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Supplementary Materials for

Water circulation in Ryugu asteroid affected the distribution of nucleosynthetic isotope anomalies in returned sample

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Supplementary Text

Mass balance calculation for the involvement of secondary minerals in Ryugu and CIs

We have conducted a mass balance calculation to derive the amounts of secondary minerals in the Ryugu samples and CI chondrites that explain the observed ε^{53}Cr and ε^{54}Cr values. In the calculation, we used the abundances of Ti, Cr, and Mn in the bulk Ryugu samples, CI chondrites, and the secondary minerals, as well as the ε^{53}Cr and ε^{54}Cr values in these samples (Table S8). Of the secondary minerals, dolomite, magnetite, and pyrrhotite are considered to be the dominant phases that can contribute to the bulk Cr isotopic composition. The abundances of Ti, Cr, and Mn of these minerals in Ryugu were taken from those reported by (*37*).

For simplification, magnetite and pyrrhotite were combined to be a single phase (magnetite + pyrrhotite) because these minerals in Ryugu have Ti, Cr, and Mn abundances similar to each other.

In the mass balance calculation, the amounts of dolomite and magnetite + pyrrhotite in individual samples were determined as relative percentages of enrichment of these minerals (*x* and *y*, respectively) compared to the mineral abundances in the most ^{53}Cr-poor/^{54}Cr-rich sample, Ryugu C0002. By using the *x* and *y* values, the values of Ti/Cr, Mn/Cr, ε^{53}Cr and ε^{54}Cr in individual samples are calculated as follows;

$$\left(\frac{\mathrm{Ti}}{\mathrm{Cr}}\right)_{calc} = \frac{\left(1-\frac{x}{100}-\frac{y}{100}\right)[\mathrm{Ti}]_{C0002}+\frac{x}{100}[\mathrm{Ti}]_{dol}+\frac{y}{100}[\mathrm{Ti}]_{mt+py}}{\left(1-\frac{x}{100}-\frac{y}{100}\right)[\mathrm{Cr}]_{C0002}+\frac{x}{100}[\mathrm{Cr}]_{dol}+\frac{y}{100}[\mathrm{Cr}]_{mt+py}}$$

$$\left(\frac{\mathrm{Mn}}{\mathrm{Cr}}\right)_{calc} = \frac{\left(1-\frac{x}{100}-\frac{y}{100}\right)[\mathrm{Mn}]_{C0002}+\frac{x}{100}[\mathrm{Mn}]_{dol}+\frac{y}{100}[\mathrm{Mn}]_{mt+py}}{\left(1-\frac{x}{100}-\frac{y}{100}\right)[\mathrm{Cr}]_{C0002}+\frac{x}{100}[\mathrm{Cr}]_{dol}+\frac{y}{100}[\mathrm{Cr}]_{mt+py}}$$

$$\varepsilon^{53}\mathrm{Cr}_{calc} = \frac{\left(1-\frac{x}{100}-\frac{y}{100}\right)[\mathrm{Cr}]_{C0002}\cdot\varepsilon^{53}\mathrm{Cr}_{C0002}+\frac{x}{100}[\mathrm{Cr}]_{dol}\cdot\varepsilon^{53}\mathrm{Cr}_{dol}+\frac{y}{100}[\mathrm{Cr}]_{mt+py}\cdot\varepsilon^{53}\mathrm{Cr}_{mt+py}}{\left(1-\frac{x}{100}-\frac{y}{100}\right)[\mathrm{Cr}]_{C0002}+\frac{x}{100}[\mathrm{Cr}]_{dol}+\frac{y}{100}[\mathrm{Cr}]_{mt+py}}$$

$$\varepsilon^{54}\mathrm{Cr}_{calc} = \frac{\left(1-\frac{x}{100}-\frac{y}{100}\right)[\mathrm{Cr}]_{C0002}\cdot\varepsilon^{54}\mathrm{Cr}_{C0002}+\frac{x}{100}[\mathrm{Cr}]_{dol}\cdot\varepsilon^{54}\mathrm{Cr}_{dol}+\frac{y}{100}[\mathrm{Cr}]_{mt+py}\cdot\varepsilon^{54}\mathrm{Cr}_{mt+py}}{\left(1-\frac{x}{100}-\frac{y}{100}\right)[\mathrm{Cr}]_{C0002}+\frac{x}{100}[\mathrm{Cr}]_{dol}+\frac{y}{100}[\mathrm{Cr}]_{mt+py}}$$

where [Ti], [Cr], and [Mn] represent the mass fractions of Ti, Cr, and Mn, respectively. The $\varepsilon^{53}\mathrm{Cr}_{dol}$ and $\varepsilon^{53}\mathrm{Cr}_{mt+py}$ values were determined from the ^{53}Mn-^{53}Cr isochron of bulk Ryugu samples (Fig. 1) using the ratios of $(\mathrm{Mn/Cr})_{dol}$ and $(\mathrm{Mn/Cr})_{mt+py}$, respectively. The ε^{54}Cr value of the aqueous fluid (i.e., $\varepsilon^{54}\mathrm{Cr}_{dol}$ and $\varepsilon^{54}\mathrm{Cr}_{mt+py}$) is unknown, but is expected to be more depleted in ^{54}Cr than that of the earlier fractions obtained by mild acid leaching in sequential acid leaching experiments of CI chondrites (*21-23*). Therefore, we set this value as a free parameter with a plausible range between –10 and –25 in the calculation. Then, the *x* and *y* values for each sample were determined by the least squares method to minimize the following Δ value;

$$\Delta = \sum_i \frac{\left(Z_i^{measured}-Z_i^{calc}\right)^2}{{\sigma_i}^2}$$

where Z_i is the Ti/Cr, Mn/Cr, ε^{53}Cr, and ε^{54}Cr values of each sample, and σ_i is the uncertainty of Z_i. The σ_i values for Ti/Cr and Mn/Cr were assumed to be 5% of the observed ratios, and those for ε^{53}Cr and ε^{54}Cr were taken from their analytical uncertainties (Table 1).

We found that the summation of all Δ values of individual samples became minimum when the $\varepsilon^{54}\mathrm{Cr}_{dol}$ and $\varepsilon^{54}\mathrm{Cr}_{mt+py}$ values were –19 (Table S6). In this case, the *x* and *y* values of individual samples ranged from 0.5 to 3.2% and from 0.8 to 4.2%, respectively, and the resulting Ti/Cr, Mn/Cr, ε^{53}Cr, and ε^{54}Cr values are generally in good agreement with the measured values (Fig. S6). The actual secondary minerals in Ryugu and CI chondrites have variations in the Ti, Cr, and Mn abundances, while we used the mean values of literature data. This may have caused the subtle differences between the calculated and measured values observed in Fig. S6. Otherwise, there could be additional secondary minerals containing Ti, Cr, and Mn that were not included in the mass balance calculation.

