru isotopic compositions most like IIAB iron 187 meteorites (ϵ^{94} Mo ~ 1.0; ϵ^{100} Ru ~ -0.5). Iron meteorites from the CC groups IID, IIF, and IIIF 188 have Mo and Ru isotopic compositions similar to IVB irons and carbonaceous chondrite metals 189 $(\epsilon^{94}$ Mo ~ 1.2; ϵ^{100} Ru ~ -1.0). The IIC irons have a Mo isotopic composition that is significantly 190 different from the other CC irons (ϵ^{94} Mo = 2.25 ± 0.10; 95% CI), but a Ru isotopic composition 191 that is identical within uncertainty (ϵ^{100} Ru = -1.04 ± 0.05; 2SD). Finally, a IIC iron meteorite, 192 Wiley, has an ϵ^{94} Mo = 3.42 ± 0.07 and an ϵ^{100} Ru = -1.07 ± 0.08 (2SD). Wiley has the largest Mo 193 isotope anomaly yet measured for an iron meteorite (Kruijer et al., 2017), but a similar Ru 194 isotopic composition to the other CC irons. The small measured CRE effect on ϵ^{196} Pt, along with 195 the shared ε^{100} Ru value of Wiley and other CC irons, indicates that the large Mo isotope anomaly 196 is not due to CRE. Additionally, the ε^{183} W of Wiley is distinct from IIC irons (Kruijer et al., 197 2017), and the relative abundances of its highly siderophile elements suggest that it and the IIC 198 irons crystallized from different parental melts (Tornabene et al., 2019). Therefore, it is likely 199 that Wiley originated on a different parent body, and in a different nebular reservoir, than the IIC 200 201 group.

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4. DISCUSSION

4.1. Comparison between the NC and CC reservoirs

205 *4.1.1. Mo-Ru cosmic correlation*

Dauphas et al. (2004) first identified that Mo and Ru isotope anomalies, which both reflect variable deficits in the *s*-process isotopes, are correlated in what are now known as NC irons and the IVB group. This linear relationship has been interpreted as two-component mixing between an *s*-enriched and an *s*-depleted endmember and taken as evidence that Mo and Ru are hosted in a common presolar carrier or a few similar carriers. Notably, this relationship is linear because the *s*-process endmember is isotopically very different from the bulk meteorites, such that the curved mixing line appears linear in the relevant range.

In agreement with previous work (Dauphas et al., 2004; Fischer-Gödde et al., 2015; 213 Bermingham et al., 2018), the ϵ^i Mo and ϵ^{100} Ru compositions of NC iron meteorite groups define 214 a roughly linear relationship (Figs. 2a, SM1). The slopes of the linear regressions through the NC 215 irons on plots of ε^{i} Mo vs. ε^{100} Ru are in good agreement with a theoretical *s*-process mixing line 216 (calculated as in Dauphas et al., 2004). However, when CC irons are considered together, some 217 plot well off the theoretical s-process mixing line on plots of ϵ^{92} Mo and ϵ^{94} Mo vs. ϵ^{100} Ru. 218 Primarily these are the IIC irons and Wiley, which plot to the right of the reference line. This is 219 also true when other Mo isotopes are plotted vs. ϵ^{100} Ru (Fig. SM1). It is also notable that IVB 220 irons and other CC irons with similar isotopic compositions (the IID, IIF, and IIIF groups and 221 chondrite metals; hereafter, the "CC cluster") plots slightly to the left of the reference line on 222 plots of ϵ^{92} Mo and ϵ^{94} Mo versus ϵ^{100} Ru. Cosmic ray exposure cannot explain why the IID, IIF, 223 and IIIF groups fall to the left of the reference line, as most of the irons examined here were not 224 exposed to high neutron fluence (Table 1), although this may explain why the IVB group does 225 (See SM). The different relative abundances of p- and r-process isotopes defining the NC-CC 226 dichotomy is most evident when p-process isotopes are included in plots (e.g., Fig. 1). For this 227

reason, the CC cluster falls to the left of the theoretical line on plots of ε^{92} Mo and ε^{94} Mo versus ε^{100} Ru, but when other Mo isotopes are plotted, the CC cluster falls closer to or on the theoretical *s*-process reference lines (Bermingham et al., 2018, Fig SM1). This indicates that, like the IIC irons and Wiley, the CC cluster likely deviates from the NC array and cannot be accounted for by a pure *s*-process deficit.

Most importantly, the CC meteorites collectively exhibit variable Mo isotopic 233 compositions, but uniform ϵ^{100} Ru. Thus, no single linear correlation can be regressed through all 234 the data. This non-linearity is not due to incomplete digestion of presolar phases in different 235 pieces used for Mo and Ru analyses, as is a concern for unequilibrated chondrites, because these 236 iron meteorites originated in equilibrated, differentiated parent bodies. Further, it is unlikely that 237 differentiation or other parent body processes decoupled Ru from Mo in CC irons. This is 238 because carbonaceous chondrites also have variable Mo isotopic compositions but ε^{100} Ru values 239 which cluster around -0.9, although they vary over a range of -0.3 to -1.5 ϵ^{100} Ru (Fischer-Gödde 240 et al., 2015; Fischer-Gödde and Kleine, 2017). As this isotopic variability is also seen within 241 carbonaceous chondrite groups, it is likely due to sampling effects (Fischer-Gödde and Kleine, 242 2017). For this reason, we suspect that the processes that acted on the sampling scale are 243 responsible for the entire isotopic range of carbonaceous chondrites, and that the average value 244 of -0.9 ϵ^{100} Ru is representative of bulk carbonaceous chondrites, consistent with the uniform 245 composition of iron meteorites (-1 ϵ^{100} Ru). 246

