

Supplementary Materials for

The Ni isotopic composition of Ryugu reveals a common accretion region for carbonaceous chondrites

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Sci. Adv. **10**, eadp2426 (2024)
DOI: 10.1126/sciadv.adp2426

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Supplementary Text

Ni isotopic composition of CAIs

The $\mu^{54}\text{Fe}$ and $\mu^{60}\text{Ni}$ values of most CAIs investigated so far are similar to those of their host chondrites, indicating that the original Fe and Ni isotope signatures of the CAIs were modified by alteration on the carbonaceous chondrite parent bodies (68, 81), most likely by oxidation and dissolution of the metal (82). Having this issue in mind, previous studies argued that the high-Fe pyroxene mineral separate from the Egg 2 CAI from Allende represents the most pristine Fe and Ni composition for CAIs because of its low FeO content, magnetic susceptibility, and CAI-like Ti and Sr isotopic composition (68, 81). The same conclusion has also been drawn based on Ni isotope measurements on CAIs separated from the reduced CV chondrite Efremovka (29), which was subject to less severe alteration than Allende (83). All these CAIs display large mass-dependent isotope fractionations, which can potentially result in residual spurious mass-independent isotope effects when using the exponential law for mass bias correction (33). To account for this effect, the measured isotope anomalies were corrected using the approach of Tang and Dauphas (2012). Table S5 summarizes the Fe and Ni isotope data of the CAIs which show little effects of parent body alteration. These CAIs have been used to determine the current best estimate of the Fe and Ni isotope composition of CAIs used in this study.

Ni isotopic variations among Ryugu, CI and other CC chondrites

Given that the number of analyzed samples is small, a Welch's t-test was conducted to evaluate whether there are statistically significant Ni isotopic differences between CI chondrites/ Ryugu ($n = 5$; Ryugu A, Ryugu C, Orgueil, Alais, Ivuna) and other carbonaceous chondrites ($n = 8$; CM, CO, CV, CR, CH, CB, TL, TD). The Welch's t-test is generally applied when there is a difference between the variations of two populations and also when their sample sizes are unequal. The designated null hypothesis (H_0) is that the average $\mu^i\text{Ni}$ values of both populations are equal, where i stand for 60, 62, or 64 when normalized to $^{61}\text{Ni}/^{58}\text{Ni}$. If the two-tailed P value is less than the defined significance threshold (α) of 0.05, then H_0 can be rejected at 95% confidence. We find that the Welch's t-test yields two-tailed P values <0.05 for all three $\mu^i\text{Ni}$ values, indicating that we can confidently reject the null hypothesis (H_0) and conclude that CI chondrites/ Ryugu are isotopically distinct from the other carbonaceous chondrites.

