DISTINCT EVOLUTION OF THE CARBONACEOUS AND NON-CARBONACEOUS RESERVOIRS: INSIGHTS FROM Ru, Mo, AND W ISOTOPES. E.A. Worsham[1], C. Burkhardt[1], G. Budde[1], M. Fischer-Gödde[1,2], T. S. Kruijer[1,3] and T. Kleine[1], [1]University of Münster, Institut für Planetologie, Wilhelm-Klemm-Str. 10, 48149 Münster, Germany (worsham@uni-muenster.de), [2]University of Köln, Institut für Geologie und Mineralogie, Köln, Germany, [3]Lawrence Livermore National Laboratory, Livermore, CA, USA.

Introduction: Nucleosynthetic isotope anomalies at the bulk meteorite scale reflect the heterogeneous distribution of isotopically diverse presolar materials in the early solar nebula, and have been identified for a variety of elements, including Mo, Ru, and W [1-5]. The origin of this heterogeneous distribution remains unclear, but may be related to inhomogeneous mixing and/or thermal processing of presolar materials [e.g., 6-7].

Molybdenum, Ru, and W are created by a combination of *p*-process, *s*-process, and *r*-process nucleosynthesis, and, therefore, these elements are ideal tracers of the relative proportions of diverse presolar carriers in solar system materials. Recent work has identified a dichotomy of Mo isotopic compositions between “carbonaceous” (CC) and “non-carbonaceous” (NC) meteorites caused by an excess of *r*-process Mo to the CC formation region [8].

To elucidate the conditions and processes responsible for the generation of nucleosynthetic isotopic variations among different nebular reservoirs we make use of the distinct physicochemical behaviors of Mo, Ru, and W under different nebular conditions. We obtained Ru isotopic data for magmatic iron meteorite groups belonging to both the NC (IC and IIIE) and CC suites (IIC, IID, IIF, and IIIF). In conjunction with Mo and W data, the Ru data provide new constraints on the processes leading to the distinct isotope heterogeneities in the NC and CC reservoirs, and the environmental conditions under which these heterogeneities were produced.

Experimental Methods: Much of the Mo data used here, along with the W data, are from [9]. The same iron meteorite samples were used to obtain Ru and additional Mo isotope data. The digestion and chromatography methods for Mo and Ru analyses are similar to those of [2, 4]. Platinum isotopes were used to correct the effects of cosmic ray exposure (CRE), which can modify the Mo, Ru, and W isotopic compositions. Platinum isotope compositions were determined for adjacent pieces from each iron meteorite [9, this study]. The digestion and chromatographic methods for Pt are described in [10].

Isotopic compositions of Mo, Ru, W, and Pt were determined using a *Thermo-Fisher Neptune Plus* MC-ICP-MS at Münster. The Mo, Ru, W, and Pt isotopic compositions are reported in ε notation (parts-per-10^4 deviations from terrestrial standards) and normalized to ^{98}Mo/^{96}Mo, ^{99}Ru/^{101}Ru, ^{186}W/^{184}W, and ^{198}Pt/^{195}Pt.

Results: The new ε^{94}Mo and ε^{100}Ru isotopic compositions of IC, IIC, IID, IIF, IIIE, and IIIF irons are shown in Fig. 1a, supplemented with Mo data from [9]. Mo and W data from [9] are shown in Fig. 1b. The Ru, Mo, and W isotopic compositions are corrected for CRE where necessary.