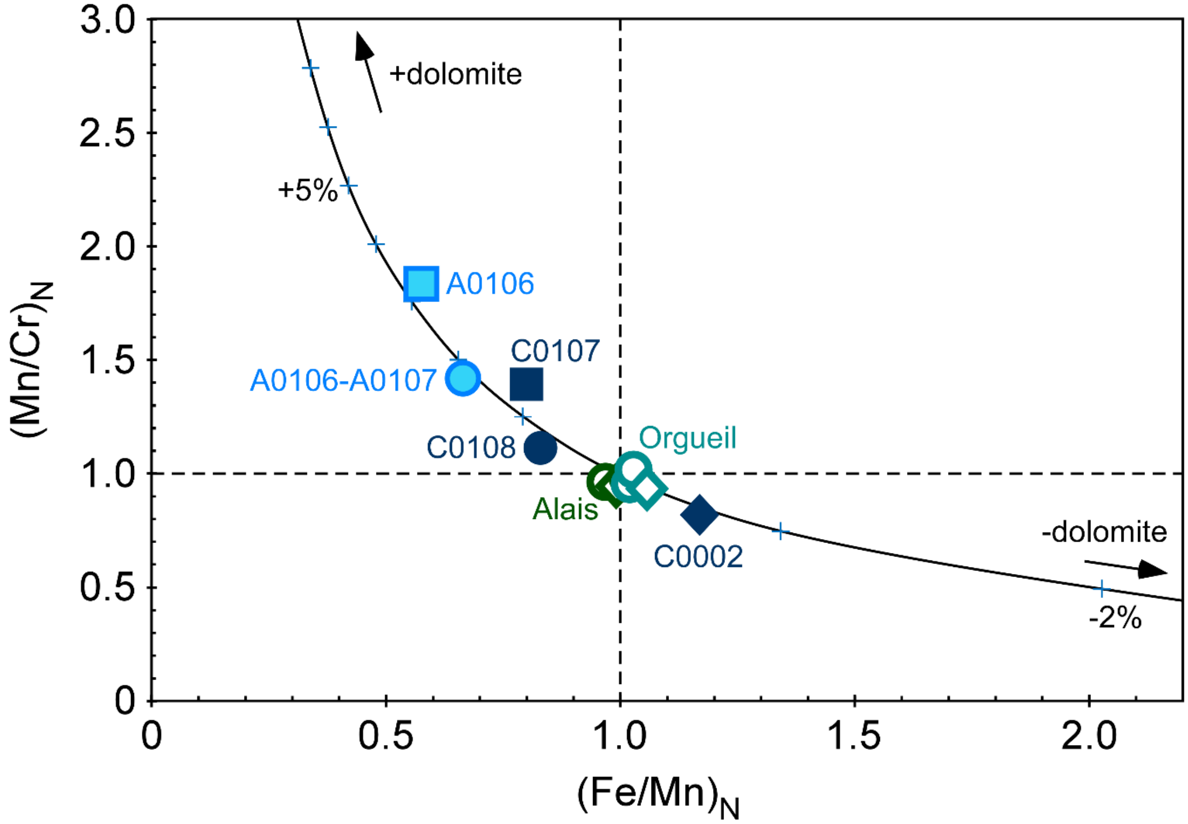

Fig. S1. Correlation between CI-normalized Mn/Cr and Fe/Mn ratios for bulk samples of Ryugu and CI chondrites examined in this study. The CI composition of Lodders (*6*) was used for the normalization. Data are listed in Table S9. The curve shows a calculated mixing relationship between bulk CI and dolomite in Ryugu (*37*).

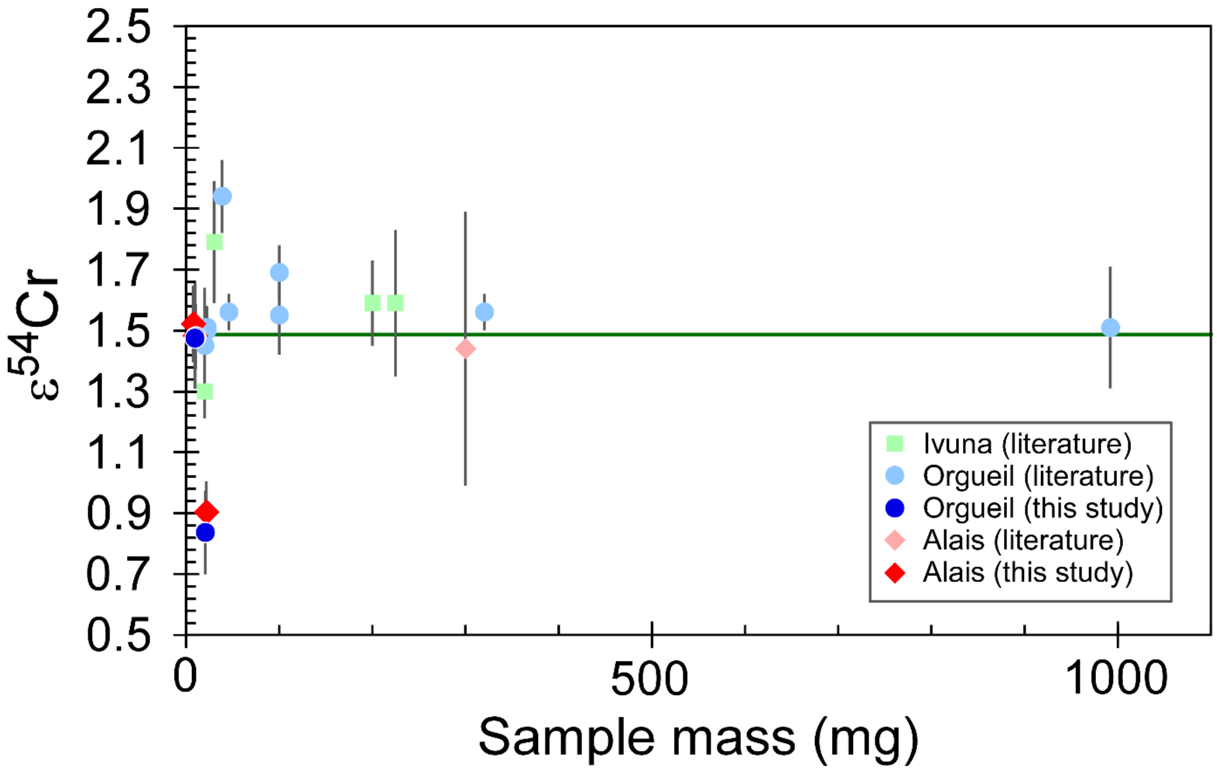

Fig. S2. ε^{54}Cr values of CI chondrites. Data are plotted against the mass (mg) of sample used.

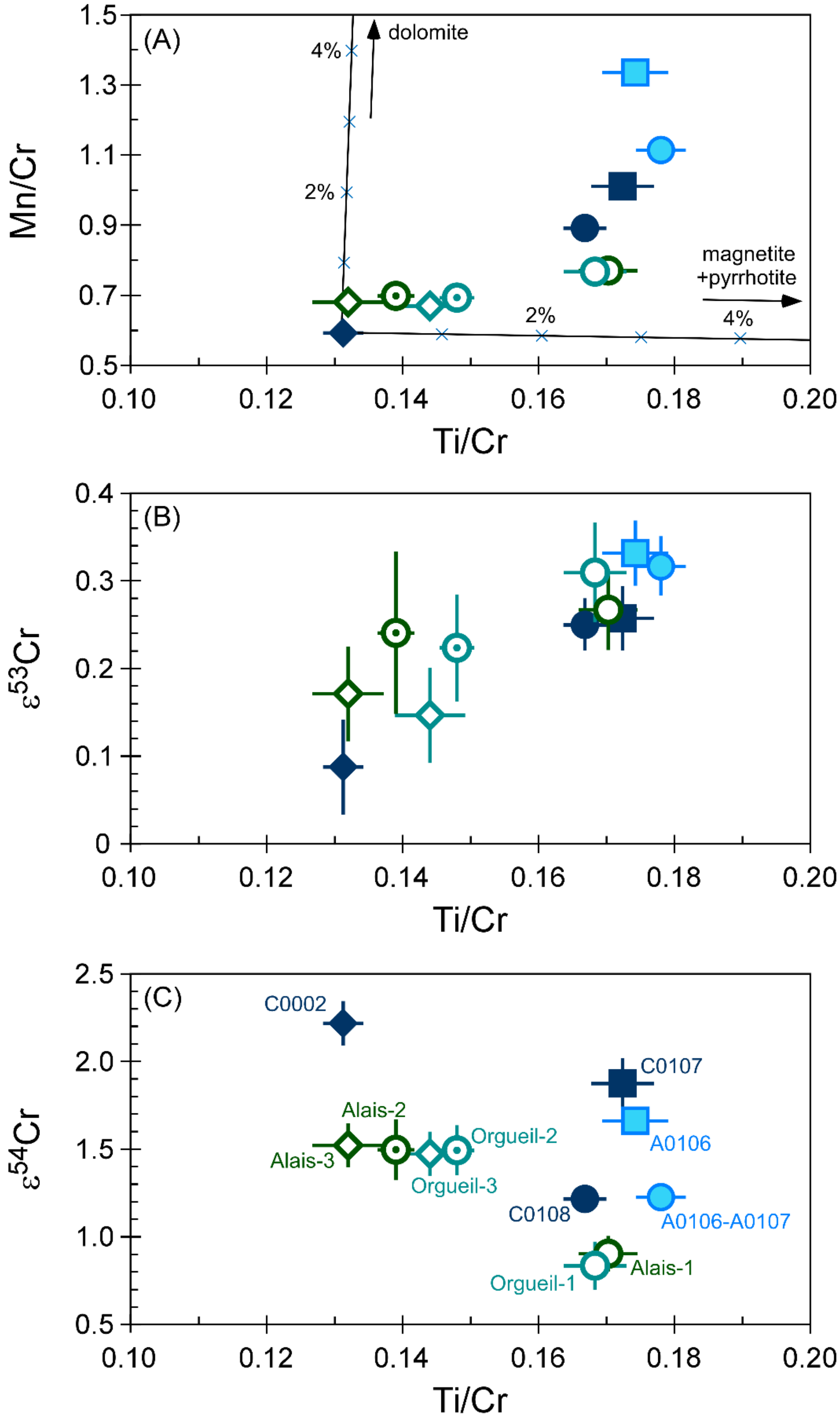

Fig. S3. Variation of Mn/Cr, Ti/Cr, ε^{53}Cr, and ε^{54}Cr values for Ryugu and CI chondrites. (A) Mn/Cr versus Ti/Cr diagram for Ryugu and CI chondrites. Vertical line is calculated mixing relationship between Ryugu C0002 and dolomite in Ryugu and horizontal line is that between C0002 and magnetite + pyrrhotite in Ryugu using the mineral compositions of (*37*). (B) ε^{53}Cr versus Ti/Cr for Ryugu and CI chondrites. (C) ε^{54}Cr versus Ti/Cr diagrams for Ryugu and CI chondrites.