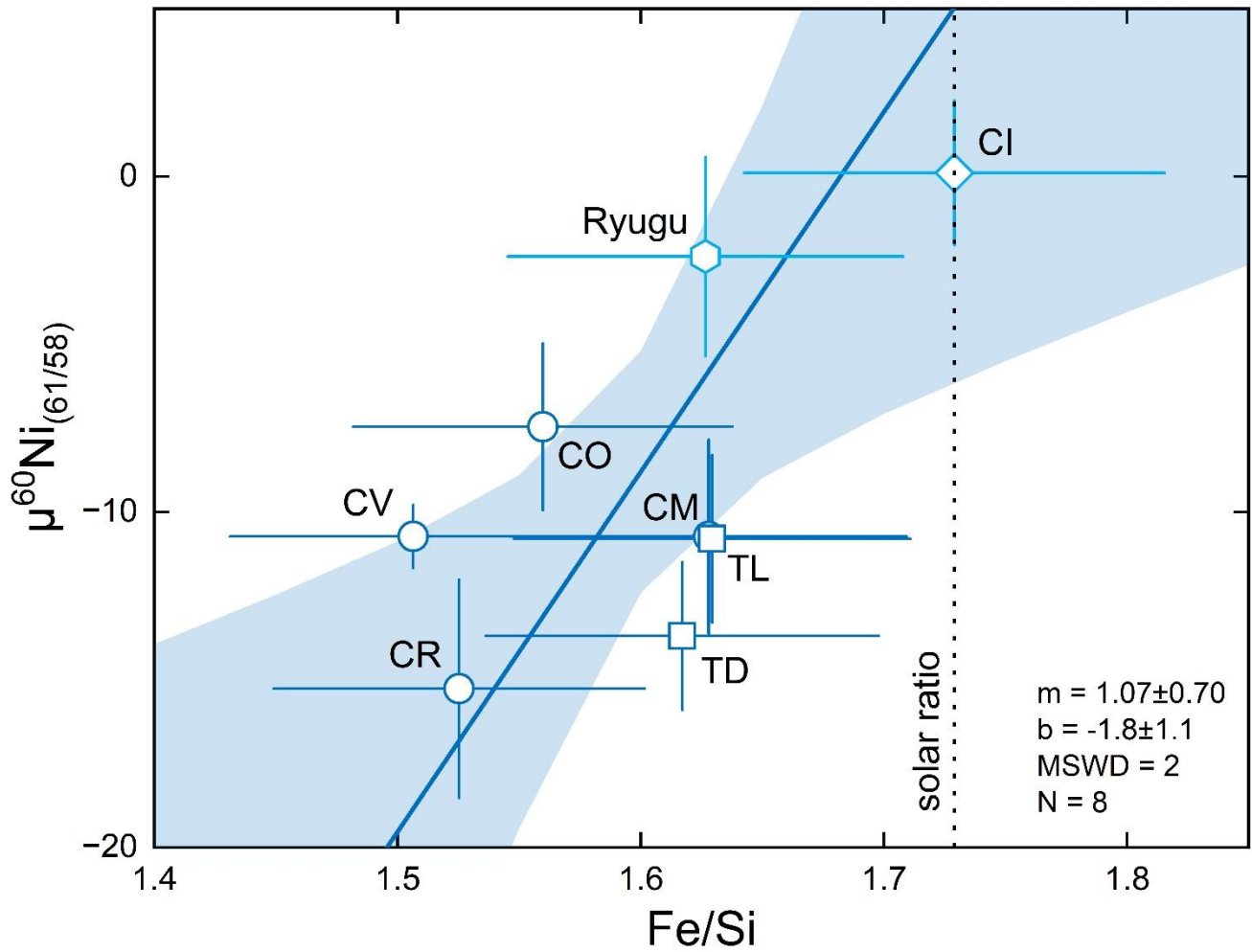


Fig. S1. Diagram of $\mu^{60}\text{Ni}_{(61/58)}$ versus Fe/Si mass ratios of carbonaceous chondrites. The weak correlation suggests that the observed ~15% variations in bulk Fe/Si ratios among carbonaceous chondrites may indeed be directly related to the variation in $\mu^{60}\text{Ni}_{61/58}$, which we attribute to abundance variations of ~5 wt% of FeNi metal grains among their parent bodies.

Table S1. Ni isotopic data of samples investigated in this study. Uncertainties of individual samples represent two standard errors (2 s.e.), where N is the number of measurements. Data is internally normalized to either $^{61}\text{Ni}/^{58}\text{Ni} = 0.016744$ or $^{62}\text{Ni}/^{61}\text{Ni} = 3.1884$. The chemical procedure used at the Tokyo Institute of Technology is described in the methods and (84). Matrix aliquots containing the bulk Ni of samples processed at the Institut für Planetologie Münster (IfP) were digested HF-HNO₃-HCl-HClO₄ mixtures and then separated from Sr or W using cation (AG50W-X8) or anion exchange (AG1-X8) exchange resin, respectively. All samples, regardless of their processing at the Tokyo Institute of Technology or Institut für Planetologie Münster were further processed at IfP to purify Ni.

