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1 **Tracing the origin and core formation of the enstatite achondrite**
2 **parent bodies using Cr isotopes**

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20

21 **Abstract:**

22 Enstatite achondrites (including aubrites) are the only differentiated meteorites
23 that have similar isotope compositions to the Earth-Moon system for most of the
24 elements. However, the origin and differentiation of enstatite achondrites and their
25 parent bodies remain poorly understood. Here, we report high-precision
26 mass-independent and mass-dependent Cr isotope data for 10 enstatite achondrites,
27 including eight aubrites, Itqiy and one enstatite-rich clast in Almahatta Sitta, to further
28 constrain the origin and evolution of their parent bodies. The $\epsilon^{54}\text{Cr}$ (per 10,000
29 deviation of the mass bias corrected $^{54}\text{Cr}/^{52}\text{Cr}$ ratio from a terrestrial standard)
30 systematics define three groups: main-group aubrites with $\epsilon^{54}\text{Cr} = 0.06 \pm 0.12$ (2SD, N
31 =7) that is similar to the enstatite chondrites and the Earth-Moon system, Shallowater
32 aubrite with $\epsilon^{54}\text{Cr} = -0.12 \pm 0.04$ and Itqiy-type meteorites with $\epsilon^{54}\text{Cr} = -0.26 \pm 0.03$
33 (2SD, N=2). This shows that there were at least three enstatite achondrite parent bodies
34 in the Solar System. This is confirmed by their distinguished mass-dependent Cr
35 isotope compositions ($\delta^{53}\text{Cr}$ values): 0.24 ± 0.03 ‰, 0.10 ± 0.03 ‰ and -0.03 ± 0.03 ‰
36 for main-group, Shallowater and Itqiy parent bodies, respectively. Aubrites are
37 isotopically heavier than chondrites ($\delta^{53}\text{Cr} = -0.12 \pm 0.04$ ‰), which likely results from
38 the formation of an isotopically light sulfur-rich core. We also obtained the abundance
39 of the radiogenic ^{53}Cr (produced by the radioactive decay of ^{55}Mn , $T_{1/2} = 3.7$ million
40 years). The radiogenic $\epsilon^{53}\text{Cr}$ excesses correlate with the $^{55}\text{Mn}/^{52}\text{Cr}$ ratios for aubrites

41 (except Shallowater and Bustee) and also the Cr stable isotope compositions ($\delta^{53}\text{Cr}$
42 values). We show that these correlations represent mixing lines that also hold
43 chronological significance since they are controlled by the crystallization of sulfides
44 and silicates, which mostly reflect the main-group aubrite parent body differentiation
45 at 4562.5 ± 1.1 Ma (i.e., 4.8 ± 1.1 Ma after Solar System formation). Furthermore, the
46 intercept of these lines with the ordinate axis which represent the initial $\varepsilon^{53}\text{Cr}$ value of
47 main-group aubrites (0.50 ± 0.16 , 2σ) is much higher than the average $\varepsilon^{53}\text{Cr}$ value of
48 enstatite chondrites (0.15 ± 0.10 , 2SD), suggesting an early sulfur-rich core formation
49 that effectively increased the Mn/Cr ratio of the silicate fraction of the main-group
50 aubrite parent body.

51

52 **1. Introduction**

53 Enstatite achondrites, e.g., aubrites, are the only differentiated meteorite group
54 that have similar oxygen isotope composition with that of the Earth-Moon system
55 (Clayton et al., 1984; Greenwood et al., 2017). This isotopic similarity also extends to
56 other isotope systems such as Ti, Cr or H, suggesting a close genetic link between the
57 solid precursors of aubrites and the Earth-Moon system (Barrat et al., 2016; Greenwood
58 et al., 2018; Greenwood et al., 2017; Newton et al., 2000; Piani et al., 2020; Trinquier et
59 al., 2007; Zhang et al., 2012), although aubrites have distinct chemical and
60 mineralogical composition and are more reduced than the Earth and the Moon (Keil,
61 2010a). Aubrites are more akin to the surface of Mercury (Ebel and Alexander, 2011;
62 Nittler and Weider, 2019; Udry et al., 2019; Wasson, 1988). As such, the origin and
63 evolution of aubrites may be closely linked to those of Earth, Moon and Mercury.

