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1	Chromium isotopic insights into the origin of chondrite
2	parent bodies and the early terrestrial volatile depletion
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4	Ke Zhu (朱柯) ^{1*} , Frédéric Moynier ¹ , Martin Schiller ² , Conel M. O'D. Alexander ³ ,
5	Jemma Davidson ⁴ , Devin L. Schrader ⁴ , Elishevah van Kooten ^{1,2} , Martin Bizzarro ^{1,2}
6	
7	¹ Université de Paris, Institut de Physique du Globe de Paris, CNRS UMR 7154, 1 rue
8	Jussieu, Paris 75005, France
9	² Centre for Star and Planet Formation, Globe Institute, University of Copenhagen,
10	Øster Voldgade 5–7, Copenhagen DK-1350, Denmark
11	³ Earth and Planetary Laboratory, Carnegie Institution for Science, 5241 Broad Branch
12	Road, Washington, DC 20015, USA
13	⁴ Center for Meteorite Studies, School of Earth and Space Exploration, Arizona State
14	University, 781 East Terrace Road, Tempe, AZ 85287-6004, USA
15	
16	*corresponding author: zhu@ipgp.fr
17	
18	
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21 Abstract:

22 Chondrites are meteorites from undifferentiated parent bodies that provide fundamental information about early Solar System evolution and planet formation. The 23 24 element Cr is highly suitable for deciphering both the timing of formation and the 25 origin of planetary building blocks because it records both radiogenic contributions from ⁵³Mn-⁵³Cr decay and variable nucleosynthetic contributions from the stable ⁵⁴Cr 26 27 nuclide. Here, we report high-precision measurements of the mass-independent Cr isotope compositions (ε^{53} Cr and ε^{54} Cr) of chondrites (including all carbonaceous 28 29 chondrites groups) and terrestrial samples using for the first time a multi-collection 30 inductively-coupled-plasma mass-spectrometer to better understand the formation 31 histories and genetic relationships between chondrite parent bodies. With our comprehensive dataset, the order of decreasing ε^{54} Cr (per ten thousand deviation of the 32 ⁵⁴Cr/⁵²Cr ratio relative to a terrestrial standard) values amongst the carbonaceous 33 chondrites is updated to $CI = CH \ge CB \ge CR \ge CM \approx CV \approx CO \ge CK > EC > OC$. 34 Resolvable ε^{54} Cr (with 2SE uncertainty) differences between CV and CK chondrites 35 36 rule out an origin from a common parent body or reservoir as has previously been suggested. The CM and CO chondrites share common ε^{54} Cr characteristics, which 37 38 suggests their parent bodies may have accreted their components in similar proportions. The CB and CH chondrites have low-Mn/Cr ratios and similar ε^{53} Cr values to the CI 39 chondrites, invalidating them as anchors for a bulk Mn-Cr isochron for carbonaceous 40

41	chondrites. Bulk Earth has a ε^{53} Cr value that is lower than the average of chondrites,
42	including enstatite chondrites. This depletion may constrain the timing of volatile loss
43	from the Earth or its precursors to be within the first million years of Solar System
44	formation and is incompatible with Earth's accretion via any of the known chondrite
45	groups as main contributors, including enstatite chondrites.
46	

- 47 Key words: Chondrites; genetic relationship, ⁵⁴Cr systematics; CV-CK, CH-CB and
- 48 CO-CM clans; CV subgroups; ⁵³Mn-⁵³Cr chronometry; condensation history; volatile
- 49 depletion; Early Earth; Solar System.

50 1. Introduction

51 Chondrites are the oldest cosmic sedimentary rocks, the most pristine of which preserve information about the origin of the Solar System (e.g., Krot et al., 2014). Most 52 53 of the parent bodies of chondrites did not undergo significant melting (though some 54 underwent varying degrees of thermal metamorphism) and thus did not differentiate. 55 Therefore, bulk chondrite samples have chemical compositions that are thought to be 56 representative of the bulk parent body and have been taken as proxies for the 57 composition of bulk differentiated planets such as the Earth (Allègre et al., 1995). Thus, 58 investigating the chemical and isotopic compositions of chondrites is central to better understand the evolution of the Solar System and planet formation. Chondrites also 59 60 record large chemical and isotopic variations amongst them (Alexander, 2019a, b; 61 Braukmüller et al., 2018; Hellmann et al., 2020; Palme and O'Neill, 2014). In particular, 62 O and Cr isotopes have been key in evaluating genetic links between meteorites and 63 planets (e.g., Clayton and Mayeda, 1999; Qin et al., 2010; Trinquier et al., 2007; 64 Warren, 2011). Although there are several studies utilizing Cr isotopes in this manner, 65 the kinship between different groups of carbonaceous chondrites is still debated. The possible genetic links between the Vigarano-type (CV) and Karoonda-type (CK) 66 67 chondrites (Dunn and Gross, 2017; Dunn et al., 2016; Greenwood et al., 2010), as well 68 as those of chondrites in Ornans-type (CO) and Mighei-type (CM) groups (e.g.,

69 Schrader and Davidson, 2017) and Bencubbin-type (CB) and high-metal (CH) groups
70 are still unresolved (e.g., Krot et al., 2014).

71 The Cr isotope system may be one of the best suited isotopic systems for studying 72 the timing of chondrite formation and the genetic relationships between meteorite 73 groups (Birck and Allègre, 1988; Trinquier et al., 2007). The short-lived radionuclide 74 ⁵³Mn, with a half-life of 3.7 ± 0.2 Myrs (Holden, 1990), decays to ⁵³Cr and was present in the early Solar System (Lugmair and Shukolyukov, 1998). Therefore, the ⁵³Mn-⁵³Cr 75 76 decay system is a useful chronometer to date early Solar System events such as 77 chondrule (precursor) formation (Nyquist et al., 2001; Yamashita et al., 2010; Zhu et al., 78 2019a; Zhu et al., 2020a), differentiation of planets/asteroids (Lugmair and 79 Shukolyukov, 1998; Trinquier et al., 2008b; Wadhwa et al., 2003; Yamakawa et al., 80 2010; Zhu et al., 2019b; Zhu et al., 2020b), as well as the aqueous alteration (Fujiya et 81 al., 2012; Fujiya et al., 2013) and metamorphic processes in chondrite parent bodies 82 (Trinquier et al., 2008b; Göpel et al., 2015). Furthermore, previous Cr isotope 83 measurements have suggested that the various carbonaceous chondrite (CC) groups define a bulk isochron with a slope defining an initial 53 Mn/ 55 Mn ratio of (8.5 ± 1.5) × 84 10^{-6} , which was used to suggest that a volatile fractionation in the solar nebular 85 86 occurred as early as 4568.6 ± 1.1 Ma (Shukolyukov and Lugmair 2006, Moynier et al. 87 2007) when the date is anchored to the U isotope corrected age of the D'Orbigny angrite (Amelin, 2008; Brennecka and Wadhwa, 2012; Glavin et al., 2004). This 88

89	whole-rock Mn-Cr isochron, reported in Shukolyukov and Lugmair (2006), is
90	primarily controlled by two extreme endmembers representing the Ivuna-type
91	carbonaceous (CI; with the highest ⁵⁵ Mn/ ⁵² Cr ratio) and the CB chondrites (with the
92	lowest ⁵⁵ Mn/ ⁵² Cr ratio). Based on more recent measurements of the Cr isotopes in CI
93	chondrites (Qin et al., 2010; Trinquier et al., 2008b), Qin et al. (2010) suggested a
94	lower slope of $(5.4 \pm 2.4) \times 10^{-6}$ and a correspondingly younger age of 4566.1 ± 2.4 Ma
95	age for the Mn/Cr fractionation (also anchored to U-corrected D'Orbigny), although
96	this younger age is still consistent within uncertainty with that of Shukolyukov and
97	Lugmair (2006). Finally, the slope of the isochron was later updated to $[(6.2 \pm 1.9) \times$
98	10 ⁻⁶] (Göpel et al., 2015). However, all of these isochrons are mostly controlled by the
99	data point for one CB chondrite, Hammadah al Hamra (HaH) 237 (Shukolyukov and
100	Lugmair, 2006) and to date no Cr isotope data for the CH chondrites, which have
101	comparably low Mn/Cr ratios (Lodders et al., 1998), have been reported. Therefore,
102	providing high-precision Cr isotope data for CB and CH chondrites is critical to further
103	evaluate the timing of Mn/Cr fractionation in the early Solar System.
104	Variable ⁵⁴ Cr nucleosynthetic anomalies, expressed as ε^{54} Cr (the parts per 10,000

105 deviation of the mass fractionation corrected ${}^{54}Cr/{}^{52}Cr$ ratio from a terrestrial standard) 106 have been used as a tracer of potential genetic relationships between Solar System 107 materials (Trinquier et al., 2007) including between Earth and Moon (Mougel et al. 108 2018). The published $\varepsilon^{54}Cr$ values for several chondrite groups with the mean values

109	following the sequence: CI > CB \ge CR \ge CH \ge CM > CV \ge CO \ge CK > EC (Enstatite
110	Chondrites) \approx RC (Rumuruti Chondrites) > OC (Ordinary Chondrites) (Göpel et al.,
111	2015; Mougel et al., 2018; Pedersen et al., 2019; Qin et al., 2010; Shukolyukov and
112	Lugmair, 2006; Trinquier et al., 2007; Van Kooten et al., 2016; Zhu et al., 2021). These
113	isotopic differences between chondrites have been compared to the composition of the
114	Earth's mantle (Mougel et al. 2018) to detect and identify the likely sources of the
115	impact-related extra-terrestrial materials included in terrestrial rocks (e.g., Koeberl et
116	al., 2007; Magna et al., 2017; Mougel et al., 2017; Mougel et al., 2019; Schmitz et al.,
117	2016; Trinquier et al., 2006) and to test magma ocean models for asteroids (e.g., Zhu et
118	al., 2019b; Zhu et al., 2020b). To date, the most ⁵⁴ Cr-rich phases analyzed in chondrites
119	are 10s to 100s of nanometer-size presolar spinel grains that can be concentrated in acid
120	residues (e.g., Podosek et al., 1997; Rotaru et al., 1992) and have been identified by
121	NanoSIMS (Dauphas et al., 2010; Nittler et al., 2018; Qin et al., 2011). These grains
122	probably formed in the ejecta of one or more supernova.

However, although Cr-isotopic homogeneity within each chondrite group is generally assumed, this assertion is only based on a limited number of measurements, often of the same meteorites. In several instances, when two or more meteorites from the same group have been analyzed their ε^{54} Cr values differ by more than the reported uncertainties of the measurements, including: Renazzo-type carbonaceous (CR) (1.06 ± 0.08 to 1.32 ± 0.11 [2SE] for Northwest Africa [NWA] 7837 and Graves Nunataks

129	[GRA] 06100); CO (0.57 ± 0.11 ~ 0.87 ± 0.18, Lancé and Kainsaz); CK (0.33 ± 0.12 to
130	0.63 ± 0.09 for Elephant Moraine [EET] 92002 and Karoonda) and CV (0.71 ± 0.15 to
131	1.10 ± 0.08 , for Leoville and Allende) (Qin et al., 2010; Trinquier et al., 2007; Zhu et al.,
132	2020b). Hence, a more comprehensive Cr isotope dataset is required to better
133	understand the extent of the ϵ^{54} Cr variability within and between chondrite groups and
134	to better determine the ϵ^{54} Cr sequence for chondrites, especially for the CH and CB
135	chondrites, which lack systematic $\epsilon^{54} \text{Cr}$ studies. Additionally, the CK and CV
136	chondrites share many similar features, including: chondrule sizes and abundances
137	(Weisberg et al., 2006), petrological and chemical compositions (Isa et al., 2014),
138	cosmic-ray exposure ages (Scherer and Schultz, 2000), O-isotope compositions
139	(Greenwood et al., 2010) and Ti isotopic anomalies (Trinquier et al., 2009; Zhang et al.,
140	2012) and are considered a clan (the CV-CK clan). Different models have been
141	proposed for their origins, including the single parent body hypothesis where CV and
142	CK chondrites originate from different depths within the same parent body, with the
143	CK3 to CK6 petrologic types at progressively greater depths (Greenwood et al., 2010).
144	This common origin for CV and CK chondrite groups can be tested using the $\epsilon^{54} Cr$
145	systematics. ϵ^{54} Cr can also be used to track the relationships between chondrites in
146	CM-CO (e.g., Schrader and Davidson, 2017) and CB-CH clans (e.g., Krot et al., 2014).
147	The mass-independent Cr isotope compositions of meteorites are traditionally
148	measured by thermal ionization mass spectrometry (TIMS) (Birck and Allègre, 1988;

149	Lugmair and Shukolyukov, 1998; Qin et al., 2010; Shukolyukov and Lugmair, 2006;
150	Trinquier et al., 2007; Trinquier et al., 2008a). However, it appears that there are
151	small residual mass-dependent fractionations that cannot be corrected for, which are
152	evident in the correlation of ϵ^{53} Cr and ϵ^{54} Cr for multiple measurements of standards
153	with a slope of ~2 (Bourdon and Fitoussi, 2020; Qin et al., 2010; Trinquier et al., 2006).
154	It has been suggested that such residual mass-dependent isotopic fractionations could
155	potentially arise from isotopic fractionation between different oxidized Cr gas species
156	during evaporation from the filaments during TIMS analysis (Bourdon and Fitoussi,
157	2020). This would mimic equilibrium isotope fractionation that occurred as Cr
158	evaporated during the formation of bodies such as the Moon and the asteroid 4 Vesta
159	(Sossi et al., 2018; Zhu et al., 2019c). Moreover, the column chemistry in some
160	previous studies can only reach a Cr yield of ~80% (e.g., Qin et al., 2010; Trinquier et
161	al., 2008a; Trinquier et al., 2008b; Zhu et al., 2019a), and sometimes the yield can be as
162	low as ~60% (Kruijer et al., 2020; Sossi et al., 2018; Zhu et al., 2019c). However, the
163	equilibrium Cr stable isotope fractionation on the column cannot be fully corrected if
164	the yield is low [e.g., < 70%; (Larsen et al., 2016; Qin et al., 2010; Trinquier et al.,
165	2008a)], since the different Cr cuts from the columns show mass-independent
166	fractionation with ϵ^{53} Cr ranging from -0.2 to +0.2 and ϵ^{54} Cr ranging from -0.5 to +0.4
167	(Trinquier et al., 2008a). In order to avoid this problem, we have utilized a high-yield
168	(~95%) four-step column chemistry and employ multiple-collector

169 inductively-coupled-plasma mass-spectrometry (MC-ICP-MS) to measure the mass-independent fractionation of Cr isotopes for a self-consistent comprehensive set 170 of chondrite group compositions, including CI, CB (both CBa and CBb subgroups), CH, 171 172 CR, CM, CV (including the oxidized, oxA after Allende and oxB after Bali, and 173 reduced, Red after Vigarano, subgroups), CO, CK and high-Fe enstatite (EH) chondrite 174 groups (Pedersen et al., 2019; Qin et al., 2010; Trinquier et al., 2007; Trinquier et al., 175 2008b; Zhu et al., 2021). This study aims to better constrain the genetic relationship 176 between chondrite parent bodies, chondrite parent body processes (redox, thermal metamorphism and aqueous alteration), the ⁵³Mn-⁵³Cr "isochron" for CCs and other 177 178 chondrites, and the radiogenic Cr isotopic deficits between chondrites and Earth.

