Supplementary Material

Heterogeneous accretion of Earth inferred Mo-Ru isotope systematics

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The supplementary material includes additional information about the reproducibility of the Ru isotope analyses (Table S1), additional evaluation and comparison of best-fit slopes and x-axis intercepts inferred from different datasets of the Mo-Ru correlation in NC meteorites (Table S2 and S3), and a summary of the published Mo and Ru isotope data used for plots of this study (Table S4).

S1. Precision and accuracy of the Ru isotope data

The precision and accuracy of the chemical separation and measurement protocol of Ru was evaluated by digestions of seven subsamples of a ~100 g powder of the Allende CV3 chondrite ('Allende MS-A') and three digestions of subsamples cut from a larger piece of the IIIAB iron meteorite Henbury that were also analyzed by prior studies (Fischer-Gödde et al., 2015). The replicate analyses of Allende (MS-A) and Henbury are in excellent agreement with previously published data by Fischer-Gödde et al. (2015) (Table S1). The reproducibility of ε^{100}Ru of the seven Allende Carius tube digestion is ±0.17 (2 s.d.). Considering that Carius tube digestion of primitive chondrites can lead to varying nucleosynthetic anomalies due to incomplete digestion the analytical reproducibility of the measurements of ε^{100}Ru can be expected to be smaller (Table S1). A similar reproducibility of ±0.11 (2 s.d.) is observed for the ε^{100}Ru of the three Henbury replicates. The external reproducibility of ±0.13 (2 s.d.) on ε^{100}Ru reported by Fischer-Gödde et al. (2015) was determined using the same measurement protocol for replicate measurements (n=84) of 18 Ru doped reference materials processed by micro-distillation techniques. The similarity of the reproducibility for materials processed with different techniques shows that the chemical separation used in this study does not compromise measurements of the ε^{100}Ru values. Of note, the reproducibility of ε^{96}Ru, ε^{98}Ru, and ε^{104}Ru of the Henbury and Allende digestions is larger than the external reproducibility reported by Fischer-Gödde et al. (2015). These differences may be attributed to the less efficient removal of Zr and other matrix elements of the chemical procedure used in this study compared to distillation technique of the previous study. However, nucleosynthetic isotope anomalies produced by *s*-process variations are largest on ε^{100}Ru, hence, the analytical protocol of this study allows precise determination of *s*-processes variability on ε^{100}Ru.

S2. Supplementary tables

Table S1. Comparison of the Ru isotope compositions of Allende and Henbury processed by the protocol of this study and previous published data.