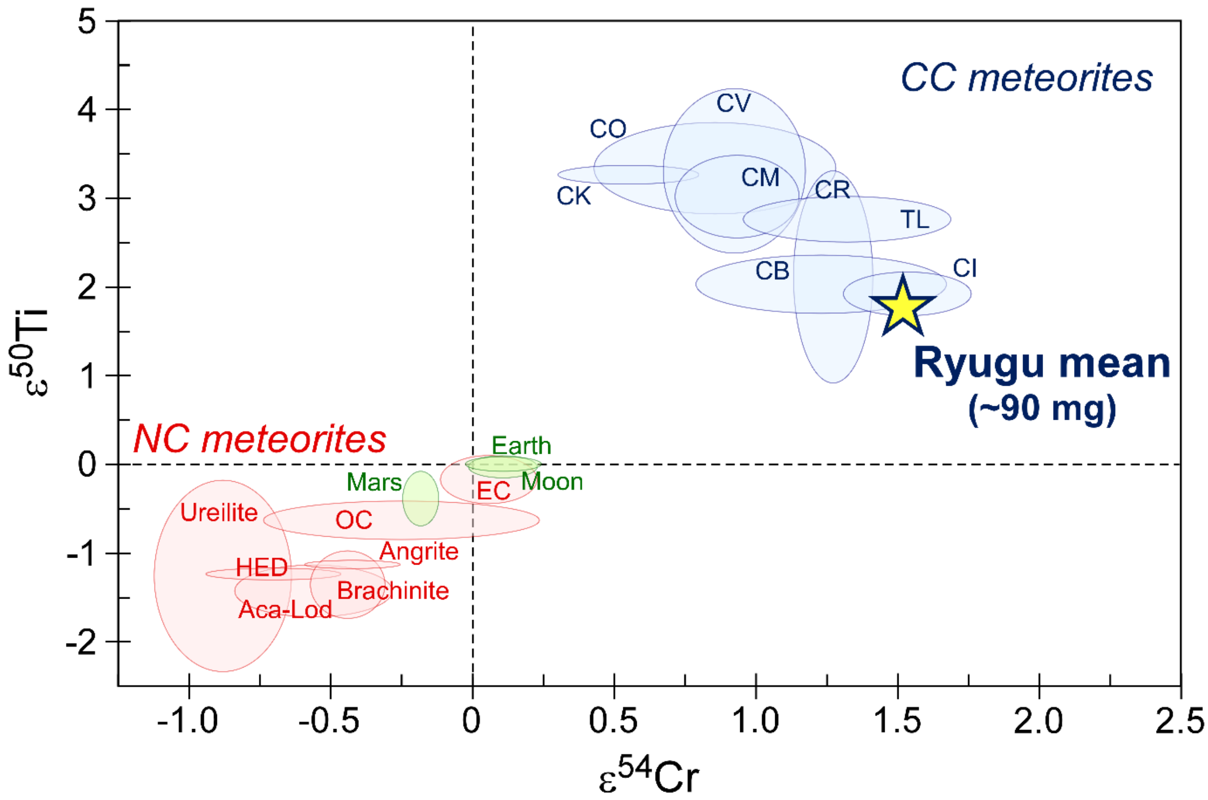

Fig. S4. Mean ε^{50}Ti and ε^{54}Cr values for the bulk Ryugu sample. The weight of the sample is equivalent to a total of ~90 mg.

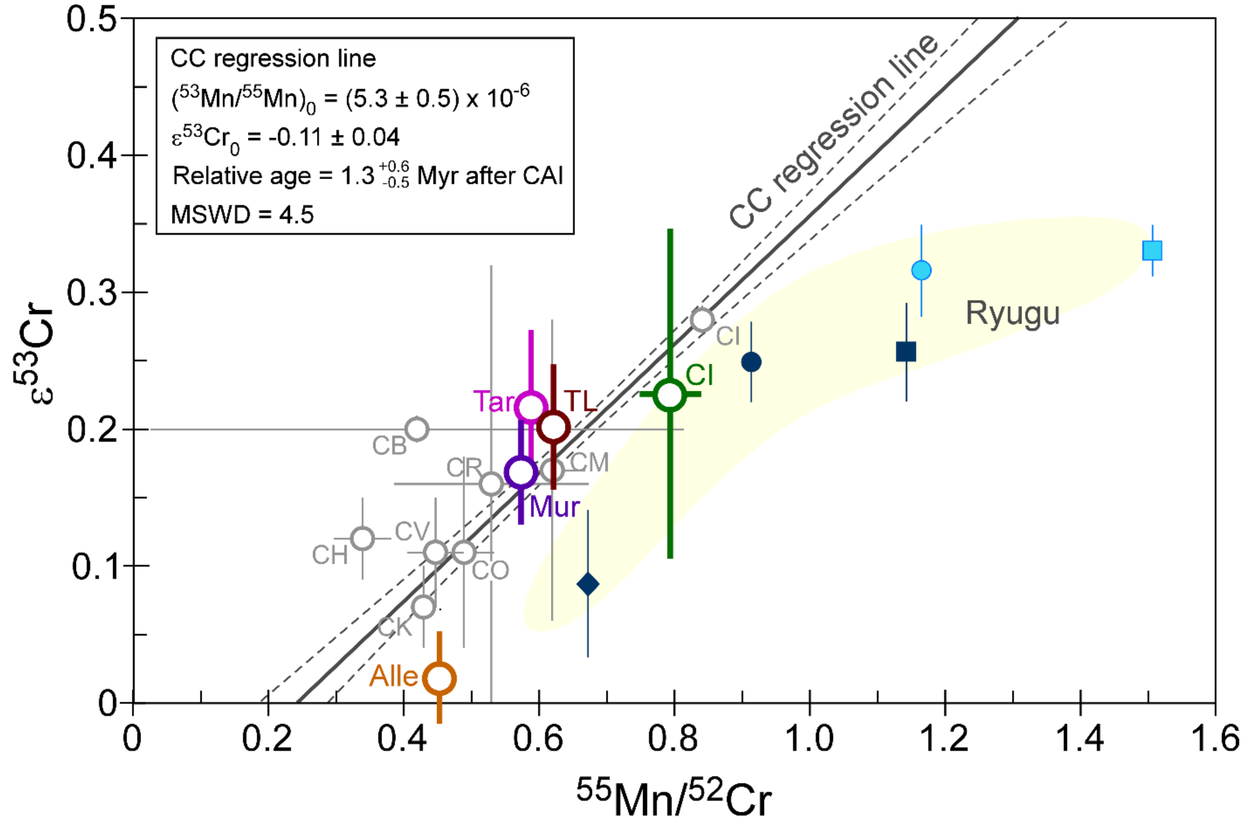

Fig. S5. ^{53}Mn-^{53}Cr isotope systematics of carbonaceous chondrites. Abbreviations are the same as Fig. 1. The yellow band represents the range of Ryugu data. The regression line determined from the six carbonaceous chondrites analyzed here and literature data for bulk carbonaceous chondrites (CI, CM, CR, CO, CV, CK, and CB: *17*) yields a slope of $(^{53}Mn/^{55}Mn)_0 = (5.3\pm0.5)\times10^{-6}$, corresponding to an older age (i.e., $\mathbf{1.3^{+0.6}_{-0.5}}$ Myr after CAI) than that determined from the Ryugu regression line. This result, however, does not necessarily indicate that planetary scale Mn-Cr fractionation for the parent bodies of carbonaceous chondrites predates the aqueous alteration on Ryugu. As discussed in (*17*), the ^{53}Mn-^{53}Cr "age" for the carbonaceous chondrites may be averaging the effects of multiple fractionation events that affected these individual chondrites, which vary with the modal abundances of their constituents. These consist of mechanical mixtures of CAIs, chondrules, metals, and matrix that may have formed at different times in the early Solar System and undergone aqueous alteration at different times.