64 In detail, aubrites formed under highly reducing conditions with oxygen fugacities
65 (fO_2) ranging from ~ 2 to ~ 6 log units below the iron-wustite (IW) buffer (Keil, 1968,
66 2010b). They consist predominately of FeO-poor enstatite (75 ~ 98% vol.) and contain
67 minor silicate phases including plagioclase, diopside and forsterite (Watters and Prinz,
68 1979). Due to their formation under low fO_2 conditions, aubrites contain S-rich
69 accessory minerals, such as troilite (FeS), alabandite (MnS) and oldhamite (CaS). Most
70 aubrites are breccias and appear to originate from one or more parent bodies that
71 underwent extensive melting and differentiation (Keil, 1989). Their highly siderophile

72 elements (HSE) depletion supports the idea that their parent body/bodies underwent
73 core formation (van Acken et al., 2012a; van Acken et al., 2012b).

74 The formation of both aubrites and enstatite chondrites (ECs) occurred under
75 extremely reduced conditions (Keil, 2010b; Lodders et al., 1993) and from precursors
76 with similar isotopic compositions, suggesting a genetic relationship between the two
77 parent bodies (e.g., Lodders et al., 1993; van Acken et al., 2012b). However, other lines
78 of evidence including mineralogy, petrology, elemental abundances and cosmic ray
79 exposure ages are difficult to reconcile with formation of aubrites from an enstatite
80 chondrite precursor [see chapter 9.1 in Keil (2010b)]. Probing their relative genetic
81 relationships may be improved by a better understanding of the chronology of
82 formation of the aubrites, which is currently poorly defined. The lack of a radiogenic
83 ^{26}Mg deficit in aubrites, while they have sub-chondritic $^{27}\text{Al}/^{24}\text{Mg}$ ratios, suggests that
84 they formed after the decay of ^{26}Al , i.e. at least 3 million years (Ma) after Solar System
85 formation (Baker et al., 2012). Iodine-Xenon systematics of Shallowater suggests it
86 formed at 4562.4 ± 0.2 Ma (Pravdivtseva et al., 2017). Given that Shallowater is an
87 unusual enstatite achondrite, which likely originates from a distinct parent body
88 relative to other aubrites (Keil, 2010b; Moynier et al., 2011a), the timing of formation
89 of Shallowater cannot be generalized to the main-group aubrites. Thus, the timing of
90 aubrites and aubrite parent body formation remains largely unconstrained.

91 Chromium isotope systematics of aubrites may improve our understanding of the
92 origin of aubrites. 1) The abundance of the neutron-rich isotope ^{54}Cr (nucleosynthetic)

93 varies among different meteorites (Trinquier et al., 2007). This feature, expressed as the
94 $\epsilon^{54}\text{Cr}$ (per 10,000 deviation of the mass bias corrected $^{54}\text{Cr}/^{52}\text{Cr}$ ratio from to the
95 terrestrial standard NIST SRM 979) signature, has been established as a tracer of the
96 genetic relationships between meteorites (Qin et al., 2010; Trinquier et al., 2007; Zhu
97 et al., 2021b). 2) The radiogenic isotope ^{53}Cr is the daughter product of the short-lived
98 isotope ^{53}Mn (half-life of 3.7 million years (Holden, 1990). This decay system can
99 provide precise relative chronology of early Solar System events while ^{53}Mn was still
100 present (Birck and Allègre, 1988; Lugmair and Shukolyukov, 1998). The ^{53}Mn - ^{53}Cr
101 chronometer is especially sensitive to volatilization and magmatic processes, because
102 Mn and Cr possess different volatility and chemical behavior during magmatic
103 differentiation (Lodders, 2003; Sossi et al., 2019). This system has been successfully
104 applied to date the differentiation of other planetary objects, including: Vesta (Lugmair
105 and Shukolyukov, 1998; Trinquier et al., 2008b), angrite (Zhu et al., 2019b) and ureilite
106 parent bodies (Van Kooten et al., 2017; Yamashita et al., 2010; Zhu et al., 2020b). 3)
107 Mass-dependent Cr stable isotope variations (expressed as $\delta^{53}\text{Cr}$, the per mil variation
108 of the $^{53}\text{Cr}/^{52}\text{Cr}$ ratio relative to the terrestrial standard NIST SRM 979) can be used to
109 trace planetary processes, such as volatile depletion (Sossi et al., 2018; Zhu et al.,
110 2019c), magmatic differentiation (Bonnand et al., 2020a; Bonnand et al., 2020b;
111 Bonnand et al., 2016a; Shen et al., 2019; Sossi et al., 2018; Zhu et al., 2019c), and
112 possibly core formation (Moynier et al., 2011b).