Sample	Class	Mass digested (g)	Mass homogenized (g)	Processed?	N	Norm. $^{61}\text{Ni}/^{58}\text{Ni}$			Norm. $^{62}\text{Ni}/^{61}\text{Ni}$		
						$\mu^{60}\text{Ni}$	$\mu^{62}\text{Ni}$	$\mu^{64}\text{Ni}$	$\mu^{58}\text{Ni}$	$\mu^{60}\text{Ni}$	$\mu^{64}\text{Ni}$
Ryugu samples											
A0106–A0107		0.0239	0.0289	Fe, U (Tokyo Tech)	24	-4 ± 3	19 ± 6	48 ± 10	58 ± 18	15 ± 9	-7 ± 12
A0106		0.0146	0.0175	Fe, U (Tokyo Tech)	20	-2 ± 4	17 ± 8	51 ± 14	51 ± 24	14 ± 11	2 ± 15
C0108		0.0222	0.0333	Fe, U (Tokyo Tech)	24	-3 ± 3	27 ± 5	76 ± 11	82 ± 17	24 ± 8	-3 ± 12
C0107		0.0128	0.0174	Fe, U (Tokyo Tech)	20	0 ± 4	25 ± 7	66 ± 8	78 ± 22	26 ± 11	-8 ± 16
Carbonaceous chondrites											
Orgueil	CI1	0.401	1.12	Sr matrix cut (IfP)	37	-3 ± 3	15 ± 4	43 ± 7	46 ± 14	12 ± 7	-1 ± 10
Orgueil (JAXA)	CI1	0.02	0.05	Fe, U (Tokyo Tech)	18	2 ± 4	22 ± 7	60 ± 13	70 ± 23	25 ± 11	-6 ± 16
Alais (JAXA)	CI1	0.022	0.051	Fe, U (Tokyo Tech)	20	2 ± 3	27 ± 7	82 ± 10	85 ± 20	30 ± 10	1 ± 13
Ivuna	CI1	0.05	0.1118	Te chemistry cut (MPS)	22	-1 ± 2	22 ± 3	62 ± 7	68 ± 11	22 ± 5	-3 ± 7
Murchison (JAXA)	CM2	0.025	1.65	Fe, U (Tokyo Tech)	16	-14 ± 4	11 ± 8	43 ± 15	35 ± 24	-2 ± 12	10 ± 17
MET 01070	CM1	0.00457	1.7667	Ni (IfP)	16	-10 ± 2	10 ± 6	26 ± 11	32 ± 18	0 ± 7	-4 ± 11
NWA 6015	CO3	0.00487	>2	Ni (IfP)	16	-7 ± 3	11 ± 7	22 ± 12	33 ± 21	4 ± 10	-10 ± 11
NWA 5933	CO3	0.00486	3.239	Ni (IfP)	16	-9 ± 3	4 ± 8	18 ± 16	12 ± 24	-6 ± 11	7 ± 14
DaG 136	CO3	0.00648	1.83	Ni (IfP)	16	-6 ± 4	13 ± 8	28 ± 12	41 ± 23	8 ± 11	-11 ± 14
Allende (JAXA)	CV3	0.025	~4000 (USNM)	Fe, U (Tokyo Tech)	16	-10 ± 4	15 ± 10	36 ± 17	46 ± 30	5 ± 14	-8 ± 15
GRA 06100	CR1	0.281	0.281	Sr matrix cut (IfP)	16	-17 ± 5	8 ± 10	27 ± 13	26 ± 31	-8 ± 15	2 ± 23
Acfer 139	CR2	0.00538	2.09	Ni (IfP)	18	-16 ± 3	14 ± 7	35 ± 13	43 ± 21	-2 ± 10	-6 ± 12
Acfer 182	CH3	0.553	0.553	W matrix cut (IfP)	12	-16 ± 5	12 ± 11	28 ± 22	38 ± 35	-4 ± 16	-9 ± 17
Tagish Lake (JAXA)	C2-ung	0.025	1.06	Fe, U (Tokyo Tech)	16	-11 ± 4	16 ± 8	44 ± 14	50 ± 25	5 ± 12	-3 ± 13
Tagish Lake	C2-ung	0.486	1.5	Sr matrix cut (IfP)	17	-11 ± 3	13 ± 7	33 ± 11	42 ± 22	3 ± 10	-7 ± 13
Tarda (JAXA)	C2-ung	0.025	0.212	Fe, U (Tokyo Tech)	16	-14 ± 3	19 ± 5	52 ± 15	60 ± 17	5 ± 8	-6 ± 12
Tarda	C2-ung	0.00619	~0.5	Ni (IfP)	19	-13 ± 3	10 ± 8	24 ± 13	31 ± 25	-3 ± 11	-6 ± 16

Table S2. Ni isotopic data for NIST SRM 361 compared to the literature.