Some deviations from a single linear regression may be due to mixing of endmembers having variable Mo/Ru, which would change the curvature of the mixing line(s) (Dauphas et al., 2004). However, if this were exclusively the cause, it is surprising that these deviations are restricted to the CC suite. Moreover, the variable Mo isotopic compositions and restricted Ru isotopic compositions of the CC irons cannot be explained this way, but rather indicate that Mo and Ru were decoupled in the CC suite, either because Mo and Ru were hosted in different presolar phases from one another in the CC reservoir, and/or because processing of the presolar hosts of Mo and Ru only modified Mo (Fischer-Gödde et al., 2015). Regardless of the cause, however, it appears that the Mo-Ru correlation is not reflected in the CC irons, indicating that the nucleosynthetic heterogeneities in the NC and CC reservoirs did not originate in the same way, or under the same conditions.

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4.1.2. Comparison to ¹⁸³W isotope anomalies

To investigate what presolar carriers or conditions were different between the two 260 reservoirs, we compared the isotopic characteristics of Mo and Ru to those of W. Like Mo and 261 Ru, W exhibits nucleosynthetic heterogeneity and is siderophile, refractory, and redox sensitive. 262 Until recently, no nucleosynthetic ¹⁸³W isotope anomalies had been identified in bulk iron 263 264 meteorites, apart from the IID and IVB iron meteorites (e.g., Qin et al., 2008; Kruijer et al., 2013). Burkhardt et al. (2012b) reported W isotope anomalies in Murchison leachates, which 265 were broadly correlated with the corresponding Mo isotope anomalies. As with Mo and Ru, this 266 suggests that Mo and W may be hosted in similar presolar carriers. However, Burkhardt et al. 267 (2012b) also observed that the large Mo isotope variations among bulk meteorites are not 268 observed for W, indicating that the two isotope systems were decoupled in the precursors of bulk 269 meteorites. Notably, the only CC irons considered in that work were IVB irons. 270

The Mo and W data reported by Kruijer et al. (2017), supplemented here with new Mo data for a larger set of CC irons, show that large ϵ^{183} W isotope anomalies are observed in the CC

irons, which are correlated with ε^{i} Mo (Fig. 2b), in contrast to ε^{i} Mo vs. ε^{100} Ru. The slope of the 273 linear relationship among CC irons is in good agreement with that of the theoretical s-process 274 mixing line of Arlandini et al. (1999). Thus, ¹⁸³W nucleosynthetic anomalies in the CC suite 275 likely reflect variable deficits in the s-process W isotopes. In contrast to correlated ε^{i} Mo and 276 ϵ^{100} Ru in the NC irons, however, isotope ratios of ϵ^{i} Mo and ϵ^{183} W are not correlated in the NC 277 suite. The lack of ¹⁸³W isotope anomalies corresponding with Mo isotope anomalies among NC 278 meteorites indicates that Mo and W are decoupled in those irons, which is generally consistent 279 280 with the conclusion of Burkhardt et al. (2012b).

The observation that W shows no nucleosynthetic heterogeneity in the NC reservoir, and 281 Ru is uniform in the CC reservoir, suggests that both reservoirs were well mixed at some stage. 282 However, both reservoirs are also characterized by variable s-process deficits in Mo and either 283 Ru or W. These seemingly conflicting observations can be reconciled if it is assumed that the 284 reservoirs were initially isotopically homogeneous (with regard to the distribution of s-process 285 carriers), and the s-process variations were generated as a secondary feature of each reservoir. 286 This relies on the assumption, however, that Mo, Ru, and W were hosted in similar s-process 287 288 carriers in both reservoirs. Alternatively, the contrasting behaviors of Ru and W relative to Mo may suggest that at least two types of s-process carriers existed hosting either Mo-Ru (in the NC 289 reservoir) or Mo-W (in the CC reservoir). However, perhaps except for a hypothesized 290 component - the addition of which may have established the NC-CC dichotomy (as discussed 291 below) - there is little reason to suspect that a specific carrier was present within one reservoir 292 293 and not the other, as mixing evidently erased the large-scale variations among presolar grains in 294 bulk meteorites. It is considered more likely that both reservoirs had similar abundances of the same *s*-process carriers and that processing under distinct conditions led to the observed
differences between the Mo-Ru-W relationships in iron meteorites.

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4.2. Implications for the evolution of the NC and CC reservoirs

4.2.1. Establishment of the NC and CC reservoirs

The Mo isotope dichotomy was suggested to result from addition of r-process enriched 300 material to the CC reservoir (Budde et al., 2016; Worsham et al., 2017), or preferential 301 302 processing of *p*-process enriched carriers (Poole et al., 2017; discussed below). To account for 303 the observation that some calcium-aluminum-rich inclusions (CAIs – some of the earliest solids formed in the solar system) have Mo isotopic signatures with stronger r-process enrichments 304 than CC meteorites (Burkhardt et al., 2011; Brennecka et al., 2013), Nanne et al. (2019) 305 suggested that the early disk, which formed by rapid viscous spreading of early infalling material 306 307 from the molecular cloud, was enriched in r-process nuclides, and that CAIs formed from this 308 material. After the formation of r-enriched CAIs, the composition of the infalling material shifted to an NC (r-depleted) composition. This material mixed within the disk with the original 309 r-process enriched material until the reservoirs were physically separated, potentially by the 310 growth of Jupiter, which set the distinct compositions of the reservoirs and resulted in the CC 311 reservoir having an intermediate composition between the NC and r-enriched components 312 313 (Kruijer et al., 2017; Nanne et al., 2019).