113 So far, the only $\epsilon^{54}\text{Cr}$ value of aubrites has been reported for Bishopville ($-0.16 \pm$

114 0.19) (Trinquier et al., 2007). This isotopic composition overlaps with the $\epsilon^{54}\text{Cr}$
115 signatures of enstatite (0.02 ± 0.11 ; 2SD, N = 13), Rumuruti (-0.06 ± 0.08 ; 2SD, N =
116 12) and ordinary (-0.39 ± 0.09 ; 2SD, N = 18) chondrites (Mougel et al., 2018;
117 Pedersen et al., 2019; Qin et al., 2010; Trinquier et al., 2007; Zhu et al., 2021a; Zhu et
118 al., 2021b). Consequently, the currently available $\epsilon^{54}\text{Cr}$ datum for aubrite limits its use
119 to test genetic links to chondrites (e.g., enstatite and Rumuruti chondrites with $\epsilon^{54}\text{Cr}$
120 of ~ 0) and other achondrites (e.g., martian meteorites and angrites with $\epsilon^{54}\text{Cr}$ of ~ -0.2
121 and ~ -0.4 respectively; Trinquier et al., 2007; Zhu et al., 2019b), as well as possible
122 mantle heterogeneity of the aubrite parent body and whether there are multiple parent
123 bodies for enstatite achondrites (e.g., Zhu et al., 2020b). Shukolyukov and Lugmair
124 (2004) reported $\epsilon^{53}\text{Cr}$ values for three bulk aubrites that established a ^{53}Mn - ^{53}Cr
125 isochron corresponding to a $^{53}\text{Mn}/^{55}\text{Mn}$ initial ratio of $(2.0 \pm 0.5) \times 10^{-6}$ and an
126 absolute age of 4560.8 ± 1.4 Ma, relative to the U isotope corrected U-Pb age of the
127 D'Orbigny angrite (Amelin, 2008; Brennecka and Wadhwa, 2012; Glavin et al., 2004).
128 Although the limitation of a three-point isochron makes the age significance of the
129 isochron somewhat uncertain, these data provide promising evidence that Mn-Cr dating
130 could fruitfully be used to better constrain the timing of aubrite formation and
131 differentiation. Combined with Cr stable isotope information, these data may shed
132 more light on the magmatic evolution and differentiation history of the aubrite parent
133 bodies.

134 Aubrites are not the only class of enstatite achondrites. Recently, Harries and

135 Bischoff (2020) observed a new type of enstatite-rich differentiated meteorite clasts in
136 Almahatta Sitta, which are believed to come from another asteroid with a size of ~500
137 km. These clasts also show petrological affinity to Itqiy, an EH7-anomalous meteorite,
138 as they both underwent partial melting and are petrologically different from aubrites
139 (Keil and Bischoff, 2008; Moynier et al., 2020; Patzer et al., 2001). The Cr isotope
140 compositions of these different types of enstatite achondrites can be used to test their
141 relationship to each other and to enstatite chondrites. Thus, here we report
142 high-precision mass-independent and mass-dependent Cr isotope data for eight aubrite
143 meteorites, the Itqiy meteorite and one enstatite achondrite clast from Almahatta Sitta.
144