179 **2. Samples and methods**

180 **2.1 Samples and digestion**

The sample suite analyzed in this study includes: one CI1 chondrite (Orgueil), three CB3 chondrites (Miller Range [MIL] 05082, Quebrada Chimborazo [QC] 001 and Hammadah al Hamra [HaH] 237 consisting of mostly silicate), two CH3 chondrites (Pecora Escarpment [PCA] 91467 and Asuka [A] 881020), two CR chondrites (Grosvenor Mountains [GRO] 95577 [CR1] and Al Rais an anomalous [CR2]), five CM chondrites (Scott Glacier [SCO] 06043 [CM1], Nogoya [CM2], Banten [CM2], Jbilet Winselwan [CM2] and Aguas Zarcas, a new CM2 fall from 2019),

188	three CO3 chondrites (Ornans, MIL 07193 and Dominion Range [DOM] 10104), one
189	CV3 _{oxA} chondrite (Allende), three CV3 _{oxB} chondrites (Bali, Mokoia, and Kaba), two
190	CV3 _{red} chondrites (Leoville and Vigarano), four CK chondrites (Allan Hills [ALH]
191	85002 [CK4], Karoonda [CK4], Elephant Moraine [EET] 92002 [CK5] and Lewis
192	Cliff [LEW] 87009 [CK6]), and one enstatite chondrite, Sahara 97096 [EH3] that is
193	from Zhu et al. (2020a). We note that, CB and CH chondrites are highly heterogeneous
194	due to their metal-rich nature (Krot et al., 2014) and that the samples analyzed here
195	(<100 mg) may not represent the bulk parent meteorites. We also selected the United
196	States Geological Survey (USGS) terrestrial rock standard DTS-1 (along with Allende)
197	as a reference material to test the precision and accuracy of the data. Furthermore, the
198	Cr isotope compositions of two widely used artificial standards, NIST 3112a and
199	SCP-Cr (ICP-MS elemental standard for Cr), were measured to test for potential
200	non-mass dependent isotopic fractionation of Cr induced during production of the
201	standards and to calibrate possible offsets between studies using different standards
202	[e.g., NIST 3112a was used by Qin et al. (2010) and Zhu et al. (2019a)]. Among these
203	samples, MIL 05082, Aguas Zarcas, Ornans and Bali were chunks, while the other
204	samples are powders. Based on our recording information, the powder of Jbilet
205	Winselwan, Mokoia, Leoville, Vigarano and Sahara 97096 were from original sample
206	masses of 1.01g, 1.08g, 1g, 0.16g, and ~0.5g respectively.

207 The samples were dissolved following the protocol described in Inglis et al. (2018) using Teflon bombs and an Analab EvapoClean, which has been successfully applied 208 209 in previous studies (Zhu et al., 2019b; Zhu et al., 2020b). The procedure involved 210 heating the samples in concentrated HF and HNO₃ (2:1) at 140 °C for two days, drying 211 down the samples and subsequent dissolution of the solid residues in 6N HCl (also at 212 140 °C) for another two days to ensure complete digestion of fluorides, and refractory 213 phases such as chromite and spinel. The combination of Teflon bombs and Analab 214 EvapoClean for chondrite dissolution is simple and convenient, resulting in lower blanks compared to traditional dissolution methods, such as PARRTM bomb dissolution 215 216 (e.g., Zhu et al., 2019c) and alkaline fusion (e.g., Qin et al., 2010). Before the chemical separation of Cr (see below), ~10% aliquots were preserved for subsequent 217 determination of the ⁵⁵Mn/⁵²Cr ratio and major element contents. 218

219 **2.2 Determination of the ⁵⁵Mn/⁵²Cr ratios**

High-precision Mn-Cr chronology requires the accurate determination of the high-precision Mn-Cr chronology requires the accurate determination of the high-precision Mn-Cr chronology requires the accurate determination of the method that was similar to those employed in previous studies (Göpel et al., 2015; Trinquier et al., 2008a; Zhu et al., 2019b; Zhu et al., 2020b). We initially prepared three Mn-Cr doped artificial standard solutions gravitationally, with Mn–Cr contents of 10– 100 ppb, 50–100 ppb and 100–100 ppb and Mn/Cr ratios of $^{\circ}0.1$, $^{\circ}0.5$ and $^{\sim}1.0$. The unpurified sample solutions were diluted to a Cr content of ~100 ppb. The intensities

227	for ⁵⁵ Mn and ⁵² Cr on Faraday detectors obtained when analyzing the standard and
228	sample solutions ranged from 0.5 V to 5 V, and 10 cycles of 4 seconds each were
229	measured in each analysis to obtain a target precision for the ⁵⁵ Mn/ ⁵² Cr ratios better
230	than 0.1%. After establishing a calibration curve ($R^2 > 0.999$) based on the true and
231	measured ⁵⁵ Mn/ ⁵² Cr ratios of the three artificial standards, the ⁵⁵ Mn/ ⁵² Cr ratios of the
232	chondrite samples could be calculated. The external precisions for the ⁵⁵ Mn/ ⁵² Cr ratios
233	are better than 0.5% (2SD, N=6) as determined from multiple measurements of the
234	USGS standards PCC-1 and DTS-1. The final estimated precision of $<5\%$ (2 σ) was
235	determined from a comparison of the PCC-1 and Allende meteorite results with those
236	in the literature (Moynier et al., 2007; Qin et al., 2010; Shukolyukov and Lugmair,
237	2006; Trinquier et al., 2008a). The determination of the Mn/Cr ratios on Neptune Plus
238	is faster than the standard-addition method (that requires preparation and analysis of at
239	least four solutions per sample; (e.g., Qin et al., 2010; Zhu et al., 2019a) and more
240	accurate than the Mn and Cr content determination (e.g., Pedersen et al., 2019) by
241	quadrupole ICP-MS (5–10%, 2σ). Additionally, introduction of the low-concentration
242	unpurified samples into the MC-ICP-MS does not result in measurable memory effects.

243 **2.3 Column chemistry**

Low Cr yields from column chemistry, where Cr isotopes typically fractionate via equilibrium processes, can result in apparent mass-independent Cr isotope variations resulting from inappropriate mass fractionation corrections (Larsen et al., 2016; Qin et

247	al., 2010; Trinquier et al., 2008a). To avoid this issue, a four-step column chemistry for
248	Cr purification with high yield broadly following previous approaches (Bizzarro et al.,
249	2011; Larsen et al., 2018; Larsen et al., 2016; Pedersen et al., 2019; Schiller et al., 2014;
250	Trinquier et al., 2008a) was employed (Table 1). Only ~5 mg of samples were
251	dissolved in 10 M HCl and dried down three times before the following column
252	chemistry step to purify the Cr. First, we used an anion chromatographic purification
253	column to efficiently remove Fe in 6 M HCl. Prior to sample loading on cation
254	exchange columns, we used a Cr pre-treatment procedure involving dissolution in 10 M
255	HCl at >120 °C to efficiently promote the formation of Cr^{3+} -Cl species, which have a
256	low affinity for the cation exchanger and thus elute early (Larsen et al., 2016; Trinquier
257	et al., 2008a). This was followed by elution of Cr on a 1 ml cation exchange column in
258	20 ml of 0.5 M HNO3 to remove the major elements including Mg, Ca, Al, Ni (Bizzarro
259	et al., 2011) and collect all the Cr species (major Cr^0 and minor Cr^{2+} and Cr^{3+}) to reach
260	a > 99 % recovery. The samples were then exposed to $0.5M \text{ HNO}_3 + 0.6\% \text{ H}_2\text{O}_2$ at
261	room temperature for >1 day to promote the formation of Cr^{3+} (Larsen et al. 2016).
262	However, it is difficult to transform all Cr to Cr ³⁺ , so the Cr ⁰ -bearing material is
263	collected in 0.5 ml of the loading solution and 0.5 ml of 0.5N HNO ₃ elution to increase
264	the recovery to > 95 $\%$ in the next column. The third clean-up column involved Cr
265	purification from Al, Fe, V, Ti (and other high-field-strength elements) and Na, K on a
266	small (0.33 ml) cation exchange column using 0.5 M HNO ₃ , 1 M HF and 6 M HCl

267 (Larsen et al., 2018). Finally, for the fourth column, 0.7 ml of TODGA resin were used in 8N HCl to remove the residual Fe, V and Ti (stuck on the column) which have 268 isobaric isotopes with ⁵⁴Cr (⁵⁴Fe) and ⁵⁰Cr (⁵⁰V and ⁵⁰Ti) (Pedersen et al., 2019; Schiller 269 270 et al., 2014). The full procedure typically reaches a total yield between 95 % and 99 %, 271 and effectively removes any matrix, especially Fe, V and Ti. Low abundances of 272 matrix elements are important for analyses by MC-ICP-MS as all elements present are 273 ionized (unlike the selective thermal ionization of TIMS) generating potential isobaric 274 interferences and altering the mass fractionation behavior. Artificial standards 275 including NIST 979, NIST 3112a and SCP-Cr (The standard for ICP-MS 276 measurements) were passed through the first column chemistry step to eliminate residual Fe. The total blank of our full purification protocol is <5 ng, which is 277 278 negligible compared to the 10~20 µg of Cr processed through the columns. The final Cr solution was dried in $\sim 100 \,\mu$ l of concentrated HNO₃ three times to transform the acid 279 280 media and remove residual organics (i.e., those from the cation exchange resin).

281 **2.4 Isotope analysis**

The Cr isotopic compositions of all the samples were determined using an MC-ICP-MS Neptune Plus located at the Centre for Star and Planet Formation, Globe Institute, University of Copenhagen. Detailed analytical and data reduction methods are described in Schiller et al. (2014) and Pedersen et al. (2019). Each sample was measured by sample-standard bracketing using the NIST SRM 979 Cr standard. Sample solutions with ~0.5 ppm of Cr were introduced to the plasma via an ESI Apex IR resulting in 52 Cr signals of 20~40 V at an uptake rate of ~0.06 mL/min. Each sample was measured five times. The 53 Cr/ 52 Cr and 54 Cr/ 52 Cr ratios were normalized to a constant 50 Cr/ 52 Cr ratio of 0.051859 using an exponential law (Lugmair and Shukolyukov, 1998). All the measured isotopic ratios are expressed relative to NIST SRM 979 in the epsilon notations:

293
$$\epsilon^{x}Cr = \left(\frac{({}^{x}Cr/{}^{52}Cr)_{sample}}{({}^{x}Cr/{}^{52}Cr)_{NIST SRM 979}} - 1\right) \times 10000 (1),$$

294 with
$$x = 53$$
 or 54.

295 In order to control the influence of the potential isobaric interferences from Fe, V and Ti (⁵⁴Fe to ⁵⁴Cr, ⁵⁰V and ⁵⁰Ti to ⁵⁰Cr) and major alkali elements (Na and K), we also 296 297 performed doping tests for these elements (the doped samples are the corresponding SCP elemental standards of ICP-MS). The external precision was tested on five 298 299 Allende, five DTS-1 and two Orgueil from aliquots that were each individually purified 300 from the same digestion. We also provide the ε^{53} Cr data for another Ivuna sample of which ε^{54} Cr has been reported in Van Kooten et al. (2016) that described the related 301 302 analytical methods.

303 **3. Results**

321

The Cr isotope data for doping and external precision tests are reported in Table 304 305 2 and combined with literature data from Schiller et al. (2014). In Table 3, we compare the ⁵⁵Mn/⁵²Cr and Cr isotope data for the same chondrites measured in this 306 307 and previous studies (Göpel et al., 2015; Jenniskens et al., 2012; Kadlag et al., 2019; Langbroek et al., 2019; Mougel et al., 2018; Moynier et al., 2007; Petitat et al., 2011; 308 Qin et al., 2010; Sanborn et al., 2019; Schiller et al., 2014; Trinquier et al., 2007; 309 310 Trinquier et al., 2008b; van Kooten et al., 2020; Van Kooten et al., 2016; Williams et 311 al., 2020; Zhu et al., 2020a; Zhu et al., 2020b). The averaged group Cr isotope data and ⁵⁵Mn/⁵²Cr ratios of all the chondrites are reported in Table 4. We also summarize 312 313 the Cr isotope data for Rumuruti (R) chondrites, Earth, Moon, Mars, Vesta and other 314 achondrite parent bodies in Table 5. Relevant literature O isotope data are shown 315 alongside the new Cr isotope data in Tables 4 and 5 (the samples measured for O and 316 Cr isotope compositions are never from the same aliquots). The Cr isotope data 317 (including literature data) for the terrestrial samples are listed in Table 6. We also 318 re-measured the ⁵⁵Mn/⁵²Cr ratios on unprocessed dissolution aliquots for the H 319 chondrites reported in Pedersen et al. (2019) using our MC-ICP-MS approach. 320 The higher precision Cr isotope data (than those typically obtained by TIMS) for

322 are consistent with most previously reported values (Mougel et al., 2018; Qin et al.,

DTS-1 (USGS standards), Allende and as well as the Orgueil meteorites reported here

323	2010; Schiller et al., 2014; Trinquier et al., 2007; Trinquier et al., 2008b; Zhu et al.,
324	2019a; Zhu et al., 2019b; Zhu et al., 2020b), providing confidence in the accuracy of
325	our protocol. Based on multiple individually processed aliquots of Allende (5), DTS-1
326	(5) and Orgueil digestions (2), we estimate the external reproducibility of our data to
327	be better than 0.04 and 0.07 for ε^{53} Cr and ε^{54} Cr, respectively (Table 2), which is
328	consistent with the estimates from Schiller et al. (2014). The doping tests show that
329	isobaric interferences do not result in resolvable effects when Fe, V and Ti
330	interferences represent less than ~0.1% (2505 ppm to 54 Cr), ~2.5% (1438 ppm to 50 Cr)
331	and ~1% (14269 ppm to 50 Cr), respectively (Table 2). Finally, Na and K have very
332	limited effects even when their concentrations are sub similar to Cr in the analyzed
333	solutions, and they cause no drift in the Cr mass fractionation.

There are no resolvable mass-independent Cr isotope shifts between chondrite falls and finds from the same group, implying that the mass-independent Cr isotope compositions preserved in the meteorites is robust against limited terrestrial weathering. As such, we conclude that the Cr isotope data reported here is accurate within the reported uncertainties.

The ε^{54} Cr values of the studied CC groups decrease in the following sequence (mean ± 2SD): CI (1.56 ± 0.07) > CH (1.50 ± 0.07) ≥ CB (1.36 ± 0.30) ≥ CR (1.28 ± 0.23) ≥ CM (0.92 ± 0.26) ≈ CV (0.89 ± 0.30) ≈ CO (0.90 ± 0.43) ≥ CK (0.51 ± 0.15) (Fig. 1 and 2). Here, the calculated 2SD uncertainty for grouping CI and CH

343	chondrites are 0.05 and 0.01 respectively (Table 4), which are less than the external
344	uncertainty of 0.07 in this study. Thus, we quote the external reproducibility of 0.07
345	for the ε^{54} Cr uncertainty of CI and CH chondrites rather than the calculated 2SD. CC
346	reservoirs have a ε^{54} Cr +0.3 higher than Non-CC reservoirs as terrestrial samples and
347	ECs have ε^{54} Cr values in the range 0 – 0.2. Resolved intra-group ε^{54} Cr variability
348	exists within CB, CM, CV and CO chondrite data, whereas no significant intra-group
349	ϵ^{54} Cr variability was found amongst CI, CH, CR and CK chondrite data at the level of
350	our precision. The ϵ^{54} Cr values are not correlated with the degree of aqueous alteration
351	(CR2 to CR1, CM2 to CM1) and, at least amongst the CK chondrites, they are also not
352	correlated with the extent of thermal metamorphism (CK4 to CK6) (Table 3).
353	Furthermore, no systematic correlation between ϵ^{53} Cr and ϵ^{54} Cr values is observed
354	among the CCs, contradicting the claim of a correlation suggested by Shukolyukov and
355	Lugmair (2006).