Two-component mixing calculations between NC compositions and type B CAIs reveal that mixing between these compositions is consistent with the isotopic characteristics of most CC irons, in support of the Nanne et al. (2019) model (Fig. 3). Because the CC reservoir represents a 317 mixture of both components, the later-added r-depleted component was present in both reservoirs (though concentrated in the NC reservoir). Therefore, processing of presolar materials 318 constituting these components under the same conditions in each reservoir would not be 319 expected to generate the observed differences in, and reciprocal nature of, the Mo-Ru and Mo-W 320 relationships. To facilitate the following discussion, which is primarily concerned with the 321 production of the contrasting s-process variations within each reservoir, we will follow the model 322 323 of Nanne et al. (2019), although the interpretations here are not dependent on how the reservoirs 324 were established.

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4.2.2. Initial homogeneity within the two reservoirs and the evidence for thermal processing

The first suggestion that nucleosynthetic heterogeneity amongst bulk meteorites resulted from variable processing of presolar materials expanded on the observation that the abundances of certain presolar phases in chondrites was related to the type of chondrite and the degree of metamorphism (e.g., Huss et al., 2003). Trinquier et al. (2009) proposed this mechanism to account for the observed correlation of Ti isotopes, which are made by different nucleosynthetic processes, in bulk meteorites.

The thermal processing model assumes that the relative proportions of different presolar materials were initially the same throughout the protoplanetary disk due to turbulent mixing, resulting in an isotopically homogeneous disk (e.g., Trinquier et al., 2009). It is now known that the NC and CC reservoirs were isotopically distinct, but within each reservoir turbulent mixing may have resulted in isotopic homogeneity. This supposed homogeneity was not chemical in nature (i.e., not due to thermal processing), but was due to efficient mechanical mixing of the
dust. Mechanical mixing is supported by the observation that all chondrites have identical
nucleosynthetic Os isotope compositions, despite the evidence that isotopically distinct presolar
carriers of Os are revealed by chondrite leachates (Yokoyama et al., 2010).

Heretofore, the lack of primary nucleosynthetic anomalies in heavy elements, such as Os, 343 in bulk meteorites from either reservoir has argued for isotopic homogeneity within the disk and 344 against inherited nucleosynthetic heterogeneity (e.g., Walker et al., 2012). The counterargument 345 is that most of the isotopically homogeneous heavy elements are produced primarily by the r-346 process and may be hosted in different carriers, such that these elements may not reflect inherited 347 s-process heterogeneity. However, several lines of evidence suggest that s-process nuclides were 348 also initially homogeneously distributed within each reservoir. First, the results of this study 349 demonstrate that W and Ru, both produced in part by the s-process, were isotopically uniform in 350 one reservoir. For instance, the uniform Ru isotopic composition in the CC reservoir, combined 351 with the presence of s-process Ru variations in the NC reservoir, suggests that, like the CC 352 reservoir, the NC reservoir was isotopically homogeneous prior to the generation of the s-process 353 variability within it. This is supported by the W isotopic homogeneity in the NC reservoir. The 354 same argument can be made for initial W isotopic homogeneity in the CC reservoir. 355

Second, initial isotopic homogeneity in each reservoir is supported by the Mo isotope dichotomy, because CC and NC meteorites fall on two parallel regressions on plots of ε^{92} Mo and ε^{94} Mo versus ε^{j} Mo (Budde et al., 2019), reflecting identical *s*-process variations in each reservoir (Fig. 1). This indicates that the relative enrichments of *r*- and *p*-process isotopes to *s*-process isotopes are distinct between the two reservoirs, but approximately constant within them; otherwise, the regressions would have significantly different slopes, or there would be scatter about the regressions. Therefore, the *r*- and *p*-process component(s) in each reservoir must have been homogeneously distributed. It is difficult to envision how these constant relative enrichments of *r*- and *p*-process to *s*-process isotopes could be achieved if the *s*-process variations were pre-existing and maintained during the establishment of the two reservoirs. However, if each reservoir had distinct, but homogeneous isotopic compositions, the pure *s*process variations could easily be explained if they were generated independently after the two reservoirs were established.

Finally, the preponderance of chemically diverse irons and chondritic metals in the CC 369 cluster suggests widespread homogenization of the precursor materials in the CC reservoir. This 370 371 was first concluded by Bermingham et al. (2018), who noted that ungrouped irons originating from three chemically diverse parent bodies cluster with the IVB group on plots of ε^{i} Mo vs. 372 ϵ^{100} Ru. Including meteorites that have Mo isotopic compositions within uncertainty of the IVB 373 group, and the rare groups reported here, at least 15 parent bodies have tightly clustered Mo and 374 Ru isotopic compositions (Fig. 4). The variety and number of parent bodies, which include 375 differentiated, undifferentiated, volatile-depleted, and volatile-enriched parent bodies, suggests 376 377 that they formed over a range of heliocentric distances and over an extended period of time. This would require that a large portion of the disk had a homogeneous isotopic composition when the 378 parent bodies representing the CC cluster formed. If this homogenized region was representative 379 380 of the CC reservoir, then it implies that the isotopic composition of the CC cluster of meteorites is close to the initial composition of the CC reservoir as a whole and that chemical or thermal 381 382 processing of the precursor materials is responsible for generating more isotopically anomalous compositions (e.g., Wiley). 383

Given the isotopic homogeneity of both *r*- and *s*-process synthesized elements in both reservoirs, the parallel trends of the NC and CC suites on Mo isotope plots, and the clustered isotopic compositions of most CC irons, it is likely that both reservoirs were initially isotopically homogeneous and that processing of presolar material in both the NC and CC reservoirs generated the *s*-process nucleosynthetic heterogeneity at the bulk meteorite scale. Moreover, the contrasting behaviors of Ru and W relative to Mo in the NC and CC reservoirs likely require that processing occurred under distinct conditions in the two reservoirs.