Sample	Weight (g)	N[a]	ε^{96}Ru[b]	ε^{98}Ru[b]	ε^{100}Ru[b]	ε^{102}Ru[b]	ε^{104}Ru[b]
This study							
Henbury (IIIAB iron)	0.125	2	0.89 ± 0.45	0.72 ± 0.52	-0.65 ± 0.13	-0.16 ± 0.15	0.57 ± 0.31
Replicate	0.135	4	0.98 ± 0.38	0.51 ± 0.43	-0.52 ± 0.09	-0.11 ± 0.06	0.33 ± 0.19
Replicate	0.127	3	1.02 ± 0.45	0.99 ± 0.52	-0.58 ± 0.13	-0.20 ± 0.15	0.23 ± 0.31
Mean (± 2 s.d.)		(3)	0.96 ± 0.13	0.74 ± 0.48	-0.58 ± 0.11	-0.16 ± 0.07	0.38 ± 0.28
Allende MS-A	0.501	3	0.33 ± 0.45	-0.51 ± 0.52	-1.41 ± 0.13	-0.44 ± 0.15	0.40 ± 0.31
Replicate	0.505	4	1.14 ± 0.19	0.19 ± 0.49	-1.34 ± 0.17	-0.50 ± 0.29	0.37 ± 0.31
Replicate	0.501	2	0.23 ± 0.45	0.65 ± 0.52	-1.39 ± 0.13	-0.62 ± 0.15	0.16 ± 0.31
Replicate	0.503	3	-0.29 ± 0.45	1.39 ± 0.52	-1.28 ± 0.13	-0.52 ± 0.15	0.01 ± 0.31
Replicate	0.503	3	0.22 ± 0.45	0.13 ± 0.52	-1.29 ± 0.13	-0.59 ± 0.15	-0.27 ± 0.31
Replicate	0.252	1	0.21 ± 0.45	0.56 ± 0.52	-1.49 ± 0.13	-0.62 ± 0.15	0.35 ± 0.31
Replicate	0.504	3	0.98 ± 0.45	1.49 ± 0.52	-1.23 ± 0.13	-0.71 ± 0.15	0.04 ± 0.31
Mean (± 2 s.d.)		(7)	0.40 ± 0.98	0.56 ± 1.31	-1.35 ± 0.17	-0.57 ± 0.17	0.15 ± 0.45
literature data[c]							
Ru-doped reference samples		84	0.06 ± 0.45	0.02 ± 0.52	-0.01 ± 0.13	0.02 ± 0.15	0.08 ± 0.31
Henbury (IIIAB iron)		5	0.90 ± 0.15	0.53 ± 0.42	-0.62 ± 0.04	-0.17 ± 0.05	0.39 ± 0.06
Allende MS-A (CT)[d]		2	0.38 ± 0.45	-0.16 ± 0.52	-1.32 ± 0.13	-0.55 ± 0.15	0.11 ± 0.31
Replicate (CT)[d]		2	0.34 ± 0.45	0.57 ± 0.52	-1.41 ± 0.13	-0.62 ± 0.15	0.00 ± 0.31
Allende MS-A (AF)[e]		9	0.57 ± 0.19	0.52 ± 0.21	-1.01 ± 0.05	-0.38 ± 0.05	0.19 ± 0.10

[a] Number of analyses of the sample solution.

[b] Ruthenium isotope ratios are internally normalized to ^{99}Ru/^{101}Ru = 0.7450754. Given uncertainties represent the external reproducibility (2 s.d.) or the 2 s.d. of the sample analyses (if N>3).

[c] Data of Fischer-Gödde et al. (2015).

[d] Samples processed by Carius tube digestion. Of note, for primitive chondrites (e.g., CV3) Carius tube digestion is incomplete, which can result in varying and apparently higher nucleosynthetic isotope anomalies compared to complete digestion methods (e.g., alkaline fusion). Hence, the external reproducibility (2 s.d. = 0.17) of the seven Allende digestions of this study incorporate possible variations induced by incomplete digestion. Our results are in very good agreement with literature data of the same powder digested by Carius tube.

[e] Samples processed by alkaline fusion described in Fischer-Gödde et al. (2015). A difference in the nucleosynthetic isotope anomalies between Carius tube digested and alkaline fusion digested Allende powder aliquots suggests incomplete digestion for primitive chondrites with presolar phases in Carius tubes.

Table S2: Ruthenium and Mo isotope data of non-carbonaceous meteorites used for the regression calculations (Table S3).