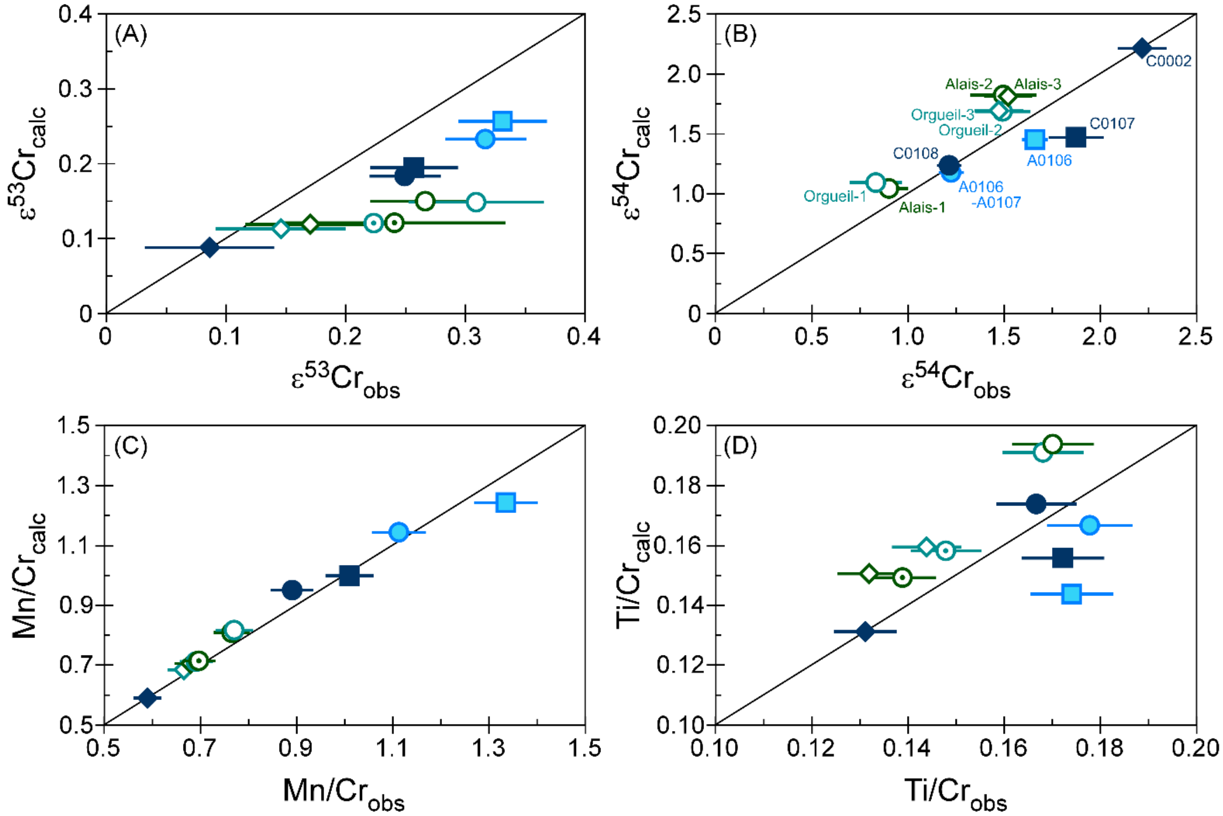

Fig. S6. Comparison between observed and calculated values for Ryugu samples and CI chondrites. (A) ε^{54}Cr, (B) ε^{54}Cr, (C) Mn/Cr, and (D) Ti/Cr. The calculated values were obtained by mass balance calculation considering the influence of secondary minerals.

Table S1. Repeated measurements of Cr isotopic ratios for individual samples measured using TIMS at Tokyo Tech.

Sample	$\varepsilon^{53}Cr$	2SE	$\varepsilon^{54}Cr$	2SE
Ryugu				
A0106-A0107#	0.42	0.02	1.43	0.04
Replicate #	0.34	0.02	1.28	0.03
Replicate	0.33	0.02	1.23	0.03
Replicate	0.29	0.01	1.11	0.03
Replicate	0.34	0.01	1.21	0.03
Replicate	0.32	0.02	1.24	0.04
Replicate	0.32	0.02	1.18	0.03
Average	0.34	0.04	1.24	0.09
C0108#	0.28	0.02	1.26	0.04
Replicate #	0.27	0.01	1.24	0.03
Replicate	0.26	0.01	1.26	0.03
Replicate	0.23	0.01	1.18	0.03
Replicate	0.27	0.02	1.20	0.03
Replicate	0.30	0.02	1.34	0.04
Replicate	0.26	0.02	1.23	0.04
Average	0.27	0.04	1.24	0.09
A0106	0.29	0.01	1.56	0.03
Replicate	0.33	0.01	1.61	0.03
Replicate	0.31	0.01	1.61	0.03
Replicate	0.35	0.02	1.59	0.04
Average	0.32	0.06	1.59	0.11
C0107	0.23	0.01	1.85	0.03
Replicate	0.24	0.02	1.86	0.03
Replicate	0.22	0.01	1.81	0.03
Replicate	0.25	0.02	1.89	0.03
Replicate	0.23	0.02	1.93	0.04
Average	0.23	0.06	1.87	0.11
C0002	0.09	0.02	2.27	0.03
Replicate	0.08	0.02	2.16	0.03
Average	0.09	0.05	2.22	0.13
Meteorites				
Orgueil (Batch-1: Acid)	0.40	0.02	1.03	0.04
Replicate	0.31	0.02	0.87	0.03
Replicate	0.28	0.02	0.74	0.03
Replicate	0.26	0.02	0.79	0.04
Average	0.31	0.05	0.86	0.11
Orgueil (Batch-2: Acid)	0.25	0.02	1.56	0.04
Replicate	0.20	0.02	1.38	0.04
Average	0.22	0.06	1.49	0.14
Orgueil (Batch-3: Alkali fusion)	0.17	0.02	1.52	0.03
Replicate	0.12	0.02	1.43	0.03
Average	0.15	0.05	1.47	0.13
Alais (Batch-1: Acid)	0.29	0.02	0.97	0.03
Replicate	0.23	0.02	0.82	0.03
Replicate	0.28	0.02	0.95	0.04
Replicate	0.24	0.02	0.93	0.04
Average	0.26	0.05	0.92	0.11
Alais (Batch-2)	0.19	0.02	1.41	0.04
Replicate	0.29	0.02	1.58	0.04
Average	0.24	0.09	1.50	0.17
Alais (Batch-3: Alkali fusion)	0.19	0.01	1.58	0.03
Replicate	0.15	0.01	1.47	0.03
Average	0.17	0.05	1.52	0.13
Tarda	0.19	0.01	0.95	0.03
Replicate	0.20	0.01	1.01	0.03
Replicate	0.17	0.01	0.91	0.03
Replicate	0.23	0.01	1.06	0.03
Replicate	0.26	0.02	1.15	0.03
Average	0.21	0.06	1.02	0.13
Tagish Lake	0.17	0.01	0.93	0.03
Replicate	0.23	0.02	1.04	0.04
Replicate	0.21	0.02	0.96	0.03
Average	0.21	0.06	0.97	0.13
Murchison	0.16	0.02	0.88	0.04
Replicate	0.19	0.01	0.98	0.03
Replicate	0.19	0.01	0.95	0.03
Replicate	0.22	0.02	1.03	0.03
Average	0.19	0.04	0.96	0.09
Allende (Batch-1)	0.07	0.02	0.87	0.03
Replicate	0.07	0.02	0.84	0.03
Replicate	0.08	0.02	0.84	0.03
Replicate	0.06	0.02	0.80	0.03
Average	0.07	0.04	0.84	0.09

Reported in (*3*).

εCr values are reported relative to NIST 979 Cr standard.

Table S2. Repeated measurements of Ti isotopic ratios for individual samples measured using MC-ICP-MS at U Tokyo.