145 **2. Samples and analytical methods**

146 **2.1 Sample description.**

147 The eight aubrites selected for this study are Khor Temiki, Bustee, Norton County,
148 Allan Hills [ALH] A78113, ALH 84007, Larkman Nunatak [LAR] 04316, Shallowater
149 and Cumberland Falls. We also studied two other enstatite-rich achondrite meteorites,
150 Itqiy (EH7-anomalous) and one enstatite-rich clast in Almahatta Sitta, MS-MU-19
151 (Harries and Bischoff, 2020). Itqiy and MS-MU-19 are enstatite-rich rocks, both of
152 which lack plagioclase and high-Ca pyroxene, suggesting they underwent higher
153 degrees of partial melting. Their detailed petrology are described in Patzer et al. (2001)
154 and Harries and Bischoff (2020), respectively. The fractions of Itqiy and MS-MU-19
155 that we used had been previously partially processed and leached by hot (120°C) 6N
156 HCl in order to remove metal and sulfides for trace element studies. While a fraction of
157 the Cr must have been lost during this process, the Cr of the acid residues that we
158 obtained (mostly from the silicates) likely represent the primitive $\epsilon^{54}\text{Cr}$ value, since
159 enstatite is the major Cr-host of these samples and magmatic process would have had
160 homogenized the $\epsilon^{54}\text{Cr}$ signatures at the meteorite scale in any cases. We also dissolved
161 a bulk Itqiy from another chip for mass-dependent Cr isotope study.

162 **2.2 Sample dissolution**

163 Small fragments of eight aubrite meteorites were crushed to powders using an
164 agate mortar. From this powdered material, 30–50 mg were transferred into Teflon

165 bombs for digestion. We also dissolved a small chip (26.4 mg) of LAR 04316 for a
166 replicate. Samples were dissolved following the protocol described in Inglis et al. (2018)
167 using Teflon bombs and an Analab EvapoClean, which has been successfully applied in
168 previous Cr isotopic studies of meteorites (Zhu et al., 2019b; Zhu et al., 2020b). The
169 procedure involved heating in concentrated HF and HNO₃ (2:1) at 140 °C for two days,
170 and subsequent dissolution in 6N HCl (also at 140 °C) for another two days to ensure
171 complete digestion of fluorides, and refractory phases such as chromite and spinel.
172 After complete digestion a ~2% aliquot was used for determination of the ⁵⁵Mn/⁵²Cr
173 ratios and major element contents by ICP-MS in IPGP, ~50% of the digestion were
174 purified for Cr by the following four-step column chemistry described in (Zhu et al.,
175 2021b) for mass-independent Cr isotopic analyses, and the residual 40% were spiked
176 with a ⁵⁰Cr-⁵⁴Cr double spike before a two-step column Cr purification for
177 mass-dependent Cr isotopic analyzes of which method is described in the literature
178 (Sossi et al., 2018; Zhu et al., 2021a; Zhu et al., 2019c).

179 **2.3 Mass-independent Cr isotope ($\epsilon^{53}\text{Cr}$ and $\epsilon^{54}\text{Cr}$) analyses**

180 We first used an anion chromatographic purification scheme to efficiently remove
181 Fe from the remaining sample in 6M HCl, followed by elution of Cr on a 1-mL cation
182 exchange column in 20 mL of 0.5M HNO₃ to remove the major elements including Mg,
183 Ca, Al, Ni (Bizzarro et al., 2011) and collect all the Cr species (major Cr⁰ and minor
184 Cr²⁺ and Cr³⁺) to reach a ~100% recovery. Prior to sample loading on the cation column,
185 we used a Cr pre-treatment procedure involving dissolution in 10M HCl at >120 °C to