The carbonaceous chondrites have decreasing ⁵⁵Mn/⁵²Cr ratios in the order: CI > CM \ge CR \ge CO > CV = CK \ge CB = CH (Fig. 3a). Despite these differences, the CCs with the highest and lowest ⁵⁵Mn/⁵²Cr ratios, the CI and CB-CH chondrites, respectively, have the highest ε^{54} Cr values (Fig. 1). More importantly, when considering all CC groups there is no systematic increase in ε^{53} Cr values with increasing ⁵⁵Mn/⁵²Cr ratios, based on the data in this study (Fig. 3a). Most CCs have ε^{53} Cr values ranging from 0 to 0.2, with an average value of 0.15 (± 0.13, 2SD; ± 0.02,

363	2SE; N = 41; excluding the silicate separate of CB chondrites). The ε^{53} Cr values of CI,
364	CR, CM and CB chondrites tend to be slightly higher compared to CH, CO, CV and CK
365	chondrites, although some CO and CM chondrites exhibit indistinguishable $\epsilon^{53}\text{Cr}$
366	values. Limited intra-group differences in ε^{53} Cr values that are not correlated with their
367	respective ⁵⁵ Mn/ ⁵² Cr ratios also exist in, for example, the CR, CM and CO groups. The
368	$^{55}\text{Mn}/^{52}\text{Cr}$ and $\epsilon^{53}\text{Cr}$ values for most OCs are correlated (Fig. 3b) and a model 1
369	regression of <i>IsoplotR</i> (Vermeesch, 2018) of these data results in a slope of 0.48 \pm
370	0.20 that corresponds to an initial 53 Mn/ 55 Mn of 5.39 ± 2.23 (MSWD = 2.2, 2SE, N =
371	23) and an initial ϵ^{53} Cr (intercept of y axis) of -0.16 ± 0.05. All the regressions
372	reported in this paper are calculated in the same way. The Cr data for all the ECs
373	(with literature data) reveals no (positive) correlation between Mn/Cr ratio and ϵ^{53} Cr
374	(Fig. 3c).

When combined with literature data (Mougel et al., 2018; Trinquier et al., 2007; Trinquier et al., 2008b), the terrestrial samples show minor Cr isotopic heterogeneity (Fig. 4), with average ε^{53} Cr = 0.04 (± 0.08, 2SD; ± 0.02, 2SE; N =15) and ε^{54} Cr = 0.07 (± 0.12, 2SD; ± 0.03, 2SE; N = 15). There is no obvious difference between samples with different petrology or chemistry (e.g., the basalt and peridotite). The Cr metal standards NIST SRM 3112a and SCP-Cr have indistinguishable Cr isotope compositions from the NIST SRM 979 standard.

382 **4. Discussion**

383 4.1 Comparison of Cr isotope data for chondrites with literature

384 CI chondrites are dominated by matrix materials and contain very few 385 chondrules and refractory inclusions (Barrat et al., 2012; Krot et al., 2014), making 386 them more homogenous than other types of chondrites. Hence, CI chondrites are good candidates to compare the Cr isotope data from this study with those in the literature. 387 388 We have listed all the reported Mn/Cr ratios and Cr isotope data for Orgueil in Table 3. For Orgueil, ε^{54} Cr values generally fall between 1.50 – 1.60, except for the data 389 reported in Kadlag et al. (2019), which are higher (ε^{54} Cr =1.94 ± 0.12). For ε^{53} Cr, some 390 391 studies (Trinquier et al. 2008, Qin et al. 2010, Schiller et al. 2014) report values of around 0.20, except for those in Kadlag et al. (2019), Moynier et al. (2007) and 392 393 Shukolyukov and Lugmair (2006), all of which report slightly higher values around 394 0.40. These elemental and isotope inconsistencies cannot be attributed to the influence from carbonates that have high Mn/Cr ratios and high ε^{53} Cr values, because the 395 ⁵⁵Mn/⁵²Cr ratios of CI chondrites reported in all studies are similar (0.80-0.85). As for 396 ⁵⁵Mn/⁵²Cr ratios, the data for Allende (0.51), Orgueil (0.94) and Ivuna (0.93) in Kadlag 397 398 et al. (2019) are systematically higher than those reported in this study and other literatures (see Table 3). The cause of the inconsistency between Kadlag et al. (2019) 399 and other recent studies is unknown. Stracke et al. (2012) showed that all the 38 400

401 Allende samples have an similar 55 Mn/ 52 Cr values, averaging at 0.41 ± 0.02 (that is 402 consistent with the value from our study, 0.42 ± 0.02), while Kadlag et al. (2019) 403 reported a value of 0.51 ± 0.03. Since Kadlag et al. (2019) used Parr bombs in their 404 dissolution procedures, it is unlikely that incomplete sample dissolution can account for 405 the data inconsistency.

406 A similar data inconsistency is also be observed in another CI chondrite, Ivuna. Kadlag et al. (2019) reported a ε^{54} Cr value (1.79 ± 0.20) for Ivuna, which is higher than 407 that reported in other literature (Schiller et al., 2014; Shukolyukov and Lugmair, 2006; 408 Van Kooten et al., 2016), while Williams et al. (2020) reported a lower ε^{54} Cr value of 409 1.30 ± 0.09 . As for ε^{53} Cr values, Shukolyukov and Lugmair (2006) report a higher 410 411 ε^{53} Cr value than that of Schiller et al. (2014) and this study, and the data in Kadlag et al. (2019) has a large error (0.17). Both higher ε^{53} Cr and ε^{54} Cr values in Kadlag et al. 412 413 (2019) might be caused by a residual mass-dependent Cr isotope fractionation using 414 TIMS. However, since the authors do not provide data for terrestrial samples, it is 415 difficult to evaluate this hypothesis. Similarly, Shukolyukov and Lugmair (2006) and 416 Williams et al. (2020) did not report data for terrestrial samples and, as such, the accuracy of their data is difficult to evaluate. The reason for the slightly lower ϵ^{54} Cr 417 data in Williams et al. (2020) is also not clear since the authors did not report the ε^{53} Cr 418 data. Note that a residual mass-dependent fractionation effect would shift both ε^{53} Cr 419 and ε^{54} Cr with a factor of ~1:2 [see discussion in section 4.5, and literature (Bourdon 420

421 and Fitoussi, 2020; Qin et al., 2010; Shukolyukov and Lugmair, 2006; Trinquier et al.,
422 2006, 2008a)].

Despite some inconsistency between the various studies, we list and consider all 423 424 published data in Table 3. Averaging the all the data in literature and this study gives: Orgueil: ϵ^{53} Cr = 0.29 ± 0.07 and ϵ^{54} Cr = 1.60 ± 0.10; Ivuna: ϵ^{53} Cr = 0.28 ± 0.10 and 425 ε^{54} Cr = 1.56 ± 0.15 (the uncertainty reflects the 95% confidence interval, which is also 426 427 used for other chondrite samples with multiple literature data; Table 3, 4). 428 Allende, which is a large fall (mass of ~ 2 tons), has been the subject of extensive Cr isotopic studies. All the Allende data show relatively homogeneous ε^{53} Cr values 429 (ranging from 0.04 ± 0.06 to 0.16 ± 0.06) but heterogeneous ε^{54} Cr values, ranging from 430 0.86 ± 0.09 to 1.10 ± 0.08 (Trinquier et al., 2007; Zhu et al., 2020b). Since the CVs are 431 432 the chondrites with the highest abundances of refractory inclusions (CAIs - Ca, Al-rich inclusions and AOAs – amoeboid olivine aggregates), which have extreme ε^{54} Cr values 433 (Trinquier et al., 2009), and they also have chondrules with variable ε^{54} Cr values 434 435 (Olsen et al., 2016), the ε^{54} Cr variability between Allende measurements likely reflect sample heterogeneity at the scales sampled. The average data for Allende: ε^{53} Cr 0.11 ± 436 0.02 and ε^{54} Cr = 0.95 ± 0.07 is reported in Table 4. 437

We also compared the Cr isotope data for other chondrites analyzed by various
workers (samples are listed in Table 3). In Table 4, we list the averages and the 95%
confidence interval uncertainties for the averaged values. Within the uncertainties,

441	most analyses of the same meteorites are consistent. Two exceptions are the $\epsilon^{54} Cr$
442	values of Jbilet Winselwan (CM2) (van Kooten et al., 2020) and the ε^{53} Cr values of
443	Lancé (CO3) (Moynier et al., 2007; Trinquier et al., 2008b). The small-degree of ϵ^{54} Cr
444	heterogeneity $(0.82 \pm 0.04 - 1.01 \pm 0.12)$ in Jbilet Winselwan could also result from
445	sample heterogeneity, given that CM chondrites also contain abundant refractory
446	inclusions (Krot et al., 2014), and their chondrules possess variable ϵ^{54} Cr values (van
447	Kooten et al., 2020). Note that the ε^{53} Cr of Lancé, -0.04 ± 0.07 in Trinquier et al.
448	(2008b), is lower than that of most other CO chondrites and even those of all other
449	measured chondrites, and its ϵ^{54} Cr value is also lower than all the other CO chondrites.
450	Because the mass of the Lancé aliquot analyzed by Trinquier et al. (2008b) is only 9 mg
451	it is possible that it is not representative of the bulk parent meteorite. As such, we have
452	not included the ϵ^{53} Cr and ϵ^{54} Cr values for Lancé in Table 4 and do not consider it in
453	further discussion.
454	It is also noteworthy to discuss the Cr isotope variation in CB chondrites. The
455	three CB chondrites measured in this study (with different subgroups, i.e., CB, CBa

456 and CBb), MIL 05082, QC 001 and HaH 237 (mostly silicate), have homogeneous

457 ϵ^{54} Cr values (1.46 ± 0.08; 2SD, N = 3), which is inconsistent with those of other

458 previously reported CB chondrites HaH 237 (0.87 ± 0.19) (Shukolyukov and Lugmair,

459 2006) and Bencubbin (bulk, average of silicate and metal; 1.12 ± 0.03) (Trinquier et al.,

460 2007). Here, since the metal and silicate parts for Bencubbin (Trinquier et al., 2007)

461	and Gujba (Yamashita et al., 2010) have consistent ϵ^{54} Cr values, we averaged the ϵ^{54} Cr
462	values of their different components to represent their bulk ϵ^{54} Cr compositions. The
463	bulk ϵ^{54} Cr values of HaH 237 and Bencubbin are different from the average ϵ^{54} Cr value
464	of the Gujba components, 1.29 ± 0.07 , (Yamashita et al., 2010), but overlap within
465	uncertainty with the ϵ^{54} Cr value (1.07 ± 0.27) of the metal spherules from Gujba
466	(Trinquier et al., 2008b). We do not interpret this ϵ^{54} Cr inconsistency as
467	inter-laboratory biases because the Cr isotope data for terrestrial samples from the
468	different studies (including different instruments within the same laboratory) are all
469	consistent within error (Qin et al., 2010; Trinquier et al., 2007; Trinquier et al., 2008b;
470	Zhu et al., 2019a; Zhu et al., 2020a; Zhu et al., 2020b). Instead, it is likely that there is
471	some ϵ^{54} Cr variability between CB chondrite samples.
472	The ε^{54} Cr data for the CO chondrites analyzed in this study (Table 4), ranging

473 from 0.80 ± 0.06 to 1.22 ± 0.04 , are higher than for Félix (0.63 ± 0.09) and Lancé (0.57

 ± 0.11) reported in Trinquier et al. (2007), but similar to that of Kainsaz (0.87 ± 0.18)

475 reported in Qin et al. (2010) (Table 4).

476 **4.2 Updated** ε^{54} Cr sequence and intra-group ε^{54} Cr heterogeneity of 477 carbonaceous chondrites

478 Chondrites typically have low cosmic ray exposure ages (CREA, less than 100 Ma)
479 and relatively low Fe/Cr ratios (Eugster, 2003; Weber et al., 2001), which limits the
480 potential cosmogenic effects on their Cr isotope compositions. Even amongst angrites

481 with Fe/Cr ratios of up to 600 and CREA up to 60 Ma (Eugster, 2003; Zhu et al., 2019b) and mesosiderites with CREA up to 300 Ma (Eugster, 2003; Trinquier et al., 2007), 482 their ε^{54} Cr values remain relatively homogeneous and show no correlation with CREA. 483 484 As for the Fe-rich CB chondrites, the metal and silicate parts of Bencubbin have the same ε^{54} Cr values, again showing that cosmogenic effects are not detectable in them 485 with current measurement precisions. Therefore, ε^{54} Cr signatures are a robust tool for 486 487 tracing general genetic relationships of chondrite parent bodies. In Table 4, combining these new measurements with literature data (Göpel et al., 488 489 2015; Qin et al., 2010; Trinquier et al., 2007; van Kooten et al., 2020; Zhu et al., 2021) results in an updated ε^{54} Cr sequence: CI \geq CH \geq CB \geq CR > CM \approx CV \approx CO \geq CK > 490 491 $EC \approx RC > OC$ (Fig. 1). We note that the CB and CH chondrite samples in this study 492 may not represent a bulk sample due to the sample heterogeneity. However, the CB chondrite components (e.g., chondrules, silicates and metal) have homogeneous ϵ^{54} Cr 493 494 values (Trinquier et al., 2007; Yamashita et al., 2010), perhaps owing to their formation 495 mechanism by impact (Krot et al., 2005). Thus, the ε^{54} Cr data reported here for CB 496 chondrites can be used to estimate the bulk value of the parent meteorite. We note that the ε^{54} Cr systematics in CH chondrite components needs to be studied in the future. 497 This updated sequence provides new insights into confirming the classification of 498 499 meteorites linked to recognized meteorite groups and determining potential genetic affinities between ungrouped chondrites and recognized meteorite groups. However, 500

501	since our new data show an overlap in the ϵ^{54} Cr values of CM, CV and CO chondrites,
502	the applicability of the ϵ^{54} Cr systematics as a tracer in carbonaceous meteorites is
503	weaker than originally suggested (e.g., Trinquier et al., 2007). Aguas Zarcas fell on
504	April 23, 2019 in Costa Rica and was classified as a CM2 chondrite (the Meteorite
505	Bulletin #108). This chondrite is of significant interest as it contains abundant prebiotic
506	compounds similar to other CM chondrites (Glavin et al., 2020). The Cr isotopic data
507	for Aguas Zarcas from this study, with ε^{53} Cr = 0.15 ± 0.03; ε^{54} Cr = 0.86 ± 0.03, is
508	consistent with the ε^{54} Cr variation range of CM chondrites (ε^{54} Cr = 0.92 ± 0.24, 2SD).
509	The ϵ^{54} Cr and Δ^{17} O values of CCs were previously reported to be correlated
510	(Trinquier et al., 2007). However, this correlation was based on a limited data set and
511	the Cr and O isotope data were often not from the same chondrites (see Fig. 2;
512	Trinquier et al., 2007). Our new data (same chondrites, but not the same sample
513	aliquots for Cr and O isotope measurements) show that this correlation ($R^2 = 0.58$) is
514	not as robust as previously observed. Nonetheless, our data confirm the Cr isotopic
515	difference between the CCs and most other (e.g., OCs, RCs and ECs) meteorites
516	(Trinquier et al., 2007; Warren, 2011), which is also observed for Ti (Trinquier et al.,
517	2009; Zhang et al., 2012), Ca (Dauphas et al., 2014; Schiller et al., 2018), Ni (Steele et
518	al., 2012); Mo (Budde et al., 2016; Spitzer et al., 2020; Yokoyama et al., 2019), and Ru
519	(Fischer-Gödde et al., 2015; Fischer-Gödde and Kleine, 2017).