Sample	ε^{46}Ti	2SE	ε^{48}Ti	2SE	ε^{50}Ti	2SE
Ryugu						
A0106-A0107#	0.25	0.21	-0.05	0.09	1.63	0.20
replicate	0.28	0.14	-0.01	0.06	1.59	0.17
Average	0.27	0.13	-0.03	0.05	1.61	0.13
C0108#	0.09	0.18	-0.12	0.10	2.02	0.19
replicate	0.27	0.20	-0.12	0.06	1.79	0.18
Average	0.18	0.13	-0.12	0.06	1.90	0.13
A0106	0.11	0.15	0.02	0.06	1.54	0.13
C0107	0.37	0.17	-0.03	0.06	1.63	0.16
Meteorites						
Orgueil (Batch-1: Acid)	0.44	0.15	0.01	0.07	2.14	0.17
Orgueil (Batch-2: Acid)	0.33	0.16	-0.10	0.09	1.93	0.19
Alais (Batch-1: Acid)	0.35	0.22	-0.03	0.11	1.75	0.19
Alais (Batch-2: Acid)	0.47	0.15	-0.24	0.17	1.85	0.20
Tarda	0.22	0.19	-0.51	0.08	2.48	0.16
Tagish Lake	0.76	0.23	0.08	0.12	2.45	0.21
Murchison	0.47	0.17	0.30	0.07	3.02	0.15
replicate	0.42	0.20	0.25	0.06	2.83	0.19
replicate	0.22	0.24	-0.10	0.08	3.01	0.25
replicate	0.36	0.20	-0.12	0.08	2.73	0.22
Average	0.37	0.13	0.08	0.26	2.90	0.17
Allende (Batch-1)	0.75	0.22	0.19	0.10	3.29	0.20
replicate	0.65	0.20	0.22	0.07	3.29	0.18
replicate	0.65	0.20	-0.11	0.08	3.38	0.24
replicate	0.81	0.18	-0.22	0.08	3.28	0.22
replicate	0.72	0.17	-0.05	0.07	3.00	0.19
replicate	0.83	0.13	-0.04	0.07	3.08	0.17
replicate	0.60	0.14	0.03	0.06	3.35	0.13
replicate	0.62	0.13	-0.02	0.06	3.33	0.16
Average	0.70	0.07	0.00	0.11	3.25	0.10

Reported in (*3*).

εTi values are reported relative to Alfar Aesar Ti standard.
Number of replicates for each Ti isotope measurement is 1.

Table S3. Repeated measurements of Cr and Ti isotopic ratios for individual samples measured using MC-ICP-MS at ASU.

Sample	*N* (Cr)	ε^{53}Cr	2SE	ε^{54}Cr	2SE	*N* (Ti)	ε^{46}Ti	2SE	ε^{48}Ti	2SE	ε^{50}Ti	2SE
Ryugu												
A0106-A0107	8	0.30	0.05	1.21	0.09	7	0.42	0.24	-0.08	0.05	1.68	0.18
C0108	7	0.23	0.04	1.19	0.09	7	0.51	0.13	-0.10	0.09	1.98	0.15
A0106	6	0.34	0.05	1.73	0.08	5	0.41	0.12	-0.07	0.09	1.72	0.09
C0107	5	0.28	0.04	1.88	0.26	5	0.45	0.18	-0.08	0.13	2.08	0.12
Meteorites												
Orgueil (Batch-1)	5	0.30	0.06	0.81	0.17	5	0.30	0.13	-0.08	0.04	2.10	0.17
Alais (Batch-1)	5	0.27	0.04	0.89	0.10	4	0.20	0.19	-0.02	0.11	2.31	0.06
Tarda	7	0.22	0.05	0.97	0.15	7	0.41	0.21	-0.16	0.09	2.81	0.13
Tagish Lake	7	0.20	0.03	1.03	0.08	5	0.48	0.21	-0.09	0.10	2.78	0.14
Murchison	6	0.15	0.04	0.91	0.07							
Allende (Batch-1)	6	-0.03	0.03	0.87	0.07	2	0.55	0.29	-0.18	0.12	3.39	0.27
Allende (Batch-2a)						5	0.56	0.24	-0.16	0.13	3.25	0.22
Allende (Batch-2b)						4	0.84	0.09	-0.12	0.07	3.76	0.16
Average							0.65	0.23	-0.15	0.05	3.47	0.37

εTi values are reported relative to Alfar Aesar Ti standard (see the text for details).
N(Cr) and N(Ti) are the number of replicates for Cr and Ti isotope measurements, respectively.

Table S4. Sample details.

Sample Name	ID	Initial weight of sample for making powder	Weight of sample dissolved for isotopic analysis	Source
Ryugu	A0106-A0107	A0106: 1.6 mg A0107: 27.29 mg	23.88 mg (acid digestion)	JAXA
Ryugu	C0108	33.34 mg	22.24 mg (acid digestion)	JAXA
Ryugu	A0106*	17.15 mg (before SOM extraction) 14.17 mg (after SOM extraction)	14.61 mg (acid digestion)#	JAXA
Ryugu	C0107*	17.36 mg (before SOM extraction) 12.92 mg (after SOM extraction)	12.78 mg (acid digestion)	JAXA
Ryugu	C0002	6.85 mg	6.85 mg (alkali fusion)	JAXA
Orgueil, CI1	n219	50 mg	20.82 mg (acid digestion-1) 10.40 mg (acid digestion-2) 9.67 mg (alkali fusion)	MNHN
Alais, CI1	n25	51 mg	21.98 mg (acid digestion-1) 10.15 mg (acid digestion-2) 7.85 mg (alkali fusion)	MNHN
Tarda, C2-ung	not available	212.44 mg	25.10 mg (acid digestion)	Meteorite.fr
Tagish Lake, C2-ung	not available	1055 mg	24.29 mg (acid digestion)	The Meteorite Market
Murchison, CM2	not available	1645 mg	24.76 mg (acid digestion)	Michael Farmer Meteorites
Allende, CV3	USNM 3529, Split 20 Position 31	4 kg	24.92 mg (acid digestion-1) 39.76 mg (acid digestion-2)	Smithsonian

* SOM (soluble organic matter) extracted residues.

\# The dissolved weight is greater than the initial weight because of the uncertainty of two scales used in different laboratories.

Table S5. Literature data for ε^{50}Ti and ε^{54}Cr.