186 efficiently promote the formation of Cr(III)-Cl species, which have a low affinity for
187 the cation exchanger and thus elutes early (Larsen et al., 2016; Trinquier et al., 2008a).
188 The third clean-up column involved Cr purification from Al, Fe, V, Ti (and other
189 high-field-strength elements) and Na, K on a small (0.33ml) cation exchange column
190 using 0.5M HNO₃, 1M HF and 6M HCl (Larsen et al., 2018). Prior to sample loading
191 onto this last column, we used a Cr pre-treatment procedure involving exposure to
192 0.5M HNO₃ + 0.6% H₂O₂ at room temperature for >1 day to promote the formation of
193 Cr³⁺ (Larsen et al. 2016). However, it is difficult to transform all the Cr to Cr³⁺, so we
194 collect the Cr⁰ in 0.5 ml solution of sample loading and 0.5 mL 0.5N HNO₃ elution to
195 increase the recovery to >95% in this column. Finally, in the fourth column, 0.7 mL of
196 TODGA resin were used in 8N HCl to remove the residual Fe, V and Ti which are
197 isobars for ⁵⁴Cr (⁵⁴Fe) and ⁵⁰Cr (⁵⁰V and ⁵⁰Ti) (Pedersen et al., 2019). The four columns
198 can reach a total yield between 95% and 99%, and effectively remove Fe, V and Ti,
199 which benefits the isotope analysis on MC-ICPMS. The total blank of <5 ng is
200 negligible compared to the 5-20 µg of Cr processed through the columns. The final Cr
201 solution was re-dissolved in 100µL of concentrated HNO₃ and then dried again 2-3
202 times to transform the acid media and to minimize residual organics (i.e. from the
203 cation exchange resin).

204 The ⁵⁵Mn/⁵²Cr ratios with an uncertainty of 5% were measured on a Neptune plus
205 multiple-collector inductively-coupled-plasma mass-spectrometer (MC-ICP-MS)
206 following the protocol described by (Zhu et al., 2021b) and previously used in (Zhu et

207 al., 2019b; Zhu et al., 2020b). The Cr isotopic compositions were determined using a
208 Neptune Plus MC-ICP-MS located at the Centre for Star and Planet Formation, Globe
209 Institute, University of Copenhagen. Detailed analytical and data reduction method are
210 described in (Zhu et al., 2021b). Sample solutions with concentration of less than 1 ppm
211 were introduced via an ESI APEX and analyzed at a ^{52}Cr signal of 30-40 V, except for
212 Norton County, with low-Cr content (180 ppm), that was analyzed at 15 V. Each sample
213 was measured five times by sample-standard bracketing relative to the NIST SRM 979
214 Cr standard. The $^{53}\text{Cr}/^{52}\text{Cr}$ and $^{54}\text{Cr}/^{52}\text{Cr}$ ratios were normalized to a constant $^{50}\text{Cr}/^{52}\text{Cr}$
215 ratio of 0.051859 using an exponential law (Lugmair and Shukolyukov, 1998) and are
216 expressed in the epsilon notation:

$$217 \quad \varepsilon^x\text{Cr} = \left(\frac{(^x\text{Cr}/^{52}\text{Cr})_{\text{sample}}}{(^x\text{Cr}/^{52}\text{Cr})_{\text{NIST SRM 979}}} - 1 \right) \times 10000 \quad (1),$$

218 with $x = 53$ or 54 .

219 The measurements in this study are performed following Zhu et al. (2021b), and
220 the related elemental doping tests (for Ti, V, Fe, K, and Na), external precision tests
221 and the isotope data of known standard samples for data quality (Allende, DTS-1,
222 PCC-1, DTS-2b) can be also found in Zhu et al. (2021b). The data for the reference
223 standards are consistent with previously reported values (Mougel et al., 2018; Qin et
224 al., 2010; Schiller et al., 2014; Schneider et al., 2020; Trinquier et al., 2007; Trinquier
225 et al., 2008b; Zhu et al., 2019a; Zhu et al., 2019b; Zhu et al., 2020b), confirming the
226 accuracy of our new data.