To summarize this mechanism generally, thermal processing of dust in an isotopically 391 homogenized portion of the disk (i.e., within either the NC or the CC reservoir) may have 392 destroyed some isotopically anomalous presolar phase(s), vaporizing certain constituent elements 393 (but likely not all). The isotopically anomalous vapor would be removed from the dust, due to 394 settling, gas drag, and radial forces, leaving a complimentary isotopically anomalous residue 395 from which planetesimals could accrete. In an environment where this type of processing 396 occurred, an element could retain isotopic homogeneity if its host(s) was not affected, or if it was 397 not lost from the system. Thus, whether an element exhibits nucleosynthetic heterogeneity 398 399 depends, in part, on the durability of its presolar host(s) and on the volatility of that element. The volatility of a given element is dependent on many factors, including the redox conditions and its 400 proclivity to form volatile molecular species. For example, under certain nebular conditions, 401 thermodynamic calculations suggest that Mo and W readily form volatile oxides, whereas Ru 402 does not (Fegley and Palme, 1985). Notably, if thermal processing via vaporization occurred, 403 404 large-scale elemental fractionations would not be expected, as only small degrees of partial 405 evaporation of an element from anomalous presolar material would be necessary to create the 406 observed nucleosynthetic effects.

407 Given the results presented here, a model explaining the relative isotopic behaviors of Mo, Ru, and W must satisfy the requirements that Mo and Ru behaved similarly in the NC 408 reservoir, and Mo and W behaved similarly in the CC reservoir. This can be accomplished by 409 appealing to different redox conditions between the two reservoirs. Using 50% condensation 410 temperatures (T_C) as a proxy for relative volatilities under reducing conditions, Mo and Ru have 411 lower 50% T_C than W (1587, 1546, and 1790 K, respectively – Lodders et al., 2003). Similarly, 412 413 calculations done by Fegley and Palme (1985) show that Mo and Ru may be depleted in a W(Re, 414 Os) alloy formed via fractional condensation, whereas the complementary gas would be enriched in Mo and Ru, and depleted in W. Indeed, refractory metal nuggets (RMNs), some of which 415 416 likely represent primary condensates from the solar nebula, have, on average, lower CInormalized abundances of Ru and Mo relative to W (Berg et al., 2009; Daly et al., 2017). This 417 indicates that W may be condensed from the gas at higher temperatures, and incorporated into 418 419 RMNs condensed at those temperatures, more readily than Mo and Ru. The corollary is that, as metals, Mo and Ru may be volatilized under reducing conditions more readily than W from their 420 421 presolar hosts, and W may stay in the residue if it remains refractory during this process. By contrast, Mo and W form volatile oxides more readily than Ru (Fegley and Palme, 1985). Thus, 422 under oxidizing conditions, Mo and W can form oxides which may be volatilized from their 423 presolar hosts, whereas Ru may remain in the residue. 424

The disparate Mo-Ru-W isotope systematics in the NC and CC reservoirs can, therefore, be accounted for by thermal processing of presolar material under relatively reducing conditions in the NC reservoir, and under oxidizing conditions in the CC reservoir. This is also broadly consistent with the bulk chemistry of chondrites and iron meteorites. The CC suite includes more volatile-rich carbonaceous chondrites (although some CC iron meteorite groups are volatiledepleted), and the NC reservoir includes volatile-depleted and more reduced enstatite and
ordinary chondrites. In the case of iron meteorites, it has been suggested that the CC iron groups
have higher Ni and refractory siderophile element abundances due to the more oxidized
conditions of their core formation relative to NC irons (Rubin, 2018).

Given the chronological evidence that the NC and CC reservoirs were physically 434 separated for an extended period of time (Kruijer et al., 2017), the different locations of the two 435 reservoirs likely contributed to the prevailing thermal and redox conditions. Warren (2011) and 436 Kruijer et al. (2017) suggested that the CC and NC reservoirs were in the outer and inner solar 437 system, respectively, which is consistent with the implication of the present study that the CC 438 reservoir was generally more oxidizing than the NC reservoir. In addition to the bulk chemistry 439 of CC and NC meteorites, ratios of ${}^{15}N/{}^{14}N$ for iron meteorites support this conclusion as well. 440 Füri and Marty (2015) argued that enrichments in ¹⁵N are generally associated with the presence 441 of organics and ices, and ¹⁵N enrichments may increase with heliocentric distance (although 442 there are exceptions - e.g., Jupiter). Iron meteorites that are classified as CC irons here and 443 elsewhere are enriched in ¹⁵N (δ^{15} N ranges from +3 to +150‰), whereas irons that are classified 444 as NC meteorites are typically depleted in ${}^{15}N$ ($\delta^{15}N$ ranges from -90 to -3‰ – Prombo and 445 Clayton, 1993). Therefore, if the CC reservoir was in the outer solar system, hydration of the 446 dust or the presence of ice in the dust may have led to the more oxidizing conditions (Fegley and 447 448 Palme, 1985; Fedkin and Grossman, 2016).

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4.2.3. Model for the origin of nucleosynthetic heterogeneity

Based on the relative isotopic characteristics of Mo, Ru, and W in the NC and CC reservoirs, a simplified illustration of the thermal/chemical processing discussed above and the collateral isotopic effects among Mo, Ru, and W isotopes is presented here (Fig. 3). This illustration provides only one example, but other models are possible.