Name	group	ε^{50}Ti	2σ	ε^{54}Cr	2σ	ε^{50}Ti ref.	ε^{54}Cr ref.
Carbonaceous chondrites							
Alais	CI1			1.44	0.45		(60)
Ivuna	CI1	2.08	0.51	1.47	0.20	(50)	(61)
Ivuna	CI1	1.91	0.10	1.30	0.08	(51)	(50)
Ivuna	CI1	1.97	0.06	1.59	0.24	(51)	(61)
Ivuna	CI1			1.79	0.20		(62)
Ivuna	CI1			1.55	0.05		(23)
Ivuna	CI1			1.59	0.14		(17)
Orgueil	CI1	1.74	0.05	1.65	0.07	(52)	(63)
Orgueil	CI1	1.92	0.06	1.45	0.19	(51)	(64)
Orgueil	CI1			1.51	0.20		(61)
Orgueil	CI1			1.94	0.12		(62)
Orgueil	CI1			1.51	0.05		(17)
Orgueil	CI1			1.50	0.08		(17)
Orgueil	CI1			1.56	0.06		(22)
Orgueil	CI1			1.56	0.06		(65)
SCO 06043	CM1			1.13	0.12		(17)
Aguas Zarcas	CM2	2.21	0.18	0.86	0.03	(47)	(17)
Aguas Zarcas	CM2	2.86	0.23	0.98	0.13	(47)	(47)
Aguas Zarcas	CM2			0.88	0.13		(47)
Banten	CM2			0.86	0.05		(17)
Cold Bokkeveld	CM2	3.27	0.09	0.81	0.12	(51)	(66)
Jbilet Winselwan	CM2	3.04	0.05	0.82	0.04	(51)	(17)
Jbilet Winselwan	CM2			1.01	0.12		(66)
Maribo	CM2			1.13	0.15		(66)
Mighei	CM2			1.50			(60)
Mighei	CM2			0.74	0.10		(66)
Murchison	CM2	3.08	0.05	0.98	0.19	(7)	(64)
Murchison	CM2	2.81	0.15	0.89	0.08	(47)	(67)
Murchison	CM2	2.94	0.10	1.10	0.15	(53)	(47)
Murchison	CM2	3.17	0.12	1.01	0.05	(53)	(22)
Murchison	CM2			0.93	0.07		(66)
Murchison	CM2			0.97	0.20		(63)
Murray	CM2	2.84	0.05	1.13	0.21	(52)	(61)
Murray	CM2	2.67	0.15	0.84	0.13	(47)	(47)
Murray	CM2	3.01	0.09	1.01	0.12	(51)	(68)
Murray	CM2			0.85	0.10		(66)
Nogoya	CM2			0.76	0.04		(17)
NWA8157	CM2			1.01	0.18		(69)
Paris	CM2			0.93	0.09		(69)
Sutters Mill	CM2			0.95	0.09		(67)
Sutters Mill	CM2			0.88	0.07		(67)
Diepenveen	CM2-an			0.85	0.10		(70)
GRO 95566	CM-an	3.50	0.15	0.92	0.13	(47)	(47)
GRO 95577	CR1			1.25	0.06		(17)
EET 92042	CR2			1.25	0.10		(71)
EET 92048	CR2	2.25	0.09			(51)	
EET 92161	CR2			1.19	0.12		(68)
GRA 06100	CR2	3.26	0.09	1.32	0.11	(54)	(63)
GRA 06100	CR2	1.94	0.08			(51)	
GRA 95229	CR2	2.30	0.51	1.18	0.07	(50)	(50)
LAP 02342	CR2	1.50	0.51	1.49	0.11	(50)	(50)
NWA 6043	CR2			1.24	0.10		(68)
NWA 7837	CR2			1.06	0.08		(68)
NWA 801	CR2	2.35	0.04			(52)	
QUE 99177	CR2	2.27	0.51	1.43	0.12	(50)	(50)
Renazzo	CR2	1.40	2.50	1.30	0.21	(55)	(22)
Renazzo	CR2	1.63	0.07	1.22	0.10	(51)	(72)
Shisr 033	CR2	1.49	0.10			(51)	
Al Rais	CR2-an			1.24	0.11		(17)
NWA 6921	CR6	2.89	1.04	1.32	0.09	(50)	(50)
NWA 7317	CR6			1.32	0.09		(72)
Acfer 202	CO3	3.39	0.09			(51)	
DAG 005	CO3	3.44	0.08			(51)	
DOM 10104	CO3			0.80	0.06		(17)
MIL 07193	CO3			1.22	0.04		(17)
Kainsaz	CO3.1			0.87	0.18		(63)
Kainsaz	CO3.1			1.02	0.24		(61)
Felix	CO3.2	4.69	0.12	0.63	0.09	(7)	(22)
Ornans	CO3.3	3.37	0.09	1.03	0.19	(52)	(64)
Ornans	CO3.3			0.90	0.03		(17)
Lance	CO3.4	3.46	0.10	0.57	0.11	(52)	(22)
Warrenton	CO3.6						
Isna	CO3.8	3.54	0.07			(7)	
Isna	CO3.8	2.83	0.15	0.66	0.14	(47)	(47)

Table S5. (continued).

Name	group	$\varepsilon^{50}Ti$	2σ	$\varepsilon^{54}Cr$	2σ	$\varepsilon^{50}Ti$ ref.	$\varepsilon^{54}Cr$ ref.
Allende	CV3	3.49	0.04	1.04	0.19	(52)	(74)
Allende	CV3	3.39	0.18	0.86	0.09	(53)	(50)
Allende	CV3	3.77	0.06	0.95	0.10	(53)	(63)
Allende	CV3	3.25	0.10	0.87	0.08	(53)	(17)
Allende	CV3	2.59	0.05	0.90	0.02	(51)	(17)
Allende	CV3			0.94	0.02		(17)
Allende	CV3			0.95	0.08		(17)
Allende	CV3			0.96	0.05		(17)
Allende	CV3			1.10	0.08		(75)
Allende	CV3			0.88	0.17		(76)
Allende	CV3			0.86	0.09		(22)
Allende	CV3			1.24	0.24		(62)
Bali	CV3			1.10	0.06		(17)
Grosnaja	CV3			2.77	0.10	(51)	
Kaba	CV3			0.89			(60)
Kaba	CV3			0.70	0.07		(17)
Leoville	CV3	4.09	0.08	0.71	0.15	(52)	(63)
Leoville	CV3			0.81	0.10		(17)
Mokoia	CV3			4.98			(60)
Mokoia	CV3			1.00	0.01		(17)
Vigarano	CV3	3.35	0.09	0.87	0.09	(51)	(63)
Vigarano	CV3			0.84	0.04		(17)
NWA 6047	CK3			1.23	0.09		(74)
ALH 85002	CK4			0.46	0.05		(17)
Karoonda	CK4	3.26	0.10	0.63	0.09	(52)	(22)
Karoonda	CK4			0.50	0.09		(17)
NWA 7461	CK4			0.67	0.11		(74)
EET 92002	CK5			0.52	0.09		(17)
EET 92002	CK5			0.33	0.12		(67)
NWA 7704	CK5			0.71	0.11		(74)
LEW 87009	CK6			0.58	0.05		(17)
MIL 05082	CB			1.50	0.09		(17)
Bencubbin	CBa	1.84	0.43	1.11	0.09	(7)	(22)
Bencubbin	CBa	2.24	0.08	1.13	0.09	(51)	(22)
Gujba	CBa	2.04	0.07	1.07	0.27	(7)	(22)
Gujba	CBa	2.00	0.06	1.29	0.07	(51)	(77)
QC 001	CBa			1.45	0.06		(17)
HaH 237	CBb			1.42	0.04		(17)
HaH 237	CBb			0.87	0.19		(61)
Tagish Lake	C2-ung	2.76	0.26	1.19	0.15	(7)	(65)
Tagish Lake	C2-ung			1.45	0.07		(50)
Ordinary chondrites							
Bremervorde	H/L3			0.36	0.19		(64)
TIE	H3			-0.18	0.12		(63)
Roosevelt	H3.4			-0.44	0.03		(56)
Brownsflied	H3.7			-0.44	0.03		(56)
Bath	H4			0.28			(60)
Bath	H4			-0.36	0.04		(56)
Beaver Creek	H4			-0.40	0.04		(56)
Forest Vale	H4	-0.50	1.50			(55)	
Kesen	H4	-0.37	0.05			(52)	
LAP 03601	H4			-0.28	0.11		(63)
Menow	H4			-0.43	0.03		(56)
Ochansk	H4			-0.40	0.03		(56)
Ste. Marguerite	H4			-0.39	0.07		(22)
Forest City	H5	-0.43	0.08	-0.36	0.03	(51)	(56)
Juancheng	H5	-0.66	0.16			(7)	
Richardton	H5	-0.46	0.14			(53)	
Richardton	H5	-0.46	0.12			(53)	
Richardton	H5	-0.49	0.05			(51)	
Allegan	H5	-0.48	0.10			(53)	
Allegan	H5	-0.60	0.08			(51)	
Ejby	H5/6	-0.70	0.13	-0.42	0.06	(57)	(57)
Aarhus	H6			-0.41	0.03		(56)
Estacado	H6			-0.35	0.06		(56)
Kernouve	H6			-0.37	0.07		(22)
Kernouve	H6			0.27	0.19		(64)
Portales Valley	H6/7			-0.37	0.04		(56)
Watson 012	H7	-0.73	0.20	-0.14	0.10	(50)	(50)

Table S5. (continued).