520	An important new observation shown by our extended database is the intragroup
521	ϵ^{54} Cr variability among the CB, CM, CO, and CV chondrites (Figs. 1 and 2). This is
522	consistent with reported intra-group heterogeneities for bulk Ti isotope anomalies
523	(expressed as ϵ^{50} Ti) amongst the CM, CV and CO chondrites (Trinquier et al., 2009;
524	Zhang et al., 2012). One of the best examples of this is the Allende CV chondrite
525	whose published ε^{54} Cr values range from 0.86 ± 0.09 to 1.10 ± 0.08 (Qin et al., 2010;
526	Trinquier et al., 2007; Zhu et al., 2020b). Similarly, the three CB chondrites studied
527	here: MIL 05082 (CB), QC 001 (CBa), and HaH 237 (CBb) have indistinguishable
528	ϵ^{54} Cr values with a mean of 1.46 ± 0.08 (2SD, N=3). This is in contrast to the
529	previously reported data for CBs that are significantly different in their absolute values
530	and more variable, e.g., 1.29 ± 0.07 (2SD; average value of the chondrules and metal in
531	Gujba) for Gujba, and 1.12 ± 0.03 (2SD) for Bencubbin (Trinquier et al., 2007;
532	Yamashita et al., 2010), but marginally overlaps with the ε^{54} Cr data (1.07 ± 0.27) for
533	one metal chondrule in Gujba (Trinquier et al., 2008). We also find that some CH and
534	CB chondrites have comparable $\epsilon^{54} Cr$ compositions to CIs, which until now were
535	considered the most $^{54}\text{Cr}\text{-enriched}$ chondrites in bulk. The intragroup $\epsilon^{54}\text{Cr}$
536	heterogeneity of the CB, CM, CO, and CV chondrites likely result from sample
537	heterogeneity at the scale of most measured samples that is also reflected by intragroup
538	(e.g., CM, CB, CV, or CR) or even intra-chondrite (e.g., Jbilet Winselwan, Paris, and
539	NWA 8157) O isotope variability (see Fig. 2 and Table 4). However, the O isotope

540	compositions (Δ^{17} O) can also change as a result of aqueous alteration (Schrader et al.,
541	2014; Schrader et al., 2011) and terrestrial weathering (Alexander et al., 2018). Mineral
542	and acid leachates exhibit very large Cr isotope variability (Göpel et al., 2015; Podosek
543	et al., 1997; Qin et al., 2010; Rotaru et al., 1992; Schiller et al., 2014; Trinquier et al.,
544	2007; Wang et al., 2011). The CM, CO, and CV chondrites also contain large fractions
545	of CAIs and AOAs (5 vol.%, 13 vol.%, and 10 vol.%, respectively; Krot et al., 2014;
546	Weisberg et al., 2006) that have high ε^{54} Cr values of up to ~6 (Larsen et al., 2011;
547	Trinquier et al., 2009) but relatively low Cr contents. Although chondrules recorded
548	heterogeneous ϵ^{54} Cr values (e.g., Bollard et al., 2019; Olsen et al., 2016; Qin et al.,
549	2011; Schneider et al., 2020; van Kooten et al., 2020; Zhu et al., 2019a; Zhu et al.,
550	2020a), it is unlikely that they are the cause the ϵ^{54} Cr heterogeneity between bulk
551	chondrites from same groups because OCs, ECs and CK chondrites, which have
552	chondrules showing clear ϵ^{54} Cr heterogeneity (Bollard et al., 2019; Williams et al.,
553	2020; Zhu et al., 2020a), have homogeneous ε^{54} Cr values. Schneider et al. (2020) report
554	that various matrix material in Allende have a fairly narrow range of ϵ^{54} Cr values (1.06
555	\pm 0.22; 2SD, N = 3) and, as such, a variable chondrule/matrix ratio could also
556	contribute to the variability of the ϵ^{54} Cr values between CCs groups. It should be noted
557	that the OCs and ECs have high abundances (60%-80%) of chondrules (Krot et al.,
558	2014), which is consistent with the ϵ^{54} Cr homogeneity in OCs and ECs, since the
559	chondrule/matrix-ratio effect should be less in OCs and ECs than that in CCs.

560	Alternatively, the limited ε^{54} Cr heterogeneity within the OCs, ECs, RCs and CKs
561	(maybe also CR; Bunch et al., 2008) could also be the result of metamorphic
562	homogenization. Since they are not as metamorphosed, the apparent intragroup $\epsilon^{54} \mathrm{Cr}$
563	heterogeneity of CM, CO and CV chondrites (CV chondrules possess the most variable
564	ϵ^{54} Cr values, ranging from -0.79 to 2.01; Olsen et al., 2016) may be mainly due to the
565	relatively small sample size and non-representative sampling. This is consistent with
566	the observation that 0.6–1g of Allende may not be enough to be representative for the
567	average bulk composition (Stracke et al., 2012).
568	It is unlikely that metamorphism can explain the ϵ^{54} Cr homogeneity in type-3 OCs,
569	ECs and RCs. Also, the effect of sample size and CAI-AOA abundance cannot explain
570	the $\epsilon^{54}\text{Cr}$ heterogeneity amongst the CR and CB chondrites, because they have low
571	combined CAI and AOA contents (less than 0.5 vol%.; Scott and Krot, 2005) and
572	chondrules with almost homogeneous ϵ^{54} Cr values (Olsen et al., 2016; Yamashita et al.,
573	2010). Alternatively, it is possible that the intra-group ϵ^{54} Cr heterogeneity in CCs
574	(mainly CV, CO, CM, CB and CR groups) reflects ϵ^{54} Cr heterogeneity in their parent
575	bodies at scales that are larger than represented by the meteorites. This is also
576	consistent with the variable major element compositions, abundances of their
577	components (e.g., CAIs) and O isotope compositions (Fig. 2) in different chondrite
578	samples from the same group (Ebel et al., 2016; Hezel et al., 2008). Note that ϵ^{54} Cr
579	heterogeneities have also been found within achondrite groups, e.g., ureilites (Zhu et al.,

580 2020b), so it is also possible that some parent bodies (e.g., CV, CO, CM and CB) are
581 isotopically heterogeneous.

4.3 Constraints on parent body processes and possible genetic relationships (CV-CK, CM-CO and CB-CH clans) inferred from ε⁵⁴Cr systematics

The lack of correlated intragroup ε^{54} Cr values with the degree of aqueous 585 586 alteration (i.e., CR2 to CR1, CM2 to CM1) or thermal metamorphism (i.e., CK4 to CK6) also suggests that parent body processes did not redistribute the Cr at the scales we have 587 588 sampled (e.g., Qin et al., 2010; Sanborn et al., 2015; Trinquier et al., 2007). This is also 589 consistent with previous studies, indicating that petrologic types 3 to 6 in both the OCs and ECs have the same ε^{54} Cr values within their uncertainties (Mougel et al., 2018; Qin 590 591 et al., 2010; Sanborn et al., 2015; Trinquier et al., 2007). 592 Our new ε^{54} Cr data also allow us to evaluate if the CV sub-groups originate from 593 an isotopically homogeneous, common parent body. The three CV subgroups primarily 594 differed from one another in their redox states: oxidized CV_{oxA}, CV_{oxB} and the reduced CV_{red}. These CV subgroups may either originate from two or three different parent 595 596 bodies (Gattacceca et al., 2020);Greenwood, 2010 #1115} or, alternatively, from 597 different regions of a single parent body (Greenwood et al., 2010). Based on the 598 presence of CV_{oxB}-type clasts in the CV_{red} chondrite Vigarano (Krot et al., 2000) and 599 clasts of CV_{oxB} and CV_{oxA} lithologies in the oxidized CV Mokoia (Krot et al., 1998),

600	the origins of these subgroups from separate parent bodies already appears unlikely.
601	Our data show that meteorites from these three subgroups have indistinguishable ϵ^{54} Cr
602	signatures (e.g., CV_{oxA} : Allende with $\epsilon^{54}Cr$ of 0.95 ± 0.06; CV_{oxB} : Bali, Mokoia and
603	Kaba with ϵ^{54} Cr of 0.70 ± 0.07 to 1.10 ± 0.06; CV _{red} : Leoville and Vigarano with ϵ^{54} Cr
604	of 0.76 ± 0.10 and 0.85 ± 0.02 , respectively, Table 3), consistent with their origin from
605	a single parent body and that internal redox variations did not alter their bulk $\epsilon^{54}\text{Cr}$
606	compositions. This is inconsistent with the systematically different O isotope
607	compositions between oxidized and reduced CV chondrites (Clayton and Mayeda,
608	1999; Gattacceca et al., 2020). Combined with their distinct chondrule sizes and matrix
609	abundances, Gattacceca et al. (2020) argued there are multiple CV chondrite parent
610	bodies. Since CV_{ox} and CV_{red} chondrites have similar $\epsilon^{54}Cr$ values, and O isotopes are
611	sensitive to aqueous alteration (e.g., Farquhar et al., 1998), an alternative explanation
612	for the relationship between CV_{ox} and CV_{red} sub-groups is the regional heterogeneity in
613	water/rock ratio within a single parent body.

Based on chemical and petrological similarities and similar O-isotope compositions, the CV and CK chondrites have been grouped into the CV–CK clan and are considered by some to have originated from the same parent body (Greenwood et al., 2010). Nonetheless, the CV and CK chondrites are still considered different chondrite groups based on relative differences in the abundances of their bulk refractory lithophile elements and CAIs, and the presence of coarse-grained igneous

620	rims around chondrules in CV chondrites that are almost absent in the CK chondrites
621	(Kallemeyn et al., 1991). To reconcile the similar O-isotope compositions but the small
622	chemical and textural differences between CV and CK chondrites, it has been proposed
623	that the CK chondrites (which exist as petrologic types 3 to 6) formed deeper in the
624	same parent body as the CV chondrites (which are exclusively low petrologic type 3s)
625	(Greenwood et al., 2010; Wasson et al., 2013). This scenario is consistent with thermal
626	modeling (Elkins-Tanton et al., 2011). Our high-precision Cr isotope data reveal that
627	the CK chondrites have similar ε^{54} Cr values to one another, 0.51 ± 0.15 (2SD), ± 0.08
628	(2SE, N = 4), but are significantly lower than those of the CV chondrites (ϵ^{54} Cr = 0.89
629	± 0.30 [2SD], ± 0.12 [2SE, N=6]). This confirms the first resolvable difference between
630	the ε^{54} Cr values of one CV and one CK chondrite reported in Trinquier et al. (2007).
631	Given the lack of evidence that secondary parent body processes, including aqueous
632	alteration, thermal metamorphism and redox processes can significantly affect ϵ^{54} Cr at
633	the scale we sampled, the different $\epsilon^{54} Cr$ values for CV and CK chondrites imply that
634	they did not originate from the same parent reservoir. This is also supported by the
635	different chemical composition of magnetites in CV and CK chondrites, suggesting that
636	they experienced different metamorphic histories (Dunn et al., 2016).
637	Similarly, CM and CO chondrites, which are in the CM–CO clan (Weisberg et al.,
638	2006), share mineralogical and geochemical similarities, including anhydrous mineral

639 compositions and bulk elemental and O-isotope compositions (Clayton and Mayeda,

1999; Greenwood and Franchi, 2004; Kallemeyn and Wasson, 1981; Weisberg et al., 640 2006), which is consistent with their similar ε^{54} Cr compositions. As such, our Cr 641 642 isotope data further supports the idea that the CM and CO chondrite precursors formed 643 from similar materials. However, they cannot derive from the same parent body, 644 because (1) their mean chondrule sizes are resolvably different and their chondrules 645 (mainly type II chondrules) have different petrologic characteristics (Schrader and Davidson, 2017), (2) the CM chondrite parent body likely formed approximately 1 646 million years after the CO chondrite parent body (Sugiura and Fujiya, 2014). 647 648 The CB and CH chondrites form the same clan (Krot et al., 2014) and both of them have high ε^{54} Cr values (>1.1). However, the CBs have heterogeneous ε^{54} Cr 649 650 values, while those of the CHs are homogeneous. Furthermore, the CB (MIL 05082) 651 and CH (PCA 91467 and A-881020) chondrites have similar ⁵⁵Mn/⁵²Cr ratios but different ε^{53} Cr values, which is likely caused by their different initial ε^{53} Cr values 652 653 and/or formation times. This suggests different origins for the CB and CH chondrites, which is consistent with their different ε^{54} Cr features, i.e. the ε^{54} Cr heterogeneity and 654 655 homogeneity in the CB and CH chondrite parent bodies, respectively. However, the numbers of CB and CH samples that have been measured are still small and 656 657 measurements of more CBs and CHs are needed to confirm our conclusions.

34

4.4 ⁵³Mn-⁵³Cr systematics of bulk chondrites.

659	The previously postulated CC Mn-Cr isochron was mostly controlled by a single
660	CB chondrite, HaH 237 with low 55 Mn/ 52 Cr of 0.08 and ϵ^{53} Cr of -0.15 ± 0.09, and
661	two CI chondrites, Orgueil and Ivuna, with high $^{55}\text{Mn}/^{52}\text{Cr}$ of ~0.8 and $\epsilon^{53}\text{Cr}$ of ~0.4
662	(see the grey points and line in Fig. 3a) (Moynier et al., 2007; Shukolyukov and
663	Lugmair, 2006). Subsequently, Trinquier et al. (2008b) and Qin et al. (2010)
664	determined that using an updated ε^{53} Cr values for CI chondrites of ~0.2, the age of the
665	"isochron", decreases from 4568.6 \pm 1.1 Ma to 4566.1 \pm 2.4 Ma (the two ages are still
666	consistent within uncertainty). At that time, no CH chondrites had been studied. Here
667	we show that our sample of HaH 237 (mostly silicates) has a similar 55 Mn/ 52 Cr (0.09)
668	to that reported by Shukolyukov and Lugmair (2006) but a higher ϵ^{53} Cr value (0.05 ±
669	0.03). The two CB chondrites MIL 05082 and QC 001 have 55 Mn/ 52 Cr ratios of
670	0.37-0.47 and ϵ^{53} Cr values of 0.20 ± 0.01 (2SD, N =2) that are indistinguishable from
671	CI chondrites (0.21 \pm 0.04; 2SD, N = 2). Moreover, the CH chondrites in this study
672	with the lowest Mn/Cr ratios have ε^{53} Cr values of 0.12 ± 0.03 that are too high to be
673	consistent with the postulated isochron. Although our new values suggest that the CH
674	and CB chondrites are not consistent with a common CC isochron, a potential caveat
675	with this observation is the difficulty in obtaining samples that are representative of
676	the bulk meteorites. We note that while there is a broad correlation between bulk CC
⁵⁵Mn/⁵²Cr ratios and $ε^{53}$ Cr values, these data do not form a single well-defined Mn-Cr

isochron even when the CH and CB chondrites are excluded (Fig. 3a).