Sample	N	Norm. $^{61}\text{Ni}/^{58}\text{Ni}$			Norm. $^{62}\text{Ni}/^{61}\text{Ni}$		
		$\mu^{60}\text{Ni}$ ($\pm 2\text{s.e.}$)	$\mu^{62}\text{Ni}$ ($\pm 2\text{s.e.}$)	$\mu^{64}\text{Ni}$ ($\pm 2\text{s.e.}$)	$\mu^{58}\text{Ni}$ ($\pm 2\text{s.e.}$)	$\mu^{60}\text{Ni}$ ($\pm 2\text{s.e.}$)	$\mu^{64}\text{Ni}$ ($\pm 2\text{s.e.}$)
NIST 361 #1	37	-1 \pm 2	-1 \pm 4	1 \pm 8	-4 \pm 12	-2 \pm 6	4 \pm 8
NIST 361 #2	21	-2 \pm 3	7 \pm 5	26 \pm 8	22 \pm 15	5 \pm 7	4 \pm 11
NIST 361 #3	10	2 \pm 5	8 \pm 8	18 \pm 17	24 \pm 26	10 \pm 12	-5 \pm 17
NIST 361 #4	12	-1 \pm 4	6 \pm 8	18 \pm 13	20 \pm 24	5 \pm 12	-1 \pm 13
NIST 361 #5	8	3 \pm 9	11 \pm 16	16 \pm 19	35 \pm 48	15 \pm 25	-17 \pm 29
Average (± 2 s.d.)	5	0 \pm 5	6 \pm 9	16 \pm 18	19 \pm 28	7 \pm 13	-3 \pm 18
Average literature* (± 2 s.d.)	5	-1 \pm 1	5 \pm 11	8 \pm 15	16 \pm 35	4 \pm 12	-7 \pm 32
Bulk silicate Earth [†]		-1 \pm 1	4 \pm 1	12 \pm 2	11 \pm 3	3 \pm 1	1 \pm 3

Data sources: *(30–32, 34) and [†](34).

Table S4. Average Ni isotopic composition of meteoritic materials. Uncertainties are 2 standard deviations (2s.d.) for $N \leq 3$ and 95% confidence intervals (95% CI) for $N \geq 4$, where N is the number of samples. NC and CC stand for non-carbonaceous (inner disk) and carbonaceous (outer disk) reservoir, respectively. Data is internally normalized to either $^{61}\text{Ni}/^{58}\text{Ni} = 0.016744$ or $^{62}\text{Ni}/^{61}\text{Ni} = 3.1884$. Data sources: (28–34, 66, 87, 88).

Sample	Reservoir	N	Norm. $^{61}\text{Ni}/^{58}\text{Ni}$			Norm. $^{62}\text{Ni}/^{61}\text{Ni}$		
			$\mu^{60}\text{Ni}$	$\mu^{62}\text{Ni}$	$\mu^{64}\text{Ni}$	$\mu^{58}\text{Ni}$	$\mu^{60}\text{Ni}$	$\mu^{64}\text{Ni}$
Hayabusa2								
Ryugu A	CC	2	-3 ± 3	18 ± 3	50 ± 4	55 ± 10	15 ± 1	-2 ± 13
Ryugu C	CC	2	-1 ± 5	26 ± 2	71 ± 14	80 ± 7	25 ± 2	-6 ± 7
Ryugu mean	CC	4	-2 ± 3	22 ± 8	60 ± 20	67 ± 24	20 ± 9	-4 ± 8
Chondrites								
CI	CC	6	0 ± 2	23 ± 4	68 ± 7	72 ± 11	24 ± 5	-2 ± 7
CM	CC	8	-11 ± 3	11 ± 1	35 ± 18	33 ± 3	0 ± 3	3 ± 16
CO	CC	4	-7 ± 2	9 ± 6	24 ± 7	29 ± 20	2 ± 9	-4 ± 13
CV	CC	9	-11 ± 1	10 ± 4	27 ± 12	29 ± 14	-3 ± 4	-4 ± 3
CR	CC	4	-15 ± 3	9 ± 8	28 ± 15	28 ± 24	-6 ± 6	1 ± 9
CH	CC	1	-16 ± 5	12 ± 11	28 ± 22	38 ± 35	-4 ± 16	-9 ± 17
CB	CC	1	-19 ± 2	16 ± 5		49 ± 14	-3 ± 5	
Tagish Lake	CC	2	-11 ± 3	15 ± 5	37 ± 8	45 ± 16	4 ± 8	-5 ± 9
Tarda	CC	2	-14 ± 2	16 ± 4	36 ± 10	51 ± 13	3 ± 6	-6 ± 9
EH	NC	4	-2 ± 3	3 ± 2	13 ± 13	8 ± 9	1 ± 3	7 ± 24
EL	NC	3	0 ± 7	-3 ± 7	-4 ± 5	-8 ± 23	-6 ± 10	3 ± 27
OC	NC	16	-5 ± 1	-6 ± 2	-16 ± 4	-19 ± 6	-11 ± 2	2 ± 3
R	NC	1	-7 ± 2	-5 ± 3	-8 ± 10	-16 ± 8	-13 ± 3	7 ± 11
Achondrites/ Iron meteorites								
HED	NC	9	-10 ± 13	3 ± 10		11 ± 27	-7 ± 15	
Ureilites	NC	2	2 ± 8	-5 ± 16		-14 ± 48	-3 ± 7	
Angrites	NC	15	-2 ± 9	2 ± 4		5 ± 10	4 ± 13	
Aubrites	NC	1	8 ± 8	5 ± 19		16 ± 52	13 ± 19	
Main group pallasites	NC	1	-2 ± 6	-6 ± 10	-13 ± 19	-20 ± 27	-8 ± 10	6 ± 30
NWA 5363/5400	NC	1	0 ± 2	1 ± 3	4 ± 8	3 ± 8	1 ± 3	1 ± 10
IAB	NC	6	-3 ± 3	-5 ± 6	-8 ± 7	-16 ± 19	-9 ± 9	7 ± 12
IC	NC	7	-5 ± 1	-5 ± 5	-21 ± 13	-17 ± 16	-10 ± 6	-5 ± 4
IIAB	NC	2	-4 ± 1	-9 ± 9	-29 ± 24	-29 ± 28	-13 ± 8	-2 ± 3
IIIAB	NC	3	-6 ± 3	-11 ± 8	-35 ± 8	-42 ± 3	-20 ± 4	1 ± 6
IIIE	NC	5	-6 ± 2	-7 ± 3	-25 ± 13	-22 ± 11	-14 ± 4	-5 ± 5
IVA	NC	5	-6 ± 2	-9 ± 3	-29 ± 14	-27 ± 9	-15 ± 2	-3 ± 6
IIC	CC	4	-16 ± 4	15 ± 11	36 ± 33	47 ± 35	-1 ± 14	-9 ± 10
IID	CC	10	-11 ± 2	9 ± 8	23 ± 14	29 ± 25	-1 ± 10	-5 ± 15
IIF	CC	1	-9 ± 1	9 ± 4	17 ± 11	28 ± 11	0 ± 4	-10 ± 14
IIIF	CC	3	-11 ± 4	12 ± 7	25 ± 18	36 ± 22	1 ± 3	-10 ± 3
IVB	CC	21	-13 ± 1	7 ± 3	24 ± 5	23 ± 8	-6 ± 4	2 ± 5
Wiley (IIC/ungr.)	CC	1	-17 ± 3	13 ± 5	34 ± 10	40 ± 14	-4 ± 5	-4 ± 15
Earth's mantle		2	-1 ± 1	4 ± 1	12 ± 2	11 ± 3	3 ± 1	1 ± 3
Mars' mantle		5	-1 ± 1	4 ± 3		12 ± 9	3 ± 3	