455 The initial composition of the disk is taken as that of Nanne et al. (2019) (point A in Fig. 3), which is equivalent to the composition of type B CAIs. As discussed in section 4.2.1 and 456 Nanne et al. (2019), the two reservoirs were likely established when NC-like material was mixed 457 into the disk (point B). Note that these components represent the characteristic compositions of 458 two bulk disk reservoirs and are, therefore, not hosted in any specific presolar phase (Nanne et 459 al., 2019). Moreover, the approximately constant relative abundances of r- and p-process 460 isotopes within the reservoirs indicates that the r- and p-process Mo isotopes are also not hosted 461 in specific carriers, but represent a homogenized nebular component (Dauphas et al., 2002b) in 462 each reservoir (Budde et al., 2019). 463

Prior to the accretion of most meteorite parent bodies, thermal processing in the NC 464 reservoir under relatively reducing conditions may have destroyed thermally labile, homogenized 465 nebular dust (i.e., dust with an s-depletion and a relative enrichment in r- and p-process 466 467 isotopes). Molybdenum and Ru may be preferentially volatilized from this r- and p-process 468 enriched dust and separated from the residue, whereas W isotopes may remain in the residue in solar proportions. Essentially, this processing would concentrate any more robust presolar 469 materials enriched in s-process nuclides, which would drive the Mo and Ru isotopic 470 compositions toward less s-process depleted compositions (toward the terrestrial composition). 471 In this scenario, the terrestrial composition represents the most thermally processed precursor 472

473 materials yet sampled (as was also suggested by Burkhardt et al., 2012b and Poole et al., 2017)
474 (Line C_{NC} in Fig. 3).

In the CC reservoir, the more oxidizing conditions may have resulted in the destruction of presolar carriers by oxidation, during which Mo and W formed volatile oxides and Ru stayed in the residue. To accommodate the evidence for an initially homogeneous composition of the CC reservoir near the CC cluster (Fig. 4), processing likely resulted in the loss of *s*-process isotopes from an *s*-process carrier, generating the more *s*-depleted Mo and W isotopic compositions of the IIC irons and Wiley (Line C_{CC} in Fig. 3).

New open questions include what the specific presolar phases involved in processing in 481 each reservoir were. For example, Poole et al. (2017) argued for a model that is generally in 482 agreement with that presented above and in Burkhardt et al (2012b), where processing of p-483 484 process and r-process carriers resulted in decreasing s-process deficits with greater degrees of thermal processing. However, Poole et al. (2017) advocate for this type of processing in both the 485 NC and CC reservoirs, with the exception that *p*-process isotopes were preferentially lost over *r*-486 process isotopes in CC irons due to physical differences between p- and r-process material. 487 These authors also propose that this process is what established the two reservoirs, which is 488 489 difficult to reconcile with the observation that the CC and NC reservoirs form two parallel sprocess mixing lines on ε^{j} Mo plots (Fig. 1; Budde et al., 2019). If the preferential loss of p-490 process isotopes in the CC reservoir occurred, it may have led to different slopes between the NC 491 and CC irons. 492

493 Other open questions relate to the conditions at which evaporation of Mo, Ru, and W can494 occur and the actual temperatures, pressures, and oxidation states within different regions of the

495 disk. Heating of the protoplanetary disk by irradiation and viscous friction likely resulted in temperatures ranging from 500-1500 K in the inner disk to 50-150 K in the outer disk (Boss, 496 1998). Transient heating events also evidently occurred, given that temperatures required for 497 chondrule formation are 1500-2000 K (e.g., Boss, 1998). These estimates of transient disk 498 temperatures generally compare favorably with temperatures at which Mo, Ru, and W may 499 evaporate. While likely not representative of the full temperature range at which this is possible, 500 calculations by Fegley and Palme (1985) show coupled Mo and Ru behavior at ~1675 K and 501 502 coupled Mo and W behavior between 1450 - 1650 K. In terms of redox conditions, Fegley and Palme (1985) showed that Mo and W may be coupled at H_2O/H_2 ratios of 10^{-2} to 10^{-1} , below 503 which Mo and Ru may be coupled (the solar H_2O/H_2 ratio is 5×10^{-4}). Environments in which 504 these conditions may be met include localized regions of dust/ice enrichment, especially if the 505 dust is hydrated (e.g., Fegley and Palme, 1985; Fedkin and Grossman, 2016). 506

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508	5.	CONCLUSIONS
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Combined Ru, Mo, and W isotope data for iron meteorites, including the first high-509 precision mass-independent Ru isotope data from the rare iron meteorite groups IIC, IIF, IIIE, 510 and IIIF, reveal a distinct genetic heritage of CC and NC meteorites. This work, along with the 511 work of Fisher-Gödde et al. (2015), Fischer-Gödde and Kleine (2017), and Bermingham et al. 512 (2018) shows that CC iron meteorites, and potentially bulk carbonaceous chondrites, are likely 513 restricted to an ϵ^{100} Ru of -1. When considered together, these data reveal decoupled Mo-Ru 514 515 isotope systematics in the CC reservoir, in contrast to the coupled Mo-Ru systematics in the NC reservoir. Conversely, W and Mo are correlated in the CC suite, but not the NC suite. This new 516 observation of the contrasting behaviors of Ru and W, relative to Mo, in the two reservoirs 517

518 allows for constraining the distinct mechanisms and physical conditions under which 519 nucleosynthetic heterogeneity was generated in the protoplanetary disk. The data presented here not only provide further evidence that thermal/chemical processing of presolar phases generated 520 s-process nucleosynthetic heterogeneity, but it also demonstrates that the heterogeneity within 521 the CC and NC reservoirs evolved under distinct redox conditions. Specifically, the prevailing 522 conditions in the NC reservoir were likely more reducing than those in the more oxidized CC 523 524 reservoir, consistent with the inferred location of these reservoirs inside and outside the orbit of 525 Jupiter, respectively.