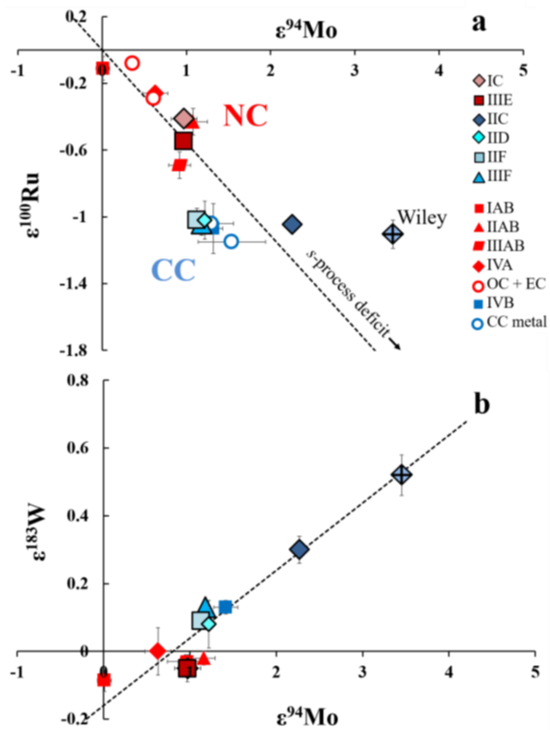

Fig. 1. ε^{94}Mo vs. ε^{100}Ru (a) and ε^{94}Mo vs. ε^{183}W (b). The new data are shown with symbols outlined in black. NC meteorite groups are shown in red, and CC groups are in blue. Other Mo and Ru data are from [e.g., 2, 4]. The dotted lines are theoretical *s*-process mixing lines [11, 12].

Also plotted is a theoretical mixing line between an endmember depleted in a pure *s*-process component and an endmember enriched in a pure *s*-process component [7, 11]. Iron meteorites from groups IC and IIIE have similar Mo and Ru isotopic compositions to IIAB iron meteorites (ε^{94}Mo ~ 1.0; ε^{100}Ru ~ -0.5). These meteorites generally plot along the *s*-process mixing line. Iron meteorites from groups IID, IIF, and IIIF have Mo and Ru isotopic compositions similar to IVB iron meteorites and some carbonaceous chondrite metals (ε^{94}Mo ~ 1.3; ε^{100}Ru ~ -1.0). This cluster of meteorites plots to the left of the mixing line on Fig. 1a. Finally, a IIC iron meteorite, Wiley, has an ε^{94}Mo = 3.45 ± 0.07 and an ε^{100}Ru

= -1.10 ± 0.08 (2SD of 3 and 2 full replicates for Mo and Ru, respectively). This meteorite plots considerably to the right of the mixing line on Fig. 1a.

Discussion: The ε^{i}Mo and ε^{100}Ru compositions of NC iron meteorite groups define a roughly linear relationship, in agreement with [4, 11]. However, most CC irons, particularly the IIC irons and Wiley, plot slightly off the trend defined by the NC meteorites. Moreover, the CC meteorites collectively exhibit variable Mo isotopic compositions, but more restricted ε^{100}Ru compositions. Thus, no single linear correlation can be regressed through all the data. This is not due to incomplete digestion of presolar phases in different pieces used for Mo and Ru analyses, as is a concern for unequilibrated chondrites, because these iron meteorites originated in differentiated parent bodies.

Dauphas et al. 2004 [11] first identified the linear relationship of what are now known as NC irons. This relationship has been taken as evidence that Mo and Ru are hosted in a common presolar carrier or a few similar carriers. As such, some of the deviations from a single linear regression may be due to mixing of this carrier with endmembers having variable Mo/Ru. However, if this were exclusively the cause, it is surprising that these deviations are largely restricted to the CC suite. Moreover, the variable Mo isotopic compositions and restricted Ru isotopic compositions of the CC irons cannot be explained in this manner, but rather indicate that either Mo and Ru were hosted in different presolar phases in the CC reservoir, and/or that processing of the presolar hosts of Mo and Ru only modified Mo. 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.