227 For acid leachates of Itqiy and the enstatite achondrite clast from Almahatta Sitta
228 (MS-MU-19), the purification followed a two-step cation column as originally
229 described in Trinquier et al. (2008a) and modified in Zhu et al. (2019c). Low-yield
230 (e.g., < 70%) Cr purification may produce large equilibrium mass-dependent Cr
231 isotope fractionation from column chemistry which cannot be well corrected using the
232 exponential law (Qin et al., 2010; Trinquier et al., 2008a). Thus, similar to the method
233 used in Moynier et al. (2011b), we used 3 (1 + 1 + 1) mL of 6M HCl for washing the
234 column and we collected the residual material from the first 1-mL cation column and
235 re-passed them in the first column to ensure yields ranging from 88% to 99% (average
236 of 93%). The blank of the full chemical purification is between 0.5 and 2 ng and is
237 negligible compared to 2 – 4 µg of Cr processed through the purification scheme. The
238 mass-independent Cr isotope analysis was obtained via the total evaporation method
239 on the Triton TIMS housed at Freie Universität Berlin based on methods described in
240 the literature (Van Kooten et al., 2016; Zhu et al., 2019b; Zhu et al., 2020a). NIST
241 3112a was used as the isotope standard, because there is no resolvable
242 mass-independent Cr isotope difference between NIST SRM 979 and NIST SRM
243 3112a (Zhu et al., 2021b). Using this method, we also measured the Cr isotope
244 composition from the same dissolution of the Allende meteorite and DTS-1 (terrestrial
245 dunite, USGS terrestrial standard) samples that were used for the MC-ICP-MS
246 analysis, and the data are consistent. The data for individual measurements of TIMS
247 total-evaporation measurements are listed in the Appendix.

248 **2.4 Mass–dependent Cr isotope ($\delta^{53}\text{Cr}$) measurements**

249 The procedure for mass–dependent Cr isotope analysis was performed at the
250 IPGP on aliquots of the same dissolutions used for mass–independent isotope
251 measurements. First, the appropriate amount of ^{50}Cr – ^{54}Cr double spike (28% of the Cr
252 content endemic to the sample) was added to the aliquot, which was then refluxed in a
253 closed beaker at 120 °C overnight to homogenize the sample and spike (e.g., Wu et al.,
254 2020). The Cr of the samples was chemically purified via a typical two–step cation
255 exchange chromatography (Sossi et al., 2018; Zhu et al., 2021a; Zhu et al., 2019c),
256 modified from Trinquier et al. (2007). This method has a total procedural yield of 60–
257 90% and a blank of ~5 ng of Cr. Compared to the total 3–40 μg of Cr from the samples,
258 the blank can be neglected. The final Cr cut was evaporated in concentrated HNO_3
259 drops 3 to 5 times to convert the HCl medium to HNO_3 and to remove leftover organics
260 (i.e., the resin) following which the samples were diluted to a concentration of 1 ppm Cr,
261 in 2% (0.317 M) HNO_3 for isotope analysis.

262 The Cr stable isotope compositions of these purified samples were measured on a
263 Thermo Scientific Neptune Plus MC–ICP–MS housed at the IPGP. Analytical details
264 are described in Zhu et al. (2021a), Sossi et al. (2018) and Zhu et al. (2019c). The
265 purified sample solution was diluted to ~ 1ppm, which returned a signal of ~ 15V of
266 ^{52}Cr with a Stable Introduction System (SIS). Every measurement is composed of 100
267 cycles, with integration time of 4.194 s for each cycle. The final Cr stable isotope data

268 ($\delta^{53}\text{Cr}$) for aubrites were corrected using their respective mass-independent Cr isotope
269 compositions that had previously been analyzed on the same aliquots. The isotopic
270 ratio for samples is reported in delta notation relative to NIST SRM 979:

$$271 \quad \delta^{53}\text{Cr}(\text{‰}) = \left(\frac{(^{53}\text{Cr}/^{52}\text{Cr})_{\text{sample}}}{(^{53}\text{Cr}/^{52}\text{Cr})_{\text{NIST SRM 979}}} - 1 \right) \times 1000 \quad (2)$$

272 The uncertainties quoted are the 2SD of individual sample measurements or the
273 2SD reproducibility of several NIST SRM 979 measurements in the same analytical
274 session ($\sim 0.03 \text{ ‰}$), whichever is the largest. Similarly, the Cr stable isotope
275 compositions of standard samples, including NIST SRM 3112a (unpurified, $\delta^{53}\text{Cr} =$
276 $-0.08 \pm 0.03 \text{ ‰}$; 2SD, N = 5), DTS-1 ($\delta^{53}\text{Cr} = -0.03 \pm 0.03 \text{ ‰}$; 2SD, N = 5), PCC-1
277 ($\delta^{53}\text{Cr} = -0.04 \pm 0.03 \text{ ‰}$; 2SD, N = 6) and BHVO-2 ($\delta^{53}\text{Cr} = -0.15 \pm 0.03 \text{ ‰}$; 2SD, N =
278 2), were also measured in this study to test the data quality. Our data are consistent with
279 previous studies (Bonnand et al., 2016b; Liu et al., 2019; Schoenberg et al., 2016; Sossi
280 et al., 2018; Wu et al., 2020; Zhu et al., 2019c), which suggests that our $\delta^{53}\text{Cr}$ data are
281 accurate to the stated uncertainties.