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4 5 Fig. 1. ϵ^{94} Mo vs. ϵ^{95} Mo, illustrating the dichotomy between NC (in red) and CC (in blue) meteorites (Budde et al. 2016; Poole et al., 2017; Worsham et al., 2017). Symbols outlined in 6 black denote rare iron meteorite groups examined in this work (data from this study and Kruijer 7 et al., 2017). Members from both suites fall along theoretical s-process mixing lines between an 8 9 s-process component and an s-process depleted component (Dauphas et al., 2004, Lugaro et al., 2003). The mixing lines for both suites are offset from the origin to align with the data. Other 10 literature data are from Burkhardt et al. (2011), Render et al. (2017), Poole et al. (2017), and 11 Bermingham et al. (2018). The offset between these lines cannot be accounted for by s-process 12 13 variability but appears to reflect a relative enrichment of r- (and p-) process isotopes in the CC 14 reservoir (Budde et al., 2016; Poole et al., 2017; Worsham et al., 2017). 15





Fig. 2. ϵ^{94} Mo vs. ϵ^{100} Ru (a) and ϵ^{94} Mo vs. ϵ^{183} W (b) for various meteorite groups. Symbols 18 outlined in black denote data for rare iron meteorite groups from this work. NC and CC 19 meteorite groups are shown in red and blue, respectively. ε^{183} W data are from Kruijer et al. 20 (2017) and Kruijer et al. (2014a) and, for the rare iron groups, were obtained from the same 21 sample set as Ru and Mo. Other literature data are from Chen et al. (2010), Burkhardt et al. 22 (2011), Fischer-Gödde et al. (2015), Fischer-Gödde and Kleine (2017), Render et al. (2017), 23 24 Poole et al. (2017), Worsham et al. (2017), and Bermingham et al. (2018). Also shown is an s-25 process mixing line, as in Fig. 1 (Dauphas et al., 2004; calculated using the same curvature coefficient used by those authors, which were estimated from Arlandini et al., 1999, and SiC 26 compositions reported by Lugaro et al., 2003 and Savina et al., 2004). In (b) the s-process 27 composition from Arlandini et al. (1999) was used to calculate the line, which is offset from the 28 29 origin to align with the CC irons.









Fig. 4. Mo isotope data for diverse meteorites and meteorite groups having similar compositions
to the IVB, IID, IIF, and IIIF iron meteorite groups. Data are from Dauphas et al., 2002a (Grand
Rapids, Eagle Station), Burkhardt et al., 2011 (Mbosi, Tafassasset, and Gujba), Burkhardt et al.,
2014 (CK), Worsham et al., 2017 (Sombrerete), Bermingham et al., 2018 (Chinga, Dronino,
Tishomingo), and Hilton et al., 2019 (South Byron Trio). Samples where Ru isotope data have

been reported are also within uncertainty of the Ru isotopic composition of the IVB group.

Meteorite	Collection (No.)	Ν	ϵ^{192} Pt ^a	±	ϵ^{194} Pt	±	ε ¹⁹⁶ Pt	±
IC								
Chihuahua City ^b	BM 1959,1011	1	0.50	1.30	0.17	0.11	0.07	0.07
Arispe ^b	Münster	6	13.33	0.27	0.52	0.03	0.35	0.03
Arispe (replicate) ^b	Münster	6	12.89	0.36	0.48	0.04	0.32	0.04
Arispe (replicate) ^b	ME 1011	3	13.69	1.30	0.67	0.11	0.42	0.07
Bendego	ME 6	4	0.62	1.00	0.37	0.06	0.49	0.06
Bendego (replicate) ^b	ME 6	3	1.08	1.30	0.32	0.11	0.47	0.07
Bendego (replicate) ^b	USNM #351	2	-0.57	1.30	0.38	0.11	0.52	0.07
Mount Dooling	USNM 5713	5	-0.39	0.98	0.07	0.05	0.00	0.03
Murnpeowie ^b	BM 2005, M179	2	3.95	1.30	0.50	0.11	0.38	0.07
Murnpeowie (replicate) ^b	BM 2005, M179	4	2.34	1.05	0.29	0.11	0.26	0.05
пс								
Kumerina	BM 1938,220	4	0.52	0.89	-0.02	0.05	-0.09	0.03
Kumerina (replicate) ^b	BM 1938,220	2	0.80	1.30	0.17	0.11	0.04	0.07
Kumerina (replicate) ^b	BM 1938,220	3	-0.02	1.30	0.04	0.11	-0.02	0.07
Perryville	USNM 428	5	2.14	1.12	0.16	0.05	0.05	0.05
D W b	Munster	2	5.20	0.60	0.18	0.04	0.12	0.02
Ballinoo	ME 980	3	-0.37	1.30	0.08	0.11	-0.01	0.07
Wiley	BM 1959, 914	5	0.97	0.45	0.04	0.07	0.01	0.02
Wiley (replicate) ^b	BM 1959, 914	4	0.55	1.34	0.12	0.05	0.05	0.07
Wiley (replicate) ^b	BM 1959, 914	5	0.58	0.65	0.11	0.06	0.08	0.06
IID	2	U	0.00	0.00	0111	0.00	0.00	0.00
Bridgewater ^b	ME 1895	5	0.80	0.90	0.02	0.08	-0.01	0.02
N'kandhla ^b	BM 1921, 17	5	0.64	0.23	0.03	0.05	0.01	0.05
IIF	,							
Monahans ^b	BM 1959,910	4	0.91	0.90	0.15	0.05	0.07	0.04
Monahans (replicate) ^b	BM 1959,911	2	1.43	1.30	0.09	0.11	0.08	0.07
Del Rio	USNM 6524	5	2.37	1.04	0.05	0.06	0.04	0.03
ше								
Willow Creek ^b	Münster	1	0.55	1.30	0.14	0.11	0.13	0.07
Kokstad ^b	ME 1015	1	1.62	1.30	0.38	0.11	0.35	0.07
Kokstad (replicate) ^b	ME 1015	2	0.82	1.30	0.29	0.11	0.26	0.07
Colonia Obreira ^b	ME 2871	1	0.17	1.30	0.06	0.11	-0.01	0.07
Colonia Obreira (replicate) ^b	ME 2871	1	-0.30	1.30	0.08	0.11	0.03	0.07
Staunton ^b	BM 1955,M239	1	-0.17	1.30	0.15	0.11	-0.07	0.07
Staunton (replicate) ^b	BM 1955,M239	2	-0.28	1.30	0.08	0.11	0.05	0.07
Paneth's iron ^b	BM 2005,M199	2	0.26	1.30	0.12	0.11	0.09	0.07
Burlington	USNM 978	3	0.17	1.30	0.18	0.11	0.07	0.07
Coopertown	USNM 1003	4	0.39	1.28	0.11	0.05	0.02	0.06
IIIF								
Klamath Falls ^b	ME 2789	1	0.39	1.30	0.06	0.11	0.06	0.07
Klamath Falls (replicate) ^b	ME 2789	1	0.99	1.30	0.13	0.11	-0.01	0.07
Clark County ^b	BM 1959,949	4	2.81	0.78	0.08	0.06	0.03	0.07
Clark County (replicate) ^b	BM 1959,949	2	3.54	1.30	0.17	0.11	0.10	0.07
Oakley	USNM 780	4	18.93	3.33	0.78	0.09	0.48	0.06