Name	group	$\varepsilon^{50}Ti$	2σ	$\varepsilon^{54}Cr$	2σ	$\varepsilon^{50}Ti$ ref.	$\varepsilon^{54}Cr$ ref.
Knyahinya	L/LL5			-0.38	0.08		(22)
QUE 97008	L3.05	-0.65	0.04	-0.42	0.14	(51)	(63)
Hedjaz	L3.7-6	-0.65	0.13			(7)	
Barratta	L4			-0.10	0.19		(64)
Ausson	L5	-0.64	0.03			(52)	
Borkut	L5			0.93			
Homestead	L5			-0.40			
Renchen	L5/6	-0.81	0.15	-0.25	0.04	(57)	(57)
Renchen	L5/6	-0.64	0.08			(57)	
Bruderheim	L6			-0.26	0.19		(64)
Dar al Gani	LL3	-0.72	0.16	-0.12		(7)	(60)
Parnallee	LL3			-0.08	0.19		(64)
Krymka	LL3.2	-0.66	0.05			(52)	
Ragland	LL3.4	-0.70	0.13			(54)	
Parnallee	LL3.6	-0.59	0.09			(51)	
Chainpur	LL3-4			-0.47	0.07		(22)
GRO95552	LL4			-0.33	0.10		(63)
Soko Banja	LL4			-0.47			(60)
Paragould	LL5	-0.61	0.07			(52)	
Saint-Severin	LL6			0.38	0.19		(64)
St. Severin	LL6	-0.74	0.20	-0.41	0.10	(7)	(22)
Stubenberg	LL6	-0.67	0.09	-0.17	0.06	(58)	(58)
Stubenberg	LL6	-0.81	0.09			(58)	
Stubenberg	LL6	-0.65	0.04			(51)	
Enstatite chondrites							
ALHA77295	EH3			0.05	0.14		(63)
Kota-Kota	EH3			0.04	0.07		(22)
Kota-Kota	EH3			0.00	0.08		(26)
Qingzhen	EH3	0.02	0.12	-0.02	0.08	(7)	(22)
Qingzhen	EH3			0.00	0.05		(26)
SAH 97096	EH3			0.17	0.08		(17)
SAH 97096	EH3			-0.01	0.14		(75)
Abee	EH4	-0.06	0.04	-0.06	0.12	(52)	(22)
Abee	EH4	-0.05	0.13	-0.02	0.08	(51)	(26)
Adhi Kot	EH4	-0.10	0.04			(52)	
Indarch	EH4	-0.13	0.05	0.05	0.14	(52)	(63)
Indarch	EH4	-0.20	0.10	0.15	0.19	(51)	(64)
Saint-Sauveur	EH5	-0.15	0.09			(52)	
QUE 94204	EH7			0.21	0.08		(63)
MAC 02837	EL3	-0.40	0.12			(54)	
MAC 88136	EL3			0.02	0.09		(63)
MAC 88184	EL3			0.11	0.07		(26)
PCA 91020	EL3			0.10			
QUE 94594	EL3			0.21			
Eagle	EL6			-0.07	0.07		(26)
Hvittis	EL6	-0.29	0.07	-0.01	0.17	(52)	(22)
Jajh deh Kot Lalu	EL6	-0.29	0.10			(52)	
Khairpur	EL6			0.14	0.19		(64)
LON 94100	EL6			-0.02	0.14		(63)
Pillistfer	EL6			0.09	0.08		(22)
Angrites							
Angra dos Ries	Angrite	-1.12	0.23	-0.36	0.07	(7)	(22)
D'Orbigny	Angrite	-1.11	0.06	-0.42	0.09	(52)	(78)
LEW 86010	Angrite	-1.10	0.34			(7)	
NWA 1296	Angrite	-1.11	0.10	-0.55	0.11	(7)	(78)
NWA 2999	Angrite	-1.13	0.04	-0.53	0.25	(52)	(79)
NWA 2999	Angrite	-1.22	0.25	-0.31	0.10	(7)	(78)
NWA 4801	Angrite	-1.16	0.06	-0.35	0.06	(52)	(80)
NWA 4931	Angrite			-0.51	0.08		(78)
NWA 6291	Angrite	-1.12	0.05			(52)	
NWA 7203	Angrite			-0.35	0.18		(78)
NWA 10463	Angrite			-0.45	0.11		(78)
Sahara 99555	Angrite	-1.16	0.04	-0.43	0.13	(52)	(78)

Table S5. (continued).

Name	group	$\varepsilon^{50}Ti$	2σ	$\varepsilon^{54}Cr$	2σ	$\varepsilon^{50}Ti$ ref.	$\varepsilon^{54}Cr$ ref.
HED							
Bereba	Eucrite			-0.74	0.11		(22)
Bouvante	Eucrite	-1.23	0.16	-0.78	0.06	(7)	(22)
Cachari	Eucrite	-1.20	1.30			(55)	
Camel Donga	Eucrite	-1.34	0.09	-0.97		(52)	(60)
Ibitira	Eucrite	-1.19	0.04			(52)	
Jonzac	Eucrite			-0.69	0.10		(22)
Juvinas	Eucrite	-1.25	0.05	-0.71	0.12	(7)	(22)
Juvinas	Eucrite	-1.22	0.07	-0.55	0.19	(53)	(64)
Juvinas	Eucrite	-1.31	0.01			(53)	
Juvinas	Eucrite	-1.24	0.17			(53)	
Juvinas	Eucrite	-1.19	0.07			(51)	
Millbillillie	Eucrite	-1.22	0.11	-0.74		(52)	(60)
Padvarninkai	Eucrite			1.22			(60)
Pasamonte	Eucrite	-1.20	0.02	-0.71	0.08	(53)	(22)
Pasamonte	Eucrite	-1.26	0.09	-0.45	0.19	(53)	(64)
Pasamonte	Eucrite	-1.26	0.08	-0.45	0.19	(51)	
Stannern	Eucrite			-0.71	0.07		(22)
Johnstown	Diogenite			-0.70	0.06		(22)
Johnstown	Diogenite			-0.56	0.19		(64)
Shalka	Diogenite			-0.80	0.06		(22)
Tatahoine	Diogenite	-1.22	0.06	-0.74	0.08		(22)
Kapoeta	Howardite	-1.23	0.04	-0.71	0.19	(52)	(64)
Martian							
ALH 84001	OPX	-0.50	1.50			(55)	
ALH 84001	Shergottite	-0.39	0.05			(51)	
DaG 476	Shergottite	-0.47	0.06			(51)	
EETA 79001	Shergottite	-0.46	0.07			(51)	
NWA 2737	Shergottite	-0.30	0.17			(7)	
NWA 7034	Shergottite	-0.43	0.18	-0.23	0.10	(50)	(50)
NWA 8159	Shergottite	-0.11	0.28	-0.04	0.07	(50)	(81)
NWA 856	Shergottite	-0.36	0.17			(7)	
Shergotty	Shergottite			-0.18	0.17		(22)
Shergotty	Shergottite	-0.08	0.18	-0.18	0.17	(50)	(50)
Tissint	Shergottite	-0.68	0.18	-0.20	0.07	(50)	(50)
Zagami	Shergottite	-0.50	0.09			(51)	
Nakhla	Nakhlite			-0.14	0.08		(22)
Nakhla	Nakhlite	-0.36	0.14	-0.14	0.08	(50)	(50)
Chassigny	Chassignite	-0.32	0.10	-0.20	0.05	(51)	(22)
Chassigny	Chassignite	-0.43	0.28	-0.20	0.05	(50)	(50)
Acapulcoite-Lodranite							
Acapulco	Acapulcoite	-1.30	0.05	-0.75		(52)	(82)
Dhofar 125	Acapulcoite	-1.39	0.18			(53)	
Dhofar 125	Acapulcoite	-1.32	0.24			(51)	
Dhofar 1222	Acapulcoite			-0.34	0.25		(79)
GRV 020043	Acapulcoite	-1.59	0.24	-0.48	0.10	(50)	(83)
NWA 2714	Acapulcoite	-1.28	0.05			(51)	
NWA 8287	Acapulcoite	-1.39	0.24	-0.62	0.10	(50)	(83)
MET 01212	Acapulcoite	-1.31	0.09			(51)	
NWA 468	Lodranite	-1.54	0.42	-0.59	0.09	(50)	(83)
NWA 8118	Lodranite	-1.68	0.31	-0.59	0.09	(50)	(83)
Brachinite							
Brachina	Brachinite	-1.31	0.24	-0.36	0.11	(50)	(84)
GRA 06128	Brachinite	-1.44	0.24	-0.43	0.11	(50)	(50)
GRA 06129	Brachinite	-1.55	0.27	-0.46	0.13	(50)	(50)
NWA 3151	Brachinite	-1.11	0.37	-0.52	0.08	(50)	(84)
Ureilites							
El Gouanem	Ureilite	-0.93	0.09	-0.93	0.09		(85)
Dhofar 132	Ureilite	-0.97	0.09	-0.97	0.09		(85)
Dhofar 836	Ureilite	-0.89	0.07	-0.89	0.07		(85)
NWA 2376	Ureilite	-0.84	0.07	-0.84	0.07		(85)
DaG 340	Ureilite	-0.98	0.10	-0.98	0.10		(85)
DaG 868	Ureilite	-0.88	0.10	-0.88	0.10		(85)
ALH77257	Ureilite	-0.90	0.80	-0.65	0.22	(55)	(86)
Almahata Sitta	Ureilite			-0.77	0.10		(87)
Kenna	Ureilite			-1.08	0.25		(79)
PCA 82506	Ureilite	-1.20	1.60			(55)	
Y791538	Ureilite			-0.97	0.16		(86)
NWA 2376	Ureilite	-1.95	0.28			(7)	
NWA766	Ureilite			-0.92	0.08		(85)
NWA1241	Ureilite			-0.98	0.08		(85)
ALH 82130	Ureilite	-2.05	0.24	-0.80	0.09	(50)	(50)
ALH 81101	Ureilite	-2.24	0.44	-0.68	0.09	(50)	(50)

Table S5. (continued).