282

283 3. Results

284 All the sample information, elemental contents, mass-independent ($\epsilon^{53}\text{Cr}$ and
285 $\epsilon^{54}\text{Cr}$) and mass-dependent ($\delta^{53}\text{Cr}$) Cr isotope data together with literature Cr and O
286 isotope data from Shukolyukov and Lugmair (2004), Barrat et al. (2016), Clayton et al.
287 (1984) and Newton et al. (2000) are reported in Table 1. The $\epsilon^{54}\text{Cr}$ variability between
288 the analyzed aubrites is rather limited with six out of the nine aubrites (including two
289 LAP 04316) having a mean $\epsilon^{54}\text{Cr} = 0.04 \pm 0.07$ (2SD, N = 6; Figure 1). The two
290 aubrites with the extreme $\epsilon^{54}\text{Cr}$ values are Shallowater (-0.12 ± 0.04) and Khor Temiki
291 (0.18 ± 0.04). Compared to aubrites, Itqiy and the enstatite achondrite clast in
292 Almahatta Sitta, MS-MU-19, have lower $\epsilon^{54}\text{Cr}$ values, averaging at -0.26 ± 0.03 (2SD,
293 N = 2).

294 The mass-dependent Cr isotope data ($\delta^{53}\text{Cr}$) for aubrites range from 0.10 ± 0.03 ‰
295 (Shallowater) to 0.30 ± 0.03 ‰ (Bustee), and bulk Itqiy possesses $\delta^{53}\text{Cr}$ of $-0.03 \pm$
296 0.03 ‰. The relationship between the $\delta^{53}\text{Cr}$ and $\epsilon^{54}\text{Cr}$ values of these enstatite
297 achondrites are shown in Figure 2a. Among main-group aubrites, $\delta^{53}\text{Cr}$ values broadly
298 increase with bulk Mg# ($\text{Mg}/(\text{Mg}+\text{Fe}) \times 100\%$, atom ratios; > 98 ; Figure 2b), but there
299 is no correlation between their $\delta^{53}\text{Cr}$ values and bulk Cr contents (Figure 2c). The
300 $\delta^{53}\text{Cr}$ values of main-group aubrites are, however, correlated with iron isotope
301 compositions [expressed as $\delta^{56}\text{Fe}$; (Wang et al., 2014)] (Figure 2d). Itqiy, Shallowater
302 and main-group aubrites also have different and correlated Mg# and $\delta^{53}\text{Cr}$ values

303 (Figure 2b and Figure 3).

304 The measured $\epsilon^{53}\text{Cr}$ values range from 0.00 ± 0.02 for Shallowater up to $2.41 \pm$
305 0.03 for Bustee. Aside from these two samples and LAR 04316 (pieces) and
306 Cumberland Falls (small aliquot in this study), the $\epsilon^{53}\text{Cr}$ values of the remaining aubrite
307 samples, including those reported by Shukolyukov and Lugmair (2004), correlate with
308 their respective Mn/Cr ratios and form a ^{53}Mn - ^{53}Cr correlation line with a slope of
309 0.225 ± 0.046 and an intercept of $\epsilon^{53}\text{Cr}$ of 0.50 ± 0.16 (MSWD = 9.8, N = 8; Figure 4a),
310 modeled by *Isoplot R* (Vermeesch, 2018), model 3 (MSWD $\gg 1$). There is also a
311 correlation between $\epsilon^{53}\text{Cr}$ values and $\delta^{53}\text{Cr}$ values (Figure 4b).