Table 1. Platinum isotope data used to monitor CRE effects.

^aData are reported in epsilon notation [($R_{sample}/R_{standard}$ -1) × 10,000], and normalized to ¹⁹⁸Pt/¹⁹⁵Pt = 0.2145. For the number of analyses of the same solution N < 4, the uncertainties are the 2SD of repeated analyses of solution standards. For N ≥ 4, uncertainties are the 95% confidence interval of the mean, according to (SD × $t_{0.95,N-1}$)/√N. ^bData from Kruijer et al. (2017)

Table 2. Ruthenium isotope data for rare iron meteorite groups. The data are not corrected for CRE exposure. Where groups include irons with ϵ^{196} Pt ≥ 0.13 , a low-exposure mean excluding those irons is given. The excluded samples are denoted with an asterisk.

Meteorite	Collection (No.)	Ν	ϵ^{100} Ru ^a	±
IC				
Chihuahua City	BM 1959,1011	5	-0.38	0.07
Arispe*	ME 1011	6	-0.24	0.05
Bendego*	ME 6	5	-0.16	0.10
Bendego (replicate)*	ME 6	2	-0.23	0.13
Mount Dooling	USNM 5713	5	-0.37	0.08
IC Average			-0.27	0.15
Low exposure IC Avera	age		-0.38	0.13
IIC				
Kumerina	BM 1938,220	5	-1.07	0.14
Perryville	USNM 428	5	-1.01	0.05
Unter-Mässing	Münster	5	-1.03	0.08
IIC Average			-1.04	0.05
Wiley	BM 1959, 914	6	-1.10	0.10
Wiley (replicate)	BM 1959, 914	5	-1.04	0.13
Wiley Average			-1.07	0.08
IID				
Bridgewater	ME 1895	5	-1.07	0.11
N'kandhla	BM 1921, 17	4	-1.02	0.13
IID Average			-1.04	0.13
IIF				
Monahans	BM 1959,910	6	-1.01	0.07
Del Rio	USNM 6524	6	-0.97	0.10
IIF Average			-0.99	0.13
IIIE				
Willow Creek	Münster	5	-0.43	0.17
Paneth's Iron	BM 2005,M199	2	-0.53	0.13
Burlington	USNM 978	4	-0.53	0.07
Coopertown	USNM 1003	5	-0.54	0.20
IIIE Average			-0.51	0.09
IIIF				
Klamath Falls	ME 2789	2	-1.06	0.13
Clark County	Münster	6	-0.93	0.04
Oakley*	USNM 780	5	-0.80	0.03
IIIF Average			-0.93	0.26
Low exposure IIIF Ave	rage		-0.99	0.13

^aData are normalized to ⁹⁹Ru/¹⁰¹Ru = 0.7450754 (Chen et al., 2010). Uncertainties for individual samples are as in Table 1. Uncertainties for group means are the 2SD of standards (N < 3) or samples (N = 3), or the 95% confidence interval (N \geq 4). For Wiley, the uncertainty is the 2SD of duplicate measurements.

Table 3. Molybdenum isotope data for rare iron meteorite groups. The data are not corrected for CRE exposure. Where irons with ϵ^{196} Pt ≥ 0.13 were not included in the group mean, a low-exposure mean is given. The samples characterized by high exposure are denoted with an asterisk.