Meteorites	**ε^{100}Ru[a]**	**95% c.i.**	**ε^{92}Mo[b]**	**2 s.d.**	**ε^{94}Mo[b]**	**2 s.d.**	**ε^{95}Mo[b]**	**2 s.d.**	**ε^{97}Mo[b]**	**2 s.d.**	**ε^{100}Mo[b]**	**2 s.d.**
Mesosiderites average	-0.42	0.02	1.21	0.14	1.04	0.08	0.46	0.05	0.25	0.03	0.19	0.04
Acapulcoite-Lodranites average	-0.33	0.05	1.05	0.11	0.92	0.07	0.48	0.03	0.23	0.02	0.24	0.07
Winonaites (HaH 193)	-0.06	0.10	0.36	0.31	0.25	0.15	0.09	0.09	0.02	0.02	0.03	0.13
Aubrites (Norton County)	-0.06	0.03	0.57	0.14	0.47	0.05	0.26	0.06	0.19	0.03	0.17	0.11
Enstatite achondrite (NWA 2526)	-0.08	0.15	0.77	0.14	0.60	0.13	0.39	0.14	0.21	0.05	0.02	0.10
Ungrouped achondrites												
NWA 6112	-0.46	0.07	1.83	0.35	1.55	0.22	0.79	0.15	0.49	0.15	0.51	0.22
NWA 1058	-0.40	0.07	1.61	0.12	1.31	0.11	0.68	0.09	0.38	0.10	0.40	0.08
NWA 5363	-0.34	0.13	0.72	0.32	0.66	0.22	0.24	0.15	0.11	0.15	0.30	0.22
Ureilites												
Dho 1519	-0.23	0.15	0.64	0.19	0.61	0.14	0.32	0.10	0.22	0.08	0.03	0.12
NWA 7630	-0.31	0.15	1.06	0.35	1.01	0.22	0.44	0.15	0.26	0.15	0.26	0.22
Leachate												
NWA 2458 L2	-1.71	0.03	3.90	0.20	3.18	0.08	1.87	0.09	0.98	0.05	1.28	0.07
Literature data[c]												
OC	-0.29	0.03	0.66	0.08	0.61	0.06	0.25	0.06	0.11	0.05	0.14	0.08
EC	-0.08	0.04	0.38	0.06	0.36	0.05	0.14	0.03	0.09	0.02	0.14	0.03
IC	-0.38	0.13	0.88	0.39	0.83	0.28	0.36	0.20	0.23	0.14	0.21	0.23
IIAB	-0.43	0.08	1.45	0.10	1.19	0.06	0.51	0.04	0.26	0.02	0.38	0.08
IIIAB	-0.58	0.15	1.27	0.10	1.04	0.05	0.43	0.05	0.18	0.01	0.36	0.05
IIIE	-0.51	0.09	1.05	0.05	0.94	0.02	0.44	0.07	0.29	0.05	0.28	0.11
IVA	-0.26	0.04	0.95	0.19	0.76	0.12	0.34	0.05	0.18	0.02	0.30	0.16
Gebel Kamil (ungr.)	-0.12	0.07	0.31	0.91	0.34	0.30	0.07	0.15	0.04	0.08	0.32	0.22
IAB	0.00	0.03	-0.03	0.30	0.04	0.10	-0.07	0.05	0.00	0.02	0.02	0.03

[a] Ruthenium isotope ratios are internally normalized to ^{99}Ru/^{101}Ru = 0.7450754. Given uncertainties represent the external reproducibility (2 s.d.) reported in Table S1 or 95% confidence interval (95% CI) for samples with N>3.

[b] Molybdenum isotope data are internally normalized to ^{98}Mo/^{96}Mo = 1.453173. Given uncertainties represent the external reproducibility (2 s.d.) obtained from repeated analyses of BHVO-2 (Budde et al., 2019) or 95% confidence intervals (95% CI) for samples with N>3.

[c] Literature data from Render et al. (2017), Fischer-Gödde et al. (2015), Poole et al. (2017), Bermingham et al. (2018), and Worsham et al. (2019).

Table S3: Comparison of predicted *s*-process slopes, best-fit slopes, and x-axis intercepts for ε^{100}Ru *vs*. ε^{i}Mo correlations calculated with different datasets (described below; Table S2). Best-fits were calculated using the York method in OriginPro (OriginLab, Northampton, MA). Uncertainties are 95% c.i. on slopes and intercepts as calculated by the linear regression. Bold values (***dataset 4***) are the most representative and precise values and used in the main text. The predicted *s*-process slopes were calculated based on equations of Dauphas et al. (2004) and SiC data (Arlandini et al., 1999; Savina et al., 2004; Stephan et al., 2019).