To investigate what presolar carriers or conditions were different between the two reservoirs we compare the isotopic characteristics of Mo and Ru to those of W. Like Mo and Ru, W exhibits nucleosynthetic heterogeneity, and is siderophile, refractory, and redox sensitive. In contrast to correlated ε^{i}Mo and ε^{100}Ru in the NC irons, isotope ratios of ε^{i}Mo and ε^{183}W are not correlated in the NC suite. In CC irons, ε^{i}Mo and ε^{100}Ru are not correlated, but ε^{i}Mo and ε^{183}W are. As W shows no nucleosynthetic heterogeneity in the NC reservoir, and Ru is uniform in the CC reservoir, it is likely that both reservoirs were initially well mixed. Thus, the contrasting behaviors of Ru and W relative to Mo in the two reservoirs likely require processing of the presolar carriers under distinct conditions. Therefore, the requirements for a new model explaining the relative isotopic behaviors of Mo, Ru, and W are that processing occurred in the NC reservoir under conditions where Mo and Ru behaved similarly, and in the CC reservoir under conditions where Mo and W behaved similarly (Fig. 2). This can be accomplished by invoking thermal processing in the NC reservoir under reducing conditions. In these conditions Mo and Ru are more volatile than W [e.g., 13]. During processing of an initially homogenized solar nebula, isotopically anomalous Mo and Ru may have been volatilized with the destruction of thermally labile presolar carriers, whereas W isotopes may remain in the nebular residue in solar proportions. Likewise, under oxidizing conditions, Mo and W form volatile oxides more readily than Ru [14]. Thus, in the CC reservoir, oxidative processing would result in the loss of isotopically anomalous Mo and W from the nebular residue containing unchanged Ru.

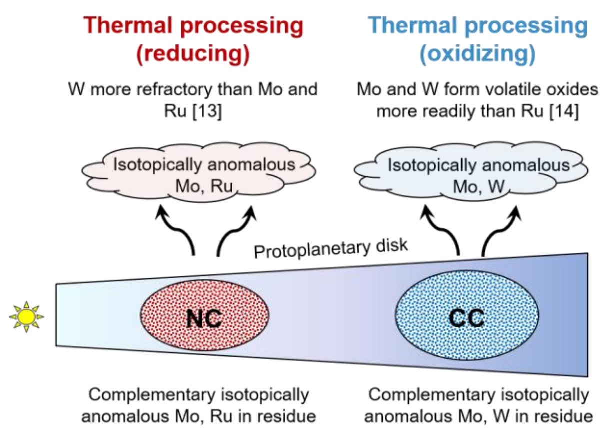

Fig. 2. Schematic of different processing in the NC and CC reservoirs.

The relative behaviors of Mo, Ru, and W imply that the NC reservoir was more reducing than the more oxidized CC reservoir, potentially due to the different ratios of ice, dust, and gas. This is consistent with the conclusions of [9], that the two reservoirs were separated by the growth of Jupiter, with the CC reservoir in the outer solar system, and the NC reservoir in the inner. This work further emphasizes that the NC and CC meteorites likely originated from spatially separated reservoirs that evolved under different thermal and redox conditions.

References: [1] Dauphas N. et al. 2002. *Astrophys. J.* 565: 640-644 [2] Burkhardt C. et al. 2011. *EPSL* 312: 390-400 [3] Chen J.H. et al. 2010. *GCA* 74: 3851-3862 [4] Fischer-Gödde M. et al. 2015. *GCA* 168: 151-171 [5] Qin L. et al. 2008. *Astrophys. J.* 674: 1234-1241 [6] Andreasen R. and Sharma M. 2007. *Astrophys. J.* 665: 874-883 [7] Trinquier A. et al. 2009. *Science* 324: 374-376 [8] Budde G. et al., 2016. *EPSL* 454, 293-303 [9] Kruijer T. et al. 2017 *PNAS* 114: 6712-6716 [10] Kruijer et al., 2013. *EPSL* 361, 162-172 [11] Dauphas N. et al., 2004. *EPSL* 226: 465-475 [12] Arlandini C. et al., 1999. *Astrophys. J.* 525, 886-900 [13] Lodders K. 2003. *Astrophys. J.* 591: 1220-1247 [14] Fegley B. & Palme H. 1985 *EPSL* 72: 311-326.