678

There are several likely reasons for the lack of a single, well-defined Mn-Cr 679 680 isochron among the CCs. Firstly, unlike other CCs, the CH and CB chondrites 681 probably formed via impacts that postdated the formation of other CCs (Krot et al., 682 2005; Yamashita et al., 2010), and the impact, e.g., ~5 Ma after CAIs (Krot et al., 2005), may potentially and secondarily modify the Mn/Cr ratios (Krot and Nagashima, 683 684 2017; Weisberg et al., 2001). Thus, there is no *a priori* reason why they should form a 685 common Mn-Cr isochron with the other CCs. Secondly, all eight CC groups have distinct photochemical and/or nucleosynthetic isotope anomalies (Clayton and Mayeda, 686 1999; Schiller et al., 2018; Trinquier et al., 2009; Zhang et al., 2012) reflecting 687 688 variability in the makeup of their precursors and/or their formation environments within the protoplanetary disk. Finally, chondrites are complex assemblages of CAIs, 689 690 AOAs, chondrules and matrix (Alexander et al. 2019a) that formed at different times 691 and under varying conditions (Connelly et al., 2012). The chondritic components (i.e., 692 chondrules, matrix, metals, CAIs and AOAs) do not all have the same ages and/or source regions (initial ε^{53} Cr values). For example, although CAIs and some 693 chondrules did form at the same time (see the internal isochron ages; Connelly et al., 694 695 2012; Bollard et al., 2017), these objects have distinct nucleosynthetic anomalies (Cr 696 and Ti: Trinquier, et al. 2009; Olsen et al., 2016; Gerber et al., 2017; Schneider et al.,

697	2020; Williams et al., 2020; Zhu et al., 2019a; Zhu et al., 2020a), and, as such, must
698	have formed in distinct reservoirs. Some CO and CM chondrites have similar
699	55 Mn/ 52 Cr ratios but distinct ϵ^{53} Cr values, which indicates that on average their
700	components experienced Mn/Cr fractionation at different times and/or conditions.
701	Their different radiogenic Cr isotopic compositions also supports CM and CO
702	chondrites originating from different parent bodies (e.g., Schrader and Davidson, 2017)
703	The matrix also has a distinct origin based on its variable nucleosynthetic isotope
704	signatures (Cr, Ti, Mo and W; Budde et al., 2016a; Budde et al., 2016b; Schneider et
705	al., 2020). This has also been discussed in Zhu et al. (2020a) and Alexander (2019a).
706	Overall, the Mn-Cr correlation for most chondrites is almost certainly a
707	multiple-component mixing trend. However, this mixing for CC Mn-Cr correlation
708	line is not limited to only chondrule-matrix (e.g., Anders, 1964), because CK
709	chondrites with high abundance of matrix (~75%; Krot et al., 2014) also have low
710	Mn/Cr ratios compared to most of other CC groups (Figure. 3a). Considering that the
711	half-life of ⁵³ Mn is comparable to the accretion ages of chondrites, it is perhaps
712	unsurprising that a general trend resembling a Mn-Cr isochron between chondrite
713	groups exists.

Such a correlation is stronger for the OCs than the CCs (Fig. 3b). The calculated initial 53 Mn/ 55 Mn ratio for this OC trend is $(5.39 \pm 2.23) \times 10^{-6}$. When anchored to D'Orbigny with the absolute age (U isotope-corrected) of 4563.37 ± 0.25 Ma and an

717	initial 53 Mn/ 55 Mn of (3.24 ± 0.04) × 10 ⁻⁶ (Amelin, 2008; Brennecka and Wadhwa,
718	2012; Glavin et al., 2004), this is equivalent to an age of 4566.1 \pm 2.3 Ma. However,
719	this apparent isochron for the OCs may not have chronological significance, even
720	considering that the $\epsilon^{54} Cr$ and $\Delta^{17} O$ values suggest a common origin for this group,
721	because the correlation line can also result from mixing of different proportions of
722	chondritic components. Compared to the CCs, the OCs have fewer refractory
723	inclusions, more chondrules, less matrix and variable amount of metal (H > L > LL).
724	The mixing end-members are thus chondrules, matrix and metal, and the metal should
725	play a more important role in the mixing. This is because the metal-rich H chondrites
726	possess lower $^{55}\text{Mn}/^{52}\text{Cr}$ ratios and lower $\epsilon^{53}\text{Cr}$ values, compared to metal-poor LL
727	chondrites, with intermediate L chondrites (Figure 3b) (Krot et al., 2014). Figure 3c
728	shows there is no resolved positive slope in a Mn-Cr correlation diagram. Note that
729	ECs are composed of chondrules, metals (Krot et al., 2014) and many Mn- or Cr-rich
730	sulfide minerals (e.g., Piani et al., 2016; Zhu et al., 2020a). Metals are poor in both
731	Mn and Cr, and silicate usually possess more Mn than Cr (Piani et al., 2016) such that,
732	their sulfide minerals have large variability for Mn and Cr contents (e.g., Cr-rich
733	troilite and Mn-rich niningerite; Piani et al., 2016), so their complicated Mn-Cr
734	mixing budgets may obscure any Mn-Cr correlation.

When comparing the ⁵³Mn-⁵³Cr data for all chondrite groups (Fig. 4), there is no
well-defined ⁵³Mn-⁵³Cr isochron. Actually, nearly half of the data points do not fit on

737	the regression line. However, all the different groups of chondrites here define a
738	positive trend between ϵ^{53} Cr values and 55 Mn/ 52 Cr ratios. For example, the CV, CK,
739	CO, and CH chondrites mostly possess low 55 Mn/ 52 Cr ratios (<~0.5) and ϵ^{53} Cr values,
740	<~0.12, while the CI, CM, CR (except one in van Kooten et al., 2020), and OCs
741	mostly have high 55 Mn/ 52 Cr ratios (>~0.5) and ε^{53} Cr values, >~0.12. These variations
742	may be caused by a multiple-endmember mixing of the different chondritic
743	components (chondrules, matrix, CAIs and AOAs) that on average experienced
744	Mn/Cr fractionations at different times, reflecting a general relationship between
745	55 Mn/ 52 Cr ratios and ϵ^{53} Cr values. Regressing all the Mn-Cr data for all the chondrites
746	[by model 3 (maximum likelihood regression with overdispersion) due to the
747	MSWD >> 1, <i>IsoplotR</i> (Vermeesch, 2018)], the slope is 0.285 ± 0.078 with initial
748	ε^{53} Cr = 0.00 ± 0.05 (MSWD = 11, N = 85). This slope corresponds to a 53 Mn/ 55 Mn =
749	$(3.23 \pm 0.89) \times 10^{-6}$ and an absolute age of 4563.4 ± 1.5 Ma, when anchored to the U
750	isotope-corrected D'Orbigny angrite. However, this ⁵³ Mn- ⁵³ Cr correlation line likely
751	represent a mixing line that does not have any chronological significance.

Trinquier et al. (2008) included a number of chondrites (OC, EC, CI, CV, and CO groups) and planets/asteroids (including Earth, Mars and Vesta) on the same 53 Mn- 53 Cr diagram with a slope of (6.53 ± 1.93) × 10⁻⁶ and an absolute age of 4567.3 ± 1.9 Ma. They interpreted this age as the last isotopic equilibration of Mn and Cr in the protoplanetary disk. The chondrites and planets had distinct origins and formation 757 times, which violates the primary assumption of an isochron that all components formed at the same time from the same reservoir. Thus, any Mn-Cr correlation is a 758 759 mixing line that does not have any chronological significance either. Here, it should also be mentioned that the age from a ⁵³Mn-⁵³Cr "isochron" established by acid 760 leachates in chondrites (Göpel et al., 2015; Trinquier et al., 2008b) are also 761 762 questionable and mostly reflect mixing lines. This is because 1) The chondritic components, e.g., CAIs, chondrules, matrix, metal and carbonates, have different 763 764 origins and formed at different periods, which has been discussed in the previous 765 section. For example, only five leachates, out of 15, of Orgueil CI chondrite fall on a 766 single Mn-Cr correlation line (Trinquier et al., 2008b). This possibly reflects the fact that CI1 chondrites experienced secondary aqueous alteration and thereby forming 767 768 resulting in the formation of younger carbonates (that would be dissolved by the weak 769 acid, e.g., acetic acid, in the first step of leaching) with high Mn/Cr ratios (e.g., Fujiya 770 et al., 2012; Fujiya et al., 2013), although CI chondrites are mostly composed of 771 matrix material, and contain few CAIs and chondrules (Krot et al., 2014). The 772 chondrites that have abundant CAIs, AOAs and chondrules, e.g., CV and CO chondrites, have a more complex Mn-Cr mixing process; 2) Mn is rich in the easily 773 dissolved minerals with high Mn/Cr ratios, e.g., carbonates, while Cr is rich in the 774 775 refractory phases with low Mn/Cr ratios, e.g., chromites, so the acid leachates potentially represent the components of high-Mn/Cr and low-Mn/Cr reservoirs, which 776

mostly reflects mixing lines that do not have chronological meaning. This is consistent with the anomalous 53 Mn/ 55 Mn [(13.64 ± 0.01) × 10⁻⁶; corresponding to 4571.1 ± 0.9 Ma that is older than CAIs] and initial ε^{53} Cr values (-0.61 ± 0.05) obtained from a Mn-Cr correlation line of acid leachates of the Paris chondrite (Göpel et al., 2015).

782 **4.5** The ε^{53} Cr deficit between chondrites and Earth suggests early 783 volatile depletion of Earth precursors.

784 The 15 terrestrial samples (including basalts, peridotites and chromites; including both MC-ICP-MS data in this study and TIMS literature data; see Table 6) measured 785 786 relative to NIST SRM 979 and corrected for mass bias using the kinetic fractionation 787 law exhibit ε^{53} Cr and ε^{54} Cr values that are slightly positive with 0.04 ± 0.08 (2SD); 788 0.02 (2SE) and 0.09 ± 0.12 (2SD); 0.03 (2SE), respectively (Fig. 5). The slightly positive ε^{53} Cr and ε^{54} Cr for terrestrial samples has been linked to an isotope 789 fractionation behavior that differs from the kinetic fractionation of atomic Cr in the 790 791 mass spectrometers that was induced during the preparation of the NIST 979 standard 792 (Schiller et al., 2014). Similar mass-independent isotopic difference between 793 terrestrial rocks and purified metal standards (i.e., NIST 3112a and SCP-Cr) have also 794 been observed for other elements like Ni (Steele et al., 2011), Sr (Moynier et al., 2012), Ti (Zhang et al., 2012), and Mo (Budde et al., 2019). As such, the reported 795 average ε^{54} Cr values of terrestrial samples are currently the best estimate of the bulk 796

terrestrial mass-independent Cr isotope composition. The residual variability in
terrestrial rock analyses highlights Cr isotope fractionation in nature and/or during Cr
purification and TIMS analysis that do not follow the kinetic law for Cr, perhaps
because they involved molecular species (e.g., CrO₂).

This is supported by the fact that theoretical calculations predict that 801 802 inappropriate fractionation correction should result in apparent mass mass-independent effects on ε^{53} Cr and ε^{54} Cr. Chromium evaporates not only as Cr⁺, 803 804 but multiple oxidized species such as CrO, CrO₂ and CrO₃ may also be present during 805 heating and evaporation during TIMS ionization. Based on masses and abundances stated in Table 7a, we firstly calculated the fractionation factors, β^{53} Cr and β^{54} Cr for 806 different Cr species following Young and Galy (2004). In detail, the factors for 807 808 equilibrium and kinetic fractionation laws are calculated from equation (9) and (12), respectively, in Young and Galy (2004). Then, we determined the ⁵³Cr/⁵²Cr and 809 ⁵⁴Cr/⁵²Cr ratios for a given ⁵⁰Cr/⁵²Cr fractionation and fractionating species. We 810 811 subsequently derive the relative deviations resulting from non-kinetic and/or 812 non-atomic Cr isotope fractionation from the predicted Cr isotope ratios resulting 813 from the typically applied mass bias correction based on kinetic laws and atomic Cr for these isotope ratios (Δ^{53} Cr and Δ^{54} Cr) (Table 7b). Irrespective of the fractionating 814 Cr species, the apparent anomalies in the ${}^{53}Cr/{}^{52}Cr$ and ${}^{54}Cr/{}^{52}Cr$ ratios ($\Delta^{53}Cr$ and 815 Δ^{54} Cr) co-vary with a factor of ~2.6, which also mimics the slope (2.6) of the reported 816

 ϵ^{53} Cr and ϵ^{54} Cr values of multiple measurements for NIST 3112a by Triton TIMS 817 (Qin et al., 2010). This ratio is also broadly consistent with the ε^{53} Cr vs. ε^{54} Cr values 818 amongst the terrestrial samples (Figure 5) that exhibit a slope of ~ 2 considering that 819 820 the magnitude of isotopic variability is typically on the order of the analytical uncertainty. However, small variations in ε^{53} Cr and ε^{54} Cr in natural terrestrial samples 821 may not solely be due to instrumental effects but may be the product of natural 822 equilibrium Cr stable isotope fractionation, i.e., the Cr stable isotope fractionation 823 during magmatic process where fractionation occurs based on exchange of Cr²⁺ and 824 Cr³⁺. 825

The average ε^{53} Cr value, 0.04 ± 0.02 (2SE, N = 15), of all terrestrial samples 826 provides the current best estimate for the Cr isotopic composition of the BSE. It should 827 828 be noted that Cr is more siderophile than Mn during core formation (Mann et al., 2009), resulting in elevated ⁵⁵Mn/⁵²Cr ratios in the BSE compared to the core. If the Earth core 829 formation process occurred early (e.g., Schiller et al., 2020), i.e. before ⁵³Mn extinction, 830 831 the higher Mn/Cr ratio of BSE compared to the core would theoretically produce higher ε^{53} Cr in the BSE relative to the core. Based on a mass balance calculation: 832 Bulk Earth = Bulk Silicate Earth + Core (2)833 the ε^{53} Cr values of the bulk earth (BE) should be similar to (if Earth core formed after 834 ⁵³Mn extinction) or slightly lower than (if Earth core formed before ⁵³Mn extinction) 835

that of BSE.

837	Since the ϵ^{53} Cr value of the BSE is lower than that of chondrites, the ϵ^{53} Cr of BE
838	must also be lower. The ε^{53} Cr value of BSE of 0.04 ± 0.02 (2SE, N = 15) is
839	systematically lower than the average ε^{53} Cr value of all the chondrites, ε^{53} Cr = 0.16 ±
840	0.01 [2SE, N = 88; see Fig. 5; (Trinquier et al., 2008b)]. We tested the significance of
841	the ϵ^{53} Cr isotopic difference between chondrites (N = 88) and Earth (N = 15) by
842	running an unpaired student t-test using Prism 8, which returned a P-value <0.0001
843	confirming the statistical difference between the two groups (statistical difference is
844	considered for P-value <0.05). The BE also has a lower ϵ^{53} Cr value compared to ECs
845	$(\epsilon^{53}Cr = 0.15 \pm 0.03; 2SE, N = 12; average {}^{55}Mn/{}^{52}Cr = 0.71)$ that have been used as
846	analogues for the isotopic composition of Earth's precursors via protracted stochastic
847	collisional accretion given their otherwise close nucleosynthetic match to the BSE
848	(Javoy et al., 2010), and CI chondrites (ϵ^{53} Cr = 0.22 ± 0.02; 2SE, N = 2; average
849	55 Mn/ 52 Cr = 0.82) that represent the bulk chemical composition of Solar System
850	(Allègre et al., 1995; Krot et al., 2014). This ε^{53} Cr deficit between chondrites and Earth
851	has been already observed in previous studies (Qin et al., 2010; Trinquier et al., 2008b),
852	and interpreted as representing early Mn/Cr volatile fractionation (Palme and O'Neill,
853	2014). Here, we combined all the chondrite data in this study and literature to better
854	constrain the early volatile depletion of the Earth.