ε^{100}Ru-ε^{i}Mo correlation	**calculated *s*-process slope**	**best-fit slopes Bulk meteorite data**			**best-fit slopes Bulk meteorite data + OC leachate**			**best-fit x-axis intercepts Bulk meteorite data**			**best-fit x-axis intercepts Bulk meteorite data + OC leachate**		
		dataset 1	*dataset 2*	*dataset 3*	***dataset 4***	*dataset 5*	*dataset 6*	*dataset 1*	*dataset 2*	*dataset 3*	***dataset 4***	*dataset 5*	*dataset 6*
ε^{100}Ru-ε^{92}Mo	-0.46	-0.38 ± 0.11	-0.38 ± 0.12	-0.44 ± 0.15	**-0.45 ± 0.06**	-0.45± 0.07	-0.46 ± 0.07	0.13 ± 0.23	0.13 ± 0.25	0.19 ± 0.22	**0.23 ± 0.15**	0.23 ± 0.16	0.22 ± 0.16
ε^{100}Ru-ε^{94}Mo	-0.57	-0.47 ± 0.11	-0.48 ± 0.12	-0.51 ± 0.14	**-0.57 ± 0.05**	-0.57 ± 0.06	-0.57 ± 0.05	0.17 ± 0.14	0.17 ± 0.15	0.19 ± 0.15	**0.25 ± 0.09**	0.25 ± 0.10	0.27 ± 0.10
ε^{100}Ru-ε^{95}Mo	-0.93	-0.87 ± 0.25	-0.87 ± 0.27	-0.92 ± 0.30	**-0.92 ± 0.12**	-0.92 ± 0.13	-0.97 ± 0.15	0.02 ± 0.10	0.02 ± 0.10	0.03 ± 0.10	**0.03 ± 0.06**	0.03 ± 0.07	0.06 ± 0.08
ε^{100}Ru-ε^{97}Mo	-1.71	-1.81 ± 0.60	-1.82 ± 0.66	-1.87 ± 0.79	**-1.81 ± 0.28**	-1.81 ± 0.30	-1.89 ± 0.39	0.04 ± 0.05	0.04 ± 0.05	0.03 ± 0.05	**0.04 ± 0.04**	0.04 ± 0.04	0.05 ± 0.05
ε^{100}Ru-ε^{100}Mo	-1.47	-1.90 ± 0.70	-1.91 ± 0.71	-2.10 ± 0.88	**-1.42 ± 0.20**	-1.42 ± 0.21	-1.41 ± 0.27	0.05 ± 0.05	0.05 ± 0.05	0.05 ± 0.06	**0.01 ± 0.05**	0.01 ± 0.05	0.00 ± 0.09

dataset 1 - All NC data excluding brachinites.
dataset 2 - All NC data excluding ureilites and brachinites.
dataset 3 - All NC data excluding ungrouped achondrites, ureilites, and brachinites.
dataset 4 **- All NC data and ordinary chondrite leachate excluding brachinites.**
dataset 5 - All NC data and ordinary chondrite leachate excluding ureilites and brachinites.
dataset 6 - All NC data and ordinary chondrite leachate excluding ungrouped achondrites, ureilites, and brachinites.

Table S4. Data and sources of Mo and Ru isotope compositions of NC and CC meteorites used in this study.