Name	group	ε^{50}Ti	2σ	ε^{54}Cr	2σ	ε^{50}Ti ref.	ε^{54}Cr ref.
Lunar							
12061				0.22	0.10		(63)
65315	anorthosite			0.12	0.04		(26)
67955	anorthosite			0.07	0.09		(26)
70017				0.08	0.12		(63)
70017	basalt			0.08	0.12		(26)
77215	norite			0.06	0.05		(26)
NWA 479	basalt	-0.03	0.13			(7)	
Earth							
AGV2	andesite	-0.07	0.12			(7)	
10PUB22-07	basalt			0.19	0.07		(26)
BCR2	basalt	0.00	0.15			(7)	
BCR2	basalt	0.00	0.13			(7)	
BCR2	basalt	0.03	0.12			(7)	
BCR2	basalt	0.01	0.07			(7)	
BCR2	basalt	-0.01	0.23			(7)	
BCR2	basalt	0.09	0.51			(7)	
BCR2	basalt	0.05	0.11			(7)	
BCR2	basalt	-0.03	0.09			(7)	
BCR2	basalt	0.04	0.03			(7)	
BCR-2	basalt	-0.04	0.15			(47)	
BCR-2	basalt	-0.03	0.17			(47)	
BCR-2	basalt	-0.02	0.26			(51)	
BE-N	basalt			0.17	0.07		(26)
BHVO_1	basalt			0.08	0.13		(63)
BHVO-2	basalt	-0.02	0.20	0.07	0.07	(51)	(26)
BIR_1	basalt			0.00	0.24		(63)
CV-SN-98-19	basalt			0.12	0.11		(26)
Deccan basalt	basalt			0.08	0.15		(22)
JB-1a	basalt			0.20	0.15		(63)
JB-1b	basalt	0.33	0.16			(59)	
KBD408729	basalt			0.14	0.07		(26)
NIST688	basalt			0.11	0.07		(26)
Tibet chromite	chromite			0.03	0.08		(22)
Tibet chromite	chromite			-0.01	0.14		(22)
DTS-1-1	dunite			0.11	0.07		(17)
DTS-1-2	dunite			0.16	0.05		(17)
DTS-1-3	dunite			0.18	0.07		(17)
DTS-1-4	dunite			0.15	0.03		(17)
DTS-1-5	dunite			0.19	0.07		(17)
BM23	peridotite			0.03	0.08		(26)
BM31	peridotite			0.13	0.08		(26)
JP-1	peridotite	0.06	0.31			(59)	
KOLA15-UB	peridotite			0.05	0.06		(26)
PCC_1	peridotite			0.13	0.08		(63)
Erta Ale tholeite	tholeiite			-0.02	0.08		(22)
ROM69				0.16	0.10		(63)

Table S6. Results for the mass balance calculation.

	$\varepsilon^{54}Cr_{fluid}$ = -19							$\varepsilon^{54}Cr_{fluid}$ = -25			$\varepsilon^{54}Cr_{fluid}$ = -15			$\varepsilon^{54}Cr_{fluid}$ = -10		
	x (%)#	y (%)#	Ti/Cr_{calc}	Mn/Cr_{calc}	$\varepsilon^{53}Cr_{calc}$	$\varepsilon^{54}Cr_{calc}$	Δ	x (%)#	y (%)*	Δ	x (%)#	y (%)*	Δ	x (%)#	y (%)*	Δ
A0106-A0107	2.8	2.3	0.167	1.14	0.232	1.18	8.9	2.6	1.6	18	3.0	3.1	6.2	3.2	4.4	12
C0108	1.8	2.9	0.174	0.950	0.182	1.24	7.4	1.7	2.1	7.0	1.9	3.6	11	2.0	5.0	26
A0106	3.2	0.8	0.144	1.24	0.255	1.45	34	3.0	0.3	53	3.4	1.2	21	3.7	2.0	6.1
C0107	2.1	1.6	0.156	0.999	0.194	1.48	15	2.0	1.2	24	2.1	1.9	10	2.2	2.4	4.7
Orgueil-1	1.2	4.0	0.191	0.810	0.147	1.10	19	1.1	3.4	12	1.2	4.5	26	1.2	5.1	42
Orgueil-2	0.6	1.8	0.158	0.713	0.120	1.69	6.8	0.6	1.6	4.6	0.7	1.9	9.1	0.7	2.0	13
Orgueil-3	0.5	1.9	0.159	0.685	0.113	1.70	7.5	0.5	1.7	4.3	0.5	2.0	11	0.5	2.1	17
Alais-1	1.2	4.2	0.194	0.818	0.149	1.05	16	1.1	3.4	9.7	1.3	5.0	24	1.3	6.3	44
Alais-2	0.6	1.2	0.149	0.715	0.120	1.83	7.5	0.6	1.2	5.4	0.6	1.2	9.3	0.6	1.1	12
Alais-3	0.6	1.3	0.151	0.708	0.118	1.81	13	0.6	1.2	9.5	0.6	1.3	16	0.6	1.2	21
Sum							135			147			144			198

Percentage of secondary mineral enrichment relative to the Ryugu sample C0002. x = dolomite, y = magnetite + pyrrhotite.

Table S7. Recovery yield of elements (%) during the SOM extraction experiment using 43.73 mg of powdered Tagish Lake.

	Na	Mg	Al	P	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn
Hexane	0.04	0.02	0.03	0.02	0.03	0.01	0.03	0.03	0.04	0.02	0.03	0.02	0.02	0.02	0.04	0.15
DCM	0.39	0.03	0.03	0.02	0.05	0.01	0.03	0.05	0.03	0.02	0.03	0.03	0.03	0.03	0.36	0.06
MeOH	8.86	0.15	0.02	0.03	0.25	0.01	0.02	0.02	0.02	0.01	0.02	0.01	0.01	0.01	0.02	0.03

Table S8. Parameters used in the mass balance calculation.

	Ti (μg/g)	Cr (μg/g)	Mn (μg/g)	$\varepsilon^{53}Cr$	$\varepsilon^{54}Cr$	Ti/Cr	Mn/Cr
Dolomite*	350	2047	49100	6.1	≦-10		
Magnetite+pyrrhotite$	3825	2525	518	0.1	≦-10		
C0002	313	2386	1411	0.1	2.2		
A0106-A0107						0.178	1.11
C0108						0.167	0.892
A0106						0.174	1.34
C0107						0.172	1.01
Orgueil-1						0.168	0.766
Orgueil-2						0.148	0.692
Orgueil-3						0.144	0.667
Alais-1						0.170	0.771
Alais-2						0.139	0.698
Alais-3						0.132	0.681

* Ti, Cr, and Mn abundances are averages of four dolomites (#4, #9, #10, #13) in Ryugu C0033 (*37*).

$ Ti, Cr, and Mn abundances are averages of three magnetite measurements (#2, #3, #5) and one pyrrhotite (#11) in Ryugu C0033 (*37*).

Table S9. CI-normalized Mn/Cr and Fe/Mn ratios in bulk samples of Ryugu and CI chondrites.

Samples	$(Mn/Cr)_N$			$(Fe/Mn)_N$		
Ryugu						
A0106-A0107	1.418	±	0.018	0.664	±	0.011
C0108	1.112	±	0.017	0.829	±	0.013
A0106	1.835	±	0.006	0.577	±	0.012
C0107	1.392	±	0.004	0.800	±	0.016
C0002	0.818	±	0.003	1.170	±	0.026
CI chondrites						
Orgueil-1	1.015	±	0.025	1.028	±	0.024
Orgueil-2	0.953	±	0.020	1.020	±	0.017
Orgueil-3	0.933	±	0.004	1.057	±	0.019
Alais-1	0.982	±	0.023	1.016	±	0.041
Alais-2	0.962	±	0.020	0.969	±	0.022
Alais-3	0.944	±	0.003	0.991	±	0.020

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