855 Variability in the initial abundance of the ⁵³Mn/⁵⁵Mn within the early Solar 856 System is unlikely to cause this discrepancy given that the CCs and non-CCs have

857	similar ϵ^{53} Cr values (Trinquier et al., 2008b), and the relative Mn-Cr ages of some
858	achondrites (e.g., Sanborn et al., 2019; Zhu et al., 2019b) and chondrules (Bollard et
859	al., 2015; Krot et al., 2005; Yamashita et al., 2010; Zhu et al., 2019a; Zhu et al., 2020a)
860	are consistent with absolute Pb-Pb chronometry. As such, the cause for this $\epsilon^{53}\text{Cr}$
861	difference between the BSE and chondrites must be due to Mn/Cr fractionation prior to
862	the extinction of 53 Mn (half-life of 3.7 Myrs). The 55 Mn/ 52 Cr ratio of the BE is difficult
863	to estimate precisely. However, it has been estimated that the 55 Mn/ 52 Cr ratio of the
864	bulk Earth is between 0.22 and 0.54 (Palme and O'Neill, 2014; Sun, 1982; Wang et al.,
865	2018; Wänke and Dreibus, 1988), which is lower than those of the CIs (\sim 0.82) and
866	ECs (~0.71). The ECs have similar isotopic compositions as Earth for multiple
867	elements (Clayton et al., 1984; Javoy et al., 2010; Mougel et al., 2018; Piani et al.,
868	2020; Trinquier et al., 2007; Zhu et al., 2020a), and CIs are proxies for the chemical
869	composition of the bulk Solar System (Allègre et al., 1995). The lower ⁵⁵ Mn/ ⁵² Cr ratio
870	for BE is likely caused by preferential evaporative loss of Mn relative to Cr either
871	prior to or during Earth's accretion. We can estimate the timing of the volatile
872	depletion by calculating a minimum age of volatile loss. To that end, we calculate the
873	evolving ϵ^{53} Cr signature of BE using the initial ϵ^{53} Cr and 53 Mn/ 55 Mn abundance of the
874	Solar System (Trinquier et al., 2008) assuming a single stage Mn-loss that reduced the
875	initial Mn/Cr ratio of the accreting Earth to its current value between 0.22 and 0.54.
876	From this estimate, it becomes apparent that lowering of the Mn/Cr ratio of the Earth

must have occurred within 0–3 Myr after CAI formation. This age is also consistent
with another volatile-sensitive chronometer, the ⁸⁷Rb-⁸⁷Sr system that indicates that
the planetary volatile depletion events happened very early, i.e., in the first few Myrs
after CAIs (Gray et al., 1973; Hans et al., 2013; Moynier et al., 2012).

881 It should be noted that decreasing the Mn/Cr ratios from chondrites to Earth 882 would be consistent with stronger volatilization of Mn than Cr (Sossi et al., 2019). In fact, this age of proto-Earth volatilization is consistent with the ⁵³Mn-⁵³Cr age of 883 chondrule precursors (Zhu et al., 2019a; Zhu et al., 2020a) that have been considered 884 885 as potential candidates for the building blocks of terrestrial planets by the mechanism of pebble accretion (Johansen et al., 2015; Schiller et al., 2018; Schiller et al., 2020). 886 887 In particular, pebble accretion inevitably results in hot planetary atmospheres and 888 proto-planets (Popovas et al., 2018) providing a natural mechanism of early volatile 889 loss of the accreting Earth at early times (Mahan et al., 2018; Rubin et al., 1988). 890 Therefore, our data suggest that the terrestrial volatile depletion more likely occurred 891 during the formation of Earth precursors as opposed to later volatile loss during 892 planetary evolution. In particular, evaporation under planetary conditions is predicted to occur under more oxidizing conditions (Visscher and Fegley Jr, 2013), which could 893 894 make Cr more volatile than Mn (Sossi et al., 2019). This would result in an increase of the Mn/Cr ratio and subsequently the ε^{53} Cr of the devolatilized planet compared to 895 chondrites, which is the opposite of what is observed. 896

898 **5. Conclusion**

915

899	This study reports a new comprehensive mass-independent Cr isotope dataset of
900	numerous chondrite types as well as Earth, using improved methods for high-yield
901	purification method and high-precision Cr isotope measurements by MC-ICP-MS.
902	The main conclusion of our work can be summarized as follows:
903	1. Our ϵ^{54} Cr chondrite data complement earlier work, and provide an updated
904	decrease of ε^{54} Cr as follows: CI \ge CH \ge CB \ge CR > CM \approx CV \approx CO \ge CK > EC > OC.
905	Our data also show that CB, CM, CV, CR and CO chondrites have intra-group $\epsilon^{54} Cr$
906	heterogeneities that are likely caused by unrepresentative sampling or heterogeneous
907	accretion of chondrite parent bodies.
908	2. The distinct $\epsilon^{54} Cr$ for CV and CK chondrites suggest that they do not come
909	from the same parent body, whereas the indistinguishable $\epsilon^{54}\mbox{Cr}$ values of CM and CO
910	chondrites suggest their parent bodies share common precursor materials and, thus,
911	may have formed under similar conditions. Moreover, heterogeneous $\epsilon^{54}\mbox{Cr}$ values for
912	CB chondrites compared to the homogeneous ϵ^{54} Cr values for CH chondrites suggests
913	they may have different origins.
914	3. The low-Mn/Cr CH and CB chondrites have indistinguishable ε^{53} Cr values

916 the proposed bulk CC Mn-Cr isochron. The broad correlation between Mn/Cr ratios

from those of high-Mn/Cr CI chondrites and, as such, these data are inconsistent with

917 and ε^{53} Cr values in chondrites may simply reflect a mixing line between high- and low-918 Mn/Cr ratio reservoirs.

919	4. When compared to the NIST SRM 979 Cr standard, bulk (silicate) Earth is
920	characterized by ε^{53} Cr and ε^{54} Cr values of 0.04 ± 0.02 and 0.09 ± 0.03 (2SE),
921	respectively. The correlated and slightly positive values reflect non-kinetic isotopic
922	fractionation of the NIST SRM 979 Cr standard and some terrestrial samples. The
923	lower ϵ^{53} Cr of bulk silicate Earth compared to chondrites requires early volatile loss of
924	Earth's precursor materials. Based on the half-life of ⁵³ Mn (3.7 Ma), this volatile loss
925	likely occurred within 3 Myr of Solar System formation.
926	

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952	respectively.

Table 1 Four-step column chemistry used in this study to purify emolinum.								
Eluent	Volume (ml)	Procedural step	Elements eluted					
Step 1: 0.5 ml Biorad AG1-X8 200–400 mesh resin								
6M HCl	2	Condition						
6M HCl	0.5	Sample Load						
6M HCl	3	Cr collection	Cr+matrix					
H_2O	5	Wash	Fe+Ni					
Step 2: 1 ml Bio	orad AG50W-	X8 200–400 mesh resin						
0.5M HCl	3	Condition						
0.5M HCl	3	* ¹ Sample Load						
0.5M HNO ₃	20	Cr collection	Cr+matrix					
6M HCl 5 Wash Mg+Ca+								
Step 3: 0.33 ml	Biorad AG50	W-X8 200–400 mesh resin						
H_2O	1	Condition						
0.5M HNO ₃	0.5	*2Sample Load and Cr collection	Cr					
0.5M HNO ₃	0.5	Cr collection	Cr					
1M HF	2.5	Matrix elution	Ti+V+K+Na+Fe+Al					
1M HCl	6	Matrix elution	K+Na					
6M HCl	3	Cr collection + Wash	Cr					
Step 4: 0.75ml Eichrom TODGA resin								
8M HCl	2	Condition						
8M HCl	0.5	Sample Load	Cr					
8M HCl	2	Cr collection	Cr					
H ₂ O	4	Wash	V+Fe+Ti					

956 Note: *1 sample pretreatment: dissolving sample in 0.25 ml 6M HCl (in 7 ml beaker) with heating 957 at >120 °C for more than 2h, then added 2.75 ml H₂O before loading. This is to transform Cr as Cr⁰. *2 958 sample pretreatment: dissolving sample in 0.125 ml 2M HNO₃ (in 3 ml beaker) with heating at 100 °C 959 for 2h, then added 0.01 ml H₂O₂ and 0.365 ml H₂O, and put the mixture at room temperature for more 960 than 24h. This is to maximize the oxidation of Cr as Cr³⁺.

Table 1 Four-step column chemistry used in this study to purify chromium.

	Interference								
Samples	Contribution	ε⁵³Cr	2se	ε⁵⁴Cr	2se	N/[ref.]			
	(ppm)*								
Cr-V-0%	0	0.00	0.00	-0.01	0.02	5			
Cr-V-S14	24	0.00	0.02	0.00	0.02	[1]			
Cr-V-0.5%	269	0.00	0.03	0.04	0.02	5			
Cr-V-2.5%	1438	-0.06	0.02	-0.07	0.03	5			
Cr-V-5%	2764	-0.09	0.01	-0.12	0.04	5			
Cr-Ti-0%	0	-0.01	0.02	0.02	0.03	5			
Cr-Ti-S14	1609	0.01	0.02	0.00	0.02	[1]			
Cr-Ti-0.5%	7590	0.01	0.05	0.03	0.05	5			
Cr-Ti-1%	14269	0.02	0.03	0.06	0.08	5			
Cr-Ti-2%	27904	0.04	0.05	0.11	0.06	5			
Cr-Fe-0%	0	0.00	0.01	0.01	0.02	5			
Cr-Fe-0.01%	205	0.00	0.01	-0.08	0.04	5			
Cr-Fe-S14	558	0.02	0.02	0.01	0.02	[1]			
Cr-Fe-0.1%	2505	0.01	0.01	-0.04	0.05	5			
Cr-Fe-0.5%	11846	-0.01	0.02	-0.19	0.04	5			
Cr-Na-50%		0.04	0.05	0.11	0.05	4			
Cr-Na-100%		0.01	0.02	0.02	0.09	4			
Cr-K-50%		0.03	0.05	0.04	0.02	4			
Cr-K-100%		0.01	0.02	0.01	0.10	4			
Allende-1		0.08	0.01	0.87	0.08	5			
Allende-2		0.10	0.02	0.90	0.02	5			
Allende-3		0.11	0.02	0.94	0.02	5			
Allende-4		0.14	0.03	0.95	0.08	5			
Allende-5		0.10	0.02	0.96	0.05	5			
average		0.10	0.04	0.92	0.07	2SD			
DTS-1-1		0.05	0.01	0.11	0.07	5			
DTS-1-2		0.05	0.01	0.16	0.05	5			
DTS-1-3		0.06	0.03	0.18	0.07	5			
DTS-1-4		0.04	0.03	0.15	0.03	5			
DTS-1-5		0.07	0.02	0.19	0.07	5			
average		0.05	0.03	0.16	0.06	2SD			
Orgueil-1		0.19	0.05	1.51	0.05	5			
Orgueil-2		0.19	0.01	1.50	0.08	5			
average		0.19	0.00	1.50	0.01	2SD			

962 Table 2 Doping test for V, Ti, Fe, Na and K and external precision tests by Allende, DTS-1 and Orgueil.

Note: The references [1] Schiller et al. (2014). The doped samples are the corresponding SCP elemental

964 standards of ICP-MS. * the corresponding V, Ti and Fe interference is for 50 Cr, 50 Cr and 54 Cr

965 respectively.

966

	Mass (mg)	⁵⁵ Mn/ ⁵² Cr	ε⁵³Cr	Error	ε⁵⁴Cr	Error	N	Instruments	Ref.
Orgueil		0.87	0.43	0.10			11	Nu Plasma HR MC-ICP-MS	(Moynier et al. 2007)
Orgueil	992	0.81	0.39	0.10	1.51	0.20	110	VG-54E and Micromass Sector	(Shukolyukov and Lugmair, 2006)
Orgueil	39.0	0.93	0.46	0.06	1.94	0.12	n.d.	Triton TIMS	(Kadlag et al., 2019)
Orgueil	22.6	0.86	0.19	0.05	1.51	0.05	5	Neptune Plus MC-ICP-MS	This study
Orgueil	22.6		0.19	0.01	1.50	0.08	5	Neptune Plus MC-ICP-MS	This study
Orgueil	46	0.80	0.25	0.06	1.56	0.06	3-5	Triton TIMS	(Trinquier et al., 2007, 2008)
Orgueil	~100	0.85	0.20	0.05	1.55	0.13	6-10	Triton TIMS	(Qin et al., 2010)
Orgueil			0.24	0.05	1.69	0.09	6-10	Triton TIMS	(Qin et al., 2010)
Orgueil	320	0.81	0.25	0.03	1.56	0.06	n.d.	Triton TIMS	(Petitat et al., 2011)
Averaged value		0.85	0.29	0.07	1.60	0.10			
Ivuna	225	0.82	0.41	0.11	1.59	0.24	71	VG-54E and Micromass Sector	(Shukolyukov and Lugmair, 2006)
Ivuna	30.5	0.94	0.30	0.17	1.79	0.20	n.d.	Triton TIMS	(Kadlag et al., 2019)
Ivuna		0.75	0.16	0.02	1.55	0.05	10	Neptune Plus MC-ICP-MS	(Schiller et al., 2014)
Ivuna	~200	0.85	0.25	0.06	1.59	0.14	12	Triton TIMS (Total Evaporation)	This study; (van Kooten et al., 2016)
					1.30	0.09		Triton Plus TIMS	(Williams et al., 2020)
Averaged value		0.84	0.28	0.10	1.56	0.15			
Allende	~100	0.42	0.08	0.01	0.87	0.08	5	Neptune Plus MC-ICP-MS	This study
Allende	~100		0.10	0.02	0.90	0.02	5	Veptune Plus MC-ICP-MS	This study
Allende	~100		0.11	0.02	0.94	0.02	5	Neptune Plus MC-ICP-MS	This study
Allende	~100		0.14	0.03	0.95	0.08	5	Neptune Plus MC-ICP-MS	This study
Allende	~100		0.14	0.00	0.96	0.00	5	Neptune Plus MC-ICP-MS	This study
Allende	~50	0.42	0.10	0.02	1 10	0.00	9		$(\mathbf{Z}_{hu} \text{ ot al. } 2020\text{ b})$
Allende	~50	0.42	0.10	0.00	1.10	0.00	0		$(Z_{\rm H})$ et al. 2020D)
Allende		0.42	0.16	0.06	0.88	0.17	15	Inton TIMS (Total Evaporation)	(Znu et al., 2020a)

Table 3 Comparison of ⁵⁵Mn/⁵²Cr and Cr isotope data for meteorites that have been measured multiple times here and/or in the literature.