Meteorites	**ε^{92}Mo**	**ε^{94}Mo**	**ε^{95}Mo**	**ε^{97}Mo**	**ε^{100}Mo**	**ε^{100}Ru**	**Ref. [Mo, Ru]**
Ordinary chondrites	0.66 ± 0.08	0.61 ± 0.06	0.25 ± 0.06	0.11 ± 0.05	0.14 ± 0.08	-0.29 ± 0.03	1,2
Enstatite chondrites	0.38 ± 0.06	0.36 ± 0.05	0.14 ± 0.03	0.09 ± 0.02	0.14 ± 0.03	-0.08 ± 0.04	1,2
IC	0.88 ± 0.39	0.83 ± 0.28	0.36 ± 0.20	0.23 ± 0.14	0.21 ± 0.23	-0.38 ± 0.13	3,3
IIAB	1.45 ± 0.10	1.19 ± 0.06	0.51 ± 0.04	0.26 ± 0.02	0.38 ± 0.08	-0.43 ± 0.08	4,2
IIIAB	1.27 ± 0.10	1.04 ± 0.05	0.43 ± 0.05	0.18 ± 0.01	0.36 ± 0.05	-0.58 ± 0.15	4,5
IIIE	1.05 ± 0.05	0.94 ± 0.02	0.44 ± 0.07	0.29 ± 0.05	0.28 ± 0.11	-0.5 ± 0.09	3,3
IVA	0.95 ± 0.19	0.76 ± 0.12	0.34 ± 0.05	0.18 ± 0.02	0.30 ± 0.16	-0.26 ± 0.04	4,2
Gebel Kamil	0.31 ± 0.91	0.34 ± 0.30	0.07 ± 0.15	0.04 ± 0.08	0.32 ± 0.22	-0.12 ± 0.07	5,5
IAB	-0.03 ± 0.30	0.04 ± 0.10	-0.07 ± 0.05	0.00 ± 0.02	0.02 ± 0.03	0.00 ± 0.03	5,5
IAB	0.00 ± 0.08	0.05 ± 0.06	-0.07 ± 0.06	-0.04 ± 0.02	0.05 ± 0.08	-0.02 ± 0.06	4,2
IIIF	1.57 ± 0.39	1.20 ± 0.28	0.96 ± 0.15	0.55 ± 0.14	0.61 ± 0.23	-0.99 ± 0.13	3,3
IIF	1.52 ± 0.39	1.10 ± 0.28	1.17 ± 0.10	0.51 ± 0.14	0.62 ± 0.23	-0.99 ± 0.13	3,3
IID	1.67 ± 0.39	1.18 ± 0.28	0.99 ± 0.04	0.51 ± 0.14	0.63 ± 0.23	-1.04 ± 0.13	3,3
IIC	2.87 ± 0.10	2.25 ± 0.10	1.56 ± 0.07	0.83 ± 0.07	0.93 ± 012	-1.04 ± 0.05	3,3
Wiley	4.26 ± 0.22	3.42 ± 0.07	2.23 ± 0.08	1.22 ± 0.07	1.47 ± 0.12	-1.07 ± 0.08	3,3
IVB	1.88 ± 0.22	1.33 ± 0.09	0.90 ± 0.15	0.49 ± 0.03	0.81± 0.09	-1.07 ± 0.15	4,2
Tafassasset	2.17 ± 0.15	1.65 ± 0.07	1.20 ± 0.05	0.62 ± 0.05	0.63 ± 0.06	-1.15 ± 0.04	6,7
Milton	-	1.30 ± 0.26	1.04 ± 0.09	0.54 ± 0.05	-	-1.14 ± 0.15	8,8
SBT triplet	-	1.27 ± 0.07	1.03 ± 0.04	0.49 ± 0.02	-	-1.07 ± 0.05	8,8
CB chondrite	1.53 ± 0.08	1.26 ± 0.04	0.99 ± 0.04	0.51 ± 0.04	0.45 ± 0.04	-1.04 ± 0.04	6,2
Allende	1.41 ± 0.27	0.97 ± 0.19	0.81 ± 0.05	0.40 ± 0.08	0.44 ± 0.12	-1.01 ± 0.04	9,2
BSE	0.08 ± 0.13	0.04 ± 0.06	0.10 ± 0.04	0.06 ± 0.02	-0.07 ± 0.04	0.01 ± 0.07	6,10

1 – Render et al. (2017), 2 – Fischer-Gödde et al. (2015), 3 – Worsham et al. (2019), 4 – Poole et al. (2017), 5 – Bermingham et al. (2018), 6 – Budde et al. (2019), 7 – Fischer-Gödde and Kleine (2017), 8 – Hilton et al. (2019), 9 – Budde et al. (2016), 10 – Bermingham and Walker (2017).

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