Meteorite	Collection (No.)	Ν	$\epsilon^{92} Mo^a$	±	ε ⁹⁴ Mo	±	ε ⁹⁵ Mo	±	ε ⁹⁷ Mo	±	ϵ^{100} Mo	±
IC												
Chihuahua City ^b	BM 1959,1011	8	0.96	0.12	0.86	0.08	0.34	0.07	0.20	0.08	0.27	0.13
Arispe*	ME 1011	5	0.77	0.20	0.75	0.14	0.21	0.10	0.14	0.07	0.27	0.07
Bendego*	ME 6	5	0.83	0.07	0.83	0.13	0.26	0.06	0.23	0.11	0.31	0.18
Mount Dooling	USNM 5713	4	0.80	0.18	0.80	0.13	0.38	0.08	0.26	0.04	0.16	0.13
Murnpeowie ^b *	BM 2005, M179	8	1.16	0.20	1.11	0.20	0.41	0.05	0.27	0.05	0.37	0.08
IC Average			0.90	0.20	0.83	0.18	0.36	0.10	0.22	0.06	0.27	0.09
Low exposure IC aver	age		0.88	0.39	0.83	0.28	0.36	0.20	0.23	0.14	0.21	0.23
IIC												
Kumerina ^b	BM 1938,220	8	2.91	0.28	2.34	0.18	1.50	0.08	0.79	0.10	0.92	0.09
Kumerina (replicate)	BM 1938,220	5	2.90	0.20	2.27	0.15	1.59	0.08	0.80	0.08	0.85	0.13
Perryville	USNM 428	6	2.89	0.13	2.27	0.11	1.59	0.07	0.83	0.03	0.85	0.08
Ballinoo ^b	ME 980	8	2.76	0.13	2.19	0.10	1.60	0.09	0.89	0.09	1.01	0.10
Unter-Mässing	Münster	5	2.87	0.36	2.19	0.27	1.54	0.13	0.83	0.09	0.99	0.23
IIC Average			2.87	0.10	2.25	0.10	1.56	0.07	0.83	0.07	0.93	0.12
Wiley ^b	BM 1959, 914	8	4.14	0.22	3.39	0.13	2.19	0.11	1.19	0.11	1.54	0.14
Wiley (replicate)	BM 1959, 914	3	4.28	0.39	3.45	0.28	2.24	0.20	1.26	0.14	1.45	0.23
Wiley (replicate)	BM 1959, 914	5	4.36	0.28	3.44	0.21	2.27	0.06	1.21	0.08	1.43	0.08
Wiley Average			4.26	0.22	3.42	0.07	2.23	0.08	1.22	0.07	1.47	0.12
IID												
Bridgewater ^b	ME 1895	7	1.63	0.10	1.16	0.16	0.96	0.15	0.51	0.12	0.67	0.17
N'kandhla	BM 1921, 17	5	1.71	0.15	1.20	0.14	1.02	0.03	0.50	0.03	0.59	0.07
IID Average			1.67	0.39	1.18	0.28	0.99	0.20	0.51	0.14	0.63	0.23
IIF												
Monahans ^b	BM 1959,910	8	1.50	0.21	1.11	0.13	0.94	0.08	0.50	0.08	0.63	0.13
Del Rio	USNM 6524	4	1.54	0.22	1.09	0.10	0.97	0.06	0.52	0.01	0.61	0.10
IIF Average			1.52	0.39	1.10	0.28	0.96	0.20	0.51	0.14	0.62	0.23
IIIE		_										
Willow Creek	Münster	5	1.10	0.30	0.94	0.23	0.41	0.11	0.24	0.10	0.28	0.17
Kokstad ¹ *	ME 1015	8	0.98	0.17	0.86	0.14	0.33	0.13	0.26	0.09	0.28	0.08
Colonia Obreira ^b	ME 2871	8	1.03	1.36	0.97	0.16	0.35	0.09	0.25	0.14	0.35	0.12
Staunton ^b	BM 1955,M239	8	1.02	0.16	0.95	0.10	0.41	0.11	0.30	0.08	0.39	0.11
Paneth's iron ^b	BM 2005,M199	8	1.09	0.15	0.93	0.20	0.42	0.07	0.35	0.05	0.37	0.12
Burlington	USNM 978	4	1.06	0.26	0.95	0.11	0.52	0.11	0.34	0.07	0.16	0.02
Coopertown	USNM 1003	4	0.98	0.32	0.91	0.28	0.51	0.10	0.29	0.06	0.16	0.15
IIIE Average			1.04	0.04	0.93	0.03	0.42	0.07	0.29	0.04	0.28	0.09
Low exposure IIIE ave	erage		1.05	0.05	0.94	0.02	0.44	0.07	0.29	0.05	0.28	0.11
IIIF												
Klamath Falls ^b	ME 2789	8	1.70	0.18	1.20	0.18	0.98	0.06	0.56	0.11	0.62	0.09
Clark County ^b	BM 1959,949	6	1.45	0.23	1.20	0.17	1.00	0.06	0.54	0.04	0.59	0.23
Oakley*	USNM 780	6	1.31	0.12	1.01	0.09	0.83	0.13	0.54	0.11	0.56	0.07
IIIF Average			1.48	0.39	1.13	0.22	0.94	0.19	0.55	0.02	0.59	0.06
Low exposure IIIF ave	erage		1.57	0.39	1.20	0.28	0.99	0.20	0.55	0.14	0.61	0.23

^aData are normalized to ${}^{98}Mo/{}^{96}Mo = 1.453173$ (Lu and Masuda, 1994). Uncertainties are as in Tables 1 and 2. ^bData from Kruijer et al. (2017)