Allende	81	0.45	0.04	0.06	0.86	0.09	3-5	Triton TIMS	(Trinquier et al., 2007, 2008)
Allende	~100		0.14	0.04	0.98	0.14	6-10	Triton TIMS	(Qin et al., 2010)
Allende	~100		0.13	0.05	0.92	0.13	6-10	Triton TIMS	(Qin et al., 2010)
Allende		0.43	0.14	0.11			5	Nu Plasma HR MC-ICP-MS	(Moynier et al., 2007)
Allende	2380	0.43	0.10	0.09	0.85	0.17	70	VG-54E and Micromass Sector	(Shukolyukov and Lugmair, 2006)
Allende	44.3	0.51	0.07	0.08	1.24	0.24	n.d.	Triton TIMS	(Kadlag et al., 2019)
Allende					0.86	0.09	4	Triton Plus TIMS	(Williams et al., 2020)
Averaged value		0.44	0.11	0.02	0.95	0.06			
Vigarano	21.0	0.42	0.08	0.02	0.84	0.04	5	Neptune Plus MC-ICP-MS	This study
Vigarano	~100	0.57	0.22	0.07	0.91	0.12	6-10	Triton TIMS	(Qin et al., 2010)
Vigarano	~100		0.14	0.05	0.82	0.13	6-10	Triton TIMS	(Qin et al., 2010)
Vigarano			0.23	0.08			12	Nu Plasma HR MC-ICP-MS	(Moynier et al. 2007)
Averaged value		0.49	0.17	0.07	0.86	0.05			
Leoville	24.8	0.41	0.08	0.04	0.81	0.10	5	Neptune Plus MC-ICP-MS	This study
Leoville	~100	0.46	0.12	0.04	0.71	0.15	6-10	Triton TIMS	(Qin et al., 2010)
Averaged value		0.43	0.10	0.04	0.76	0.10			
Renazzo	12	0.54	0.20	0.10	1.30	0.21	3-5	Triton TIMS	(Trinquier et al., 2007, 2008)
Renazzo					1.22	0.10	4	Triton Plus TIMS	(Sanborn et al., 2019)
Averaged value		0.54	0.20	0.10	1.26	0.08			
Jbilet Winselwan	19.5	0.56	0.12	0.02	0.82	0.04	5	Neptune Plus MC-ICP-MS	This study
Jbilet Winselwan	~150	0.63	0.19	0.06	1.01	0.12	16	Triton TIMS (Total Evaporation)	(van Kooten et al. 2020)
Averaged value		0.59	0.16	0.07	0.92	0.18			
Murchison		0.64	0.27	0.06	1.01	0.05	3-5	Triton TIMS	(Trinquier et al., 2007 2008)

Murchison	~150	0.67	0.19	0.04	0.93	0.07	16	Triton TIMS (Total Evaporation)	(van Kooten et al. 2020)
Murchison	~100	0.60	0.17	0.08	0.97	0.20	6-10	Triton TIMS	(Qin et al., 2010)
Murchison			0.16	0.04	0.89	0.08	4	Triton Plus TIMS	(Jenniskens et al., 2012)
Averaged value		0.64	0.20	0.05	0.95	0.05			
Murray	101	0.64	0.27	0.09	1.13	0.21	93	VG-54E and Micromass Sector	(Shukolyukov and Lugmair, 2006)
Murray	~150	0.63	0.18	0.03	0.85	0.10	16	Triton TIMS (Total Evaporation)	(van Kooten et al. 2020)
Averaged value		0.63	0.23	0.09	0.99	0.27			
Kainsaz	1030	0.54	0.20	0.10	1.02	0.24	91	VG-54E and Micromass Sector	(Shukolyukov and Lugmair, 2006)
Kainsaz	~100	0.52	0.13	0.06	0.87	0.18	6-10	Triton TIMS	(Qin et al., 2010)
Averaged value		0.53	0.17	0.07	0.95	0.15			
Lancé	8	0.47	-0.04	0.07	0.57	0.11	3-5	Triton TIMS	(Trinquier et al., 2007, 2008)
Lancé		0.50	0.20	0.09			15	Nu Plasma HR MC-ICP-MS	(Moynier et al. 2007)
Karoonda	33.9	0.41	0.04	0.02	0.50	0.09	5	Neptune Plus MC-ICP-MS	This study
Karoonda	25	0.48	0.14	0.06	0.63	0.09	3-5	Triton TIMS	(Trinquier et al., 2007, 2008)
Averaged value		0.44	0.09	0.10	0.57	0.13			
EET 92002	34.6	0.41	0.10	0.04	0.52	0.09	5	Neptune Plus MC-ICP-MS	This study
EET 92002	~100	0.46	0.02	0.05	0.33	0.12	6-10	Triton TIMS	(Qin et al., 2010)
Averaged value		0.43	0.06	0.08	0.43	0.19			
Qingzhen			0.12	0.04	0.00	0.05	5	Triton TIMS	(Mougel et al., 2018)
Qingzhen	260	0.72	0.20	0.06			3-5	Triton TIMS	(Trinquier et al., 2007, 2008)
Qingzhen	260	0.72	0.17	0.06	-0.02	0.08	3-5	Triton TIMS	(Trinquier et al., 2007, 2008)
Average value		0.72	0.16	0.05	-0.01	0.02		Triton TIMS	(Trinquier et al., 2007, 2008)

Kota-Kota		n.d.	0.11	0.04	0.00	0.08	6	Triton TIMS	(Mougel et al., 2018)
Kota-Kota		0.69	0.18	0.06	-0.02	0.21	3-5	Triton TIMS	(Trinquier et al., 2007, 2008)
Kota-Kota		0.69	0.17	0.06	0.04	0.07	3-5	Triton TIMS	(Trinquier et al., 2007, 2008)
Averaged value		0.69	0.15	0.04	0.01	0.03			
Abee			0.05	0.04	-0.02	0.08	4	Triton TIMS	(Mougel et al., 2018)
Abee	18	0.93	0.26	0.08	-0.06	0.12	3-5	Triton TIMS	(Trinquier et al., 2007, 2008)
Averaged value		0.93	0.16	0.21	-0.04	0.04			
SAH 97096	33.2	0.65	0.25	0.03	0.17	0.08	5	Neptune Plus MC-ICP-MS	This study
SAH 97096		0.65	0.19	0.04	-0.01	0.14	14	Triton TIMS (Total Evaporation)	(Zhu et al., 2020a)
Averaged value		0.65	0.22	0.06	0.08	0.18			
MIL 05082	96.4*	0.37	0.20	0.01	1.50	0.09	5	Neptune Plus MC-ICP-MS	This study
QC 001	39.8	0.47	0.19	0.04	1.45	0.06	5	Neptune Plus MC-ICP-MS	This study
HaH 237 (mostly silicate)	32.9	0.09	0.05	0.03	1.42	0.04	5	Neptune Plus MC-ICP-MS	This study
HaH 237	679	0.08	-0.15	0.09	0.87	0.19	148	VG-54E and Micromass Sector	(Shukolyukov and Lugmair, 2006)
Bencubbin (silicate < metal)		0.04	-0.05	0.06	1.11	0.09	3-5	Triton TIMS	(Trinquier et al., 2007, 2008)
Bencubbin (metal < silicate)		0.48	0.12	0.09	1.13	0.09	3-5	Triton TIMS	(Trinquier et al., 2007, 2008)
Gujba metal chondrule		0.20	-0.03	0.13	1.07	0.27	3-5	Triton TIMS	(Trinquier et al., 2007, 2008)

969 Note: The error for Mn/Cr ratios is 5%. The mass marked * means a chip. The uncertainty of average values are 95% confidence intervals.

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		Т	able 4 The	Mn/Cr ratio	, and Cr a	and O iso	tope data	a for all o	chondrites			
Carbonaceous Chondrites	⁻ all/Find	Mass (mg)	Туре	55Mn/52Cr	ε ⁵³ Cr	Error	ε ⁵⁴ Cr	Error	N/[ref.]	$\Delta^{17}O$	Error	[ref.]
Orgueil	fall	22.6	CI1	0.85	0.29	0.07	1.60	0.10	[This study, 2, 4, 5, 21, 30, 31, 32]	0.39	0.08	[9]
Ivuna	fall		CI1	0.84	0.28	0.10	1.56	0.15	[1, 22, 29, 30, 32] 0.47		0.08	[9]
CI average				0.84	0.28	0.01	1.56	0.05	2SD			
PCA 91467*	find	28.5	CH3	0.36	0.13	0.00*	1.50	0.08	5	-1.47	0.08	[9]
A-881020*	find	40.5	CH3	0.33	0.11	0.03	1.49	0.06	5			
CH average				0.34	0.12	0.03	1.50	0.01	2SD			
MIL 05082*	find	96.4	CB3	0.37	0.20	0.01	1.50	0.09	5			
QC 001*	find	39.8	CBa3	0.47	0.19	0.04	1.45	0.06	5			
Bencubbin*	find		CBa3				1.12	0.03	[2]	-1.75	0.08	[9]
Gujba*	fall		CBa3				1.29	0.07	[3]	-2.38	0.08	[10]
HaH 237*	find	32.9	CBb3	0.09	0.05	0.03	1.42	0.04	5	-0.77	0.08	[11]
CB average				0.42	0.20	0.01	1.36	0.30	2SD			
GRO 95577	find	20.5	CR1	0.51	0.15	0.02	1.25	0.06	5	-0.45	0.08	[9]
Al Rais	fall	23.2	CR2-an	0.60	0.19	0.01	1.24	0.11	5	-1.01	0.08	[9]
Renazzo	fall		CR2	0.54	0.20	0.10	1.26	0.08	[2, 4, 24]	-0.96	0.08	[9]
GRA 06100	find		CR2	0.58	0.25	0.05	1.32	0.11	[5]	-1.80	0.03	[20]
NWA 6043	find		CR2	0.58	0.07	0.07	1.24	0.10	[22, 23]			
EET 92161	find		CR2	0.50	0.21	0.04	1.19	0.12	[22, 23]			
NWA 7837	find		CR2	0.39	0.02	0.04	1.06	0.08	[22, 23]			
GRA 95229	find		CR2				1.18	0.07	[29]	-2.19	0.01	[29]
QUE 99177	find		CR2				1.43	0.12	[29]	-2.89	0.12	[29]
LAP 02342	find		CR2				1.49	0.11	[29]	-2.45	0.06	[29]
NWA 6921	find		CR6				1.32	0.09	[24,29]	-1.74	0.01	[29]
NWA 7317	find		CR6				1.32	0.09	[24]			
CR average				0.53	0.16	0.16	1.28	0.23	2SD			
SCO 06043	find	21.7	CM1	0.60	0.22	0.02	1.13	0.12	5			

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Nogoya	fall	24.3	CM2	0.59	0.18	0.05	0.76	0.04	5	-2.00	0.08	[9]
Banten	fall	21.7	CM2	0.58	0.12	0.03	0.86	0.05	5	-2.97	0.08	[9]
Aguas Zarcas	fall	87.7	CM2	0.60	0.15	0.03	0.86	0.03	5	-2.78	0.20	[18]
Jbilet Winselwan	find	19.5	CM2	0.59	0.16	0.10	0.92	0.18	[This study, 23]	-4.03	0.55	[18]
Paris	find		CM2	0.62	0.16	0.05	0.93	0.09	[6]	-3.39	0.39	[19]
NWA 8157	find		CM2	0.62	0.20	0.11	1.01	0.18	[6]	-4.05	0.55	[18]
Murchison	fall		CM2	0.64	0.20	0.11	0.95	0.04	[2, 4, 5, 23, 28]	-2.60	0.08	[12]
Mighei	fall		CM2	0.63	0.18	0.03	0.74	0.10	[23]	-2.50	0.08	[9]
Cold Bokkeveld	fall		CM2	0.65	0.07	0.03	0.81	0.12	[23]	-2.45	0.08	[9]
Murray	fall		CM2	0.63	0.23	0.09	0.99	0.27	[23, 30]	-3.07	0.08	[9]
Maribo	fall		CM2	0.65	0.29	0.04	1.13	0.15	[23]			
Diepenveen	fall		CM2-an		0.13	0.05	0.85	0.10	[27]			
CM average				0.62	0.17	0.11	0.92	0.26	2SD			
Allende	fall	~100	CV3-oxA	0.44	0.11	0.02	0.95	0.06	[This study,2,4,5,25,26,28,30,31,32]	-3.62	0.06	[13]
Bali	fall	42.3	CV3-oxB	0.45	0.13	0.04	1.10	0.06	5	-3.30	0.17	[13]
Mokoia	fall	20.5	CV3-oxB	0.45	0.11	0.04	1.00	0.01	5	-3.18	0.07	[13]
Leoville	find	24.8	CV3-red	0.43	0.10	0.04	0.76	0.10	[This study, 5]	-4.69	0.17	[13]
Vigarano	fall	21.0	CV3-red	0.49	0.13	0.10	0.85	0.02	[This study, 5, 31]	-4.25	0.03	[13]
Kaba	fall	19.7	CV3-oxB	0.43	0.08	0.05	0.70	0.07	5	-3.43	0.31	[13]
CV average				0.45	0.11	0.04	0.89	0.30	2SD			
MIL 07193	find	20.9	CO3	0.48	0.16	0.02	1.22	0.04	5			
DOM 10104	find	24.7	CO3	0.48	0.09	0.03	0.80	0.06	5			
Ornans	fall	66.2	CO3	0.50	0.12	0.01	0.90	0.03	5	-4.45	0.08	[9]
Felix	fall		CO3	0.47	0.07	0.06	0.63	0.09	[2, 4]	-4.59	0.08	[9]
Kainsaz	fall		CO3	0.53	0.17	0.07	0.95	0.15	[5, 31]	-4.72	0.08	[9]
CO average				0.49	0.11	0.07	0.90	0.43	2SD			
ALH 85002	find	45.8	CK4	0.42	0.06	0.02	0.46	0.05	5			

EET 92002	find	34.6	CK5	0.43	0.06	0.08	0.43	0.19	[This study, 5]			
LEW 87009	find	43.8	CK6	0.44	0.08	0.04	0.58	0.05	5	-4.32	0.08	[9]
CK average				0.43	0.07	0.03	0.51	0.15	2SD			
Ordinary Chondrites												
Roosevelt	find		H3.4	0.65	0.17	0.02	-0.44	0.03	[7]			
Brownsflied	find		H3.7	0.67	0.18	0.02	-0.44	0.03	[7]			
Ochansk	fall		H4	0.64	0.15	0.02	-0.40	0.03	[7]	0.82	0.08	[14]
LAP 03601	find		H4	0.78	0.21	0.06	-0.28	0.11	[5]			
Ste. Marguerite	fall		H4	0.63	0.13	0.06	-0.39	0.07	[2,4]			
Beaver Creek	fall		H4	0.70	0.17	0.02	-0.40	0.04	[7]	0.76	0.08	[14]
Bath	fall		H4	0.68	0.17	0.02	-0.36	0.04	[7]	0.71	0.08	[14]
Menow	fall		H4	0.66	0.12	0.02	-0.43	0.03	[7]			
Forest city	fall		H5	0.71	0.18	0.02	-0.36	0.03	[7]	0.75	0.08	[14]
Estacado	find		H6	0.67	0.15	0.02	-0.35	0.06	[7]			
Aarhus	fall		H6	0.68	0.17	0.01	-0.41	0.03	[7]			
Kernouve	fall		H6	0.72	0.19	0.06	-0.37	0.07	[2,4]			
Portales Valley	fall		H6/7	0.74	0.19	0.02	-0.37	0.04	[7]			
H average				0.69	0.17	0.05	-0.38	0.09	2SD			
QUE 97008	find		L3	0.72	0.17	0.06	-0.42	0.14	[5]			
Bjurböle	fall		L4	0.81	0.20	0.06			[4]	1.00	0.08	[14]
Knyahinya	fall		L5	0.70	0.15	0.06	-0.38	0.08	[2,4]	1.05	0.08	[14]
Holbrook	fall		L6	0.80	0.23	0.06			[4]			
L average				0.76	0.19	0.07	-0.40	0.06	2SD			
Chainpur	fall		LL3-4	0.85	0.24	0.06	-0.47	0.07	[2,4]			
Soko-Banja	fall		LL4	0.82	0.34	0.06			[4]	1.32	0.08	[14]
GRO 95552	find		LL4	0.78	0.19	0.04	-0.33	0.10	[5]			
Olivenza	fall		LL5	0.76	0.23	0.06			[4]	1.11	0.08	[14]