Table S5. Fe and Ni isotopic data for selected CAIs. Uncertainties represent those reported in the literature and 95% confidence intervals (95% CI) for the calculated CAI mean. Data sources: (29, 68, 81, 89).

Sample	Host chondrite	$\delta^{56}\text{Fe}$	Norm. $^{57}\text{Fe}/^{56}\text{Fe}$		$\delta^{60}\text{Ni}$	Norm. $^{61}\text{Ni}/^{58}\text{Ni}$			Norm. $^{62}\text{Ni}/^{61}\text{Ni}$		
			$\mu^{54}\text{Fe}$	$\mu^{58}\text{Fe}$		$\mu^{60}\text{Ni}$	$\mu^{62}\text{Ni}$	$\mu^{64}\text{Ni}$	$\mu^{58}\text{Ni}$	$\mu^{60}\text{Ni}$	$\mu^{64}\text{Ni}$
Egg-2	CV3 _{ox} Allende	9.8 ± 1.1	-560 ± 450	3400 ± 1200	4.15 ± 0.06	53 ± 6	143 ± 11	336 ± 21	443 ± 30	199 ± 11	-86 ± 33
Egg 2 high-Fe Px	CV3 _{ox} Allende	9.86 ± 0.04	-581 ± 58	324 ± 254	4.24 ± 0.06	60 ± 8	169 ± 18	389 ± 60	525 ± 49	232 ± 18	-110 ± 68
31E-1	CV3 _{red} Efremovka				1.33 ± 0.02	70 ± 2	159 ± 6	400 ± 13	493 ± 15	232 ± 5	-70 ± 18
31E-2	CV3 _{red} Efremovka				1.77 ± 0.03	68 ± 7	161 ± 12	428 ± 11	500 ± 33	232 ± 12	-49 ± 32
31E-4	CV3 _{red} Efremovka				1.27 ± 0.04	56 ± 2	130 ± 5	349 ± 9	402 ± 14	189 ± 5	-35 ± 15
CAI mean			-581 ± 56			61 ± 9	152 ± 20	380 ± 47	473 ± 61	217 ± 27	-70 ± 37

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