Guidder	fall		LL5	0.99	0.21	0.06			[4]	1.19	0.08	[14]
Saint-Se'verin	fall		LL6	0.77	0.28	0.06	-0.41	0.10	[2,4]	1.16	0.08	[14]
LL average				0.83	0.25	0.04	-0.40	0.14	2SD			
OC average				0.74	0.19	0.10	-0.39	0.09	2SD			
Enstatite Chondrites												
SAH 97096	find	33.2	EH3	0.65	0.22	0.06	0.08	0.18	[This study, 26]	-0.07	0.02	[17]
Qingzhen	fall		EH3	0.72	0.15	0.07	-0.01	0.02	[2,4,8]	-0.03	0.08	[15]
Kota-Kota	find		EH3	0.69	0.14	0.07	0.01	0.01	[2,4,8]	-0.15	0.08	[15]
ALHA 77295	find		EH3	0.71	0.14	0.06	0.05	0.14	[5]			
Abee	fall		EH4	0.93	0.16	0.21	-0.04	0.04	[2,4,8]	0.19	0.19	[17]
Indarch	fall		EH4	0.91	0.21	0.07	0.05	0.14	[5]	0.12	0.08	[15]
EH average				0.77	0.17	0.07	0.02	0.09	2SD			
MAC 88136	find		EL3	0.72	0.15	0.03	0.02	0.09	[5]	-0.11	0.04	[17]
MAC 88184	find		EL3		0.20	0.03	0.11	0.07	[8]			
Hvittis	fall		EL6	0.67	0.14	0.07	-0.01	0.17	[2,4]	0.07	0.08	[15]
Pillistfer	fall		EL6	0.59	0.15	0.05	0.09	0.08	[2,4]	0.02	0.08	[15]
LON 94100	find		EL6	0.56	0.17	0.04	-0.02	0.14	[5]			
Eagle	fall		EL6		0.14	0.05	-0.07	0.07	[8]			
EL average				0.63	0.16	0.05	0.02	0.14	2SD			
EC average				0.71	0.16	0.06	0.02	0.11	2SD			

973 Note: The references: [1] Schiller et al. (2014), [2] Trinquier et al. (2007), [3] Yamashita et al. (2010), [4] Trinquier et al. (2008a), [5] Qin et al. (2010), [6] Göpel et al. (2015),

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Moynier et al. (2007) and [32] Kadlag et al. (2019).
The errors for ⁵⁵ Mn/ ⁵² Cr ratios are 5%. The ⁵⁵ Mn/ ⁵² Cr for H chondrite samples in [7] Pedersen et al. (2019) are re-measured (same solution) by MC-ICP-MS in this study.
The Fe/Cr (atom) ratios for CB and CH chondrites: PCA 91467, 97; A-881020, 115; QC 001, 25; HaH 237, 235
The errors for O isotope data in [18] are 1SD of multiple O isotope data, and in [10] and [11] are quoted as 0.08 from [9].
* The ε ⁵³ Cr 2SE uncertainty for PCA 91467 is less than 0.004. The CB and CH samples in this study, including PCA 91467, MIL 05082, A-881020, QC 001 and HaH 237
may not represent the bulk composition of the parent chondrites given the clear evidence for sample heterogeneity in CB and CH chondrites and the fact that the mass of our
samples is relatively small. The ε^{54} Cr data for Bencubbin is the average (with 2SD uncertainty) of two data (silicate > metal and silicate < metal) in Trinquier et al. (2007).
The ε^{54} Cr data for Gujba is the average (with 2SD uncertainty) of the data for chondrules and metals in Yamashita et al. (2010).

 Table 5. The Cr and O isotope compositions of various Solar System materials not analyzed here
 (including: Rumuruti chondrites, terrestrial planets and achondrite parent bodies).

Meteorites/Planets	ε ⁵⁴ Cr	Error (2SD)	N	ref.	Δ ¹⁷ Ο	error	N	ref.
Rumuruti (R) chondrites	-0.06	0.08	12	[35]	2.72	0.31	24	[36]
Earth	0.09	0.12	15	[2,8,25, this study]	-0.01	0.01	14	[26]
Moon	0.09	0.08	/	[8]	-0.01	0.02	22	[26]
Aubrites	-0.16	0.19	1	[2]	-0.01	0.11	13	[26]
Mars (SNC meteorites)	-0.17	0.15	18	[2,33]	0.28	0.08		[34]
Angrite parent body (APB)	-0.42	0.13	8	[2,22]	-0.07	0.01	5	[26]
Brachinites	-0.44	0.23	2	[29]	-0.17	0.09	2	[29]
Winonaites	-0.53	0.02	3	[23]	-0.51	0.08	16	[26]
Acapulcoite-lodranite Clan	-0.61	0.19	6	[23]	-1.12	0.36	23	[26]
Vesta (HED meteorites)	-0.73	0.08	9	[2]	-0.24	0.02	105	[26]
Ureilite parent body (UPB)	-0.95	0.15	18	[24,25]	-0.96	1.00	42	[26]

992 Note: The reference sources are: [2] Trinquier et al. (2007), [8] Mougel et al. (2018), [22] Zhu et al. 993 (2019b), [23] Li et al. (2018), [24] Zhu et al. (2020b), [25] Yamakawa et al. (2010), [26] Greenwood et 994 al. (2017), [25] Zhu et al. (2020b), [29] Williams et al. (2020), [33] Kruijer et al. (2020), [34] Ireland et 995 al. (2020), [35] Zhu et al. (2021) and [36] Bischoff et al. (2011). The ε^{54} Cr values are calculated by 996 mean average. The O isotope data did not consider Cumberland falls for aubrites and some isotopically 997 anomalous eucrites for Vesta. The N (/) of ϵ^{54} Cr value of Moon means it was modeled by the 998 correlation of the ε^{53} Cr and ε^{54} Cr of lunar samples that had been influenced by cosmogenic effects. The 999 ε^{54} Cr for Nakhla (Mars) is chosen from [2], due to the smaller uncertainty.

Sample Name	Petrology	ε ⁵³ Cr	2se	ε ⁵⁴ Cr	2se	N/[ref.]
NIST 3112a	artificial standard	0.04	0.03	0.03	0.05	5
SCP-Cr	artificial standard	0.02	0.02	0.05	0.08	5
DTS-1	dunite	0.05	0.03	0.16	0.06	5, 2SD
DTS-2b	dunite	0.04	0.01	0.08	0.06	[1]
PCC-1	peridotite	0.05	0.03	0.10	0.07	[25]
KOLA15-UB	peridotite	0.00	0.03	0.05	0.06	[8]
BM31	peridotite	0.07	0.04	0.13	0.08	[8]
BM23	peridotite	-0.01	0.05	0.03	0.08	[8]
Tibet chromite	chromite	0.03	0.07	0.08	0.08	[2,4]
Deccan basalt	basalt	0.03	0.08	0.08	0.15	[2,4]
Erta Ale tholeite	basalt	-0.02	0.06	-0.02	0.08	[2,4]
10PUB22-07	basalt	0.08	0.05	0.19	0.07	[8]
KBD408729	basalt	0.07	0.03	0.14	0.07	[8]
NIST 688	basalt	0.06	0.04	0.11	0.07	[8]
CV-SN-98-19	basalt	0.01	0.05	0.12	0.11	[8]
BE-N	basalt	0.12	0.03	0.17	0.07	[8]
BHVO-2	basalt	0.02	0.03	0.07	0.07	[8]
average		0.04	0.02	0.09	0.03	2SE, N =1

1002 Note: The average data for terrestrial samples (N =15) do not include the two artificial standards. The

reference sources are [1] Schiller et al. (2014), [2] Trinquier et al. (2007), [4] Trinquier et al. (2008a), [8]
Mougel et al. (2018) and [25] Zhu et al. (2020b).

Table 7a Parameters for theoretical calculation for Cr isotope fractionation

				-	
	Mass of isotopes	Isotope abundance	CrO	CrO ₂	CrO ₃
⁵⁰ Cr	49.946	4.345%	65.941	81.936	97.931
⁵² Cr	51.941	83.789%	67.935	83.930	99.925
⁵³ Cr	52.941	9.501%	68.936	84.930	100.925
⁵⁴ Cr	53.939	2.365%	69.934	85.929	101.924
¹⁶ O	15.995				

Table 7b The calculated Cr isotope data based on different Cr oxides and different laws (kinetic and equilibrium), assuming the initial ⁵⁰Cr/⁵²Cr fractionation is 1000 ppm.

	50/52	53/52	54/52	β ⁵³ Cr	β ⁵⁴ Cr	Δ^{53} Cr	Δ^{54} Cr	54/53 factors
Kin. Cr	1000	-487.09	-964.17	-2.05	-1.04	0.00	0.00	
Kin. CrO	1000	-490.46	-972.94	-2.04	-1.03	-3.37	-8.77	2.606
Kin. CrO ₂	1000	-492.55	-978.41	-2.03	-1.02	-5.45	-14.24	2.611
Kin. CrO ₃	1000	-493.97	-982.14	-2.02	-1.02	-6.88	-17.97	2.614
Equ. Cr	1000	-473.09	-927.79	-2.11	-1.08	14.00	36.38	2.598
Equ. CrO	1000	-479.68	-944.75	-2.08	-1.06	7.42	19.41	2.617
Equ. CrO ₂	1000	-483.78	-955.40	-2.07	-1.05	3.32	8.76	2.643
Equ. CrO ₃	1000	-486.58	-962.71	-2.06	-1.04	0.51	1.46	2.838

1011 Note: The fractionation factors, β^{53} Cr and β^{54} Cr, are calculated by equations (9 and 12) in Young and

1012 Galy (2004).



1015

1016 Figure 1. The ε^{54} Cr variations in different groups of chondrites (the data and reference sources can be 1017 found in tables 4-6), which occur in the order $CI = CH \ge CB \ge CR > CM \approx CV \approx CO \ge CK > EC \approx$ 1018 RC > OC. The color shades indicate the 2SD variation of different chondrite groups (the blue and green 1019 shades show the 2SD uncertainty of all the ECs and OCs respectively; we use the external 1020 reproducibility 0.07 to represent the grouping uncertainties of CIs and CHs since the 2SD uncertainty 1021 are less than 0.07. The middle bold dashed line at $\epsilon^{54}Cr = +0.3$ is the $\epsilon^{54}Cr$ boundary for 1022 non-carbonaceous chondrite (NC) and carbonaceous chondrite (CC) reservoirs. All CK, RC, OC and 1023 EC groups have homogeneous ϵ^{54} Cr within their respective groups. The CV and CK chondrites have 1024 distinct ε^{54} Cr values from one another, whereas the ε^{54} Cr for CV, CM and CO chondrites overlap. EC 1025 and RC have similar ϵ^{54} Cr features with the Earth-Moon system.





1029 Figure 2. The ε^{54} Cr and Δ^{17} O compositions of the CCs, OCs, ECs (filled circles), as well as achondrites, 1030 terrestrial and lunar samples (gray ellipses). The data sources for planets and achondrites are shown in 1031 Table 5. NC: non-CCs; APB: angrite parent body; UPB: ureilite parent body. The colored circles 1032 represent the same samples that are shown in Figure. 1. It should be noted that the correlation between 1033 ε^{54} Cr and Δ^{17} O values in CCs is not as strong (R² = 0.58) as that described in Trinquier et al. (2007) 1034 and Warren (2011). The ε^{54} Cr gap between the CC and NC reservoirs is +0.3, because some terrestrial 1035 samples and the ECs have ε^{54} Cr values in the range 0 – 0.2, and no CCs possess ε^{54} Cr values < 0.4.

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- 1038



1040Figure 3. 55 Mn/ 52 Cr vs. ε^{53} Cr in the CCs (a), OCs (b) and ECs (c). In (a), the gray circles and dashed line are from1041Moynier et al. (2007) and Shukolyukov and Lugmair (2006), who reported well-defined Mn-Cr isochrons.1042However, the additional data presented here (colored data points; the colors for chondrite groups are the same as in1043Figure 1) do not reproduce this correlation line. (a) does not include the Mn-Cr data for the silicate parts of CB1044chondrites. The best fit Mn-Cr correlation line in (b) may be a mixing line rather than an isochron.

Slope = 0.285 ± 0.078 0.4 53 Mn/ 55 Mn = (3.23 ± 0.89) × 10⁻⁶ Initial ϵ^{53} Cr = 0.00 \pm 0.05 MSWD = 11; N = 85 Absolute age": 4563.4 ± 1.5 Ma 0.3 CB ε⁵³Cr 0.2 HO СН EH 0.1 0.0 C -0.1 0.6 0.7 0.0 0.1 0.2 0.3 0.4 0.5 0.8 0.9 1.0 1.1 1.2 55Mn/52Cr

⁵³Mn-⁵³Cr Diagram of All Chondrites

1046Figure 4. All of the ${}^{55}Mn/{}^{52}Cr$ and ${\epsilon}^{53}Cr$ data for the different chondrite groups. The warm-color circles are the1047CCs, while the black, green and blue ones are the RCs, OCs and ECs, respectively. The data are from Table 4, and1048regressed by Model 3 (maximum likelihood regression with overdispersion), *Isoplot R* (Vermeesch, 2018). This1049figure does not include the Mn-Cr data for the silicate parts of CB chondrites. There is a positive trend (gray1050dashed line) between ${}^{55}Mn/{}^{52}Cr$ and ${\epsilon}^{53}Cr$ with a slope of 0.285 ± 0.078 (MSWD =11, N = 85; some literature data1051do not have ${}^{55}Mn/{}^{52}Cr$ ratio information) regressed by model 3, IsoplotR. This Mn-Cr correlation line is interpreted1052as a mixing trend and, as such, does not have chronological significance.

1053



ε⁵³Cr Deficit between Chondrites and Bulk Earth



1055 Figure 5. The ε^{53} Cr and ε^{54} Cr values for terrestrial rocks (Table 5) and chondrites. The black circles and squares 1056 are terrestrial crustal and mantle rocks respectively, while the black triangles are artificial standards (i.e., NIST 1057 SRM 979 and NIST SRM 3112a). The ε^{53} Cr and ε^{54} Cr values for terrestrial rocks are correlated with a slope of 1058 1.37 ± 0.65 (N = 15, MSWD = 0.1), regressed by Model 1, *Isoplot R* (Vermeesch, 2018). This correlation indicates 1059 the residual mass-dependent Cr isotope fractionation. The gray and colorful bars indicate the average ε^{53} Cr values 1060 (with 2SE uncertainty) for bulk silicate Earth (BSE; N =15) and chondrites (N = 88), respectively. The two 1061 artificial standards are not considered to represent the Cr isotope composition of Earth. There is a ε^{53} Cr deficit of 1062 0.12 ± 0.02 between BE and chondrites, which potentially indicate an early volatile fractionation of proto-Earth. 1063 The ⁵⁵Mn/⁵²Cr for BE of 0.22–0.54, was estimated by Sun (1982), Wang et al. (2018), Wänke and Dreibus (1988) 1064 and (Palme and O'Neill, 2014), while the ⁵⁵Mn/⁵²Cr for chondrites are from those of the ECs (0.71; having the 1065 same isotope compositions with Earth for multiple elements) and CIs (0.82; are regarded to represent the chemical 1066 composition of the Solar System). In this figure, ⁵⁵Mn/⁵²Cr ratios of 0.71-0.82 are only from ECs and CI 1067 chondrites, but note that some other groups of chondrites (e.g., CV, CO and CK in Figure 4) may have an 1068 Earth-like ⁵⁵Mn/⁵²Cr ratios. No systematic differences were observed in data measured by MC-ICP-MS in this 1069 study and by TIMS in the literature. 1070



1073 Figure 6. Calculated Δ^{53} Cr and Δ^{54} Cr (for evaporation of only Cr⁺ by kinetic isotope fractionation) 1074 using different Cr oxides (CrO, CrO₂ and CrO₃) and different fractionation laws (kinetic and 1075 equilibrium). The data sources are from Table 7a and 7b. It should be noted that the variation factor 1076 (slope) between Δ^{53} Cr and Δ^{54} Cr is 2.61 that is highly consistent with the slope (2.6) between ϵ^{53} Cr and 1077 ϵ^{54} Cr values of multiple measurements for NIST 3112a (Qin et al., 2010).

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