312

313 **4. Discussion**

314 **4.1 $\epsilon^{54}\text{Cr}$ systematics and the number of enstatite achondrite parent** 315 **bodies**

316 One of the complications for precise Cr isotope measurements of meteorites is
317 their susceptibility to cosmic ray induced modifications. In aubrites, which are mostly
318 composed of Fe-poor enstatite, this effect is negligible because iron is the primary
319 target for producing Cr isotopes via cosmic ray exposure (Shima and Honda, 1966).
320 Aubrites also have short cosmic ray exposure ages [up to 100 Ma; (Eugster, 2003;
321 Lorenzetti et al., 2003)] compared to other meteorites such as mesosiderites which do
322 not exhibit any cosmogenic effects on Cr isotopes themselves (Eugster, 2003; Trinquier
323 et al., 2007). As such, modification of the Cr isotope systematics of aubrites by such
324 processes is unlikely and not further considered.

325 The combination of $\epsilon^{54}\text{Cr}$ and $\Delta^{17}\text{O}$ signatures has been applied to investigate the
326 kinship of Solar System materials (Trinquier et al., 2007; Warren, 2011; Zhu et al.,
327 2021b). Most aubrites for which these data exist have indistinguishable $\epsilon^{54}\text{Cr}$ and $\Delta^{17}\text{O}$
328 values (Figure 1b), supporting the idea that the majority of aubrites originated from a
329 single parent body. In addition, aubrites also have indistinguishable O and Cr isotopic
330 composition with respect to the Earth, Moon and enstatite chondrites (Figure 1a), and
331 similar $\epsilon^{54}\text{Cr}$ values but different $\Delta^{17}\text{O}$ values compared to R chondrites (Bischoff et al.,
332 2011; Zhu et al., 2021a). Aside from the main aubrite population, some isotopically

333 distinct samples exist: Shallowater, a chondritic clast within Cumberland Falls, and
334 Khor Temiki.

335 Although Shallowater has an indistinguishable $\Delta^{17}\text{O}$ value (-0.02 ± 0.02) from
336 other aubrites (Newton et al., 2000), the $\varepsilon^{54}\text{Cr}$ value reported here is lower than the ones
337 of other aubrites and enstatite chondrites, suggesting a distinct parent body. Formation
338 of Shallowater from a distinct parent body is also consistent with its higher Fe and Cr
339 content (Rubin, 2015; Watters and Prinz, 1979), heavier Zn and Fe isotope
340 compositions (Moynier et al., 2011a; Wang et al., 2014), higher HSE contents (van
341 Acken et al., 2012a) and more complex cooling history (Keil et al., 1989) relative to
342 other aubrites.

343 The distinct and variable $\Delta^{17}\text{O}$ values reported for Cumberland Falls (Clayton et
344 al., 1984; Miura et al., 2007; Newton et al., 2000) have been interpreted as representing
345 the composition of ordinary chondrite-like clasts (Barrat et al., 2016; Verkouteren and
346 Lipschutz, 1983) in this breccia. As such, the $\Delta^{17}\text{O}$ composition of Cumberland Falls
347 cannot be taken as a robust genetic signature. Nonetheless, the sample of Cumberland
348 Falls analyzed here has an indistinguishable $\varepsilon^{54}\text{Cr}$ value from other aubrites (with the
349 exception of Shallowater) and enstatite chondrites. This value differs significantly from
350 that of ordinary chondrites [-0.39 ± 0.09 , 2SD; (Pedersen et al., 2019; Qin et al., 2010;
351 Trinquier et al., 2007)] and, thus, suggests that the sample analyzed here is not
352 compromised by the presence of ordinary chondrite material. Given its
353 indistinguishable $\varepsilon^{54}\text{Cr}$ value with other aubrites, Cumberland Falls appears to belong

354 to the main group-aubrites. However, oxygen isotopes indicates that some limited
355 isotopic heterogeneity existed within this meteorite.

356 The aubrite Khor Temiki, is characterized by a $\epsilon^{54}\text{Cr}$ value of 0.18 ± 0.04 that is
357 slightly higher but overlaps within uncertainties with some other aubrites, including
358 Norton County (0.06 ± 0.09), LAR 04316 (0.06 ± 0.09) and Cumberland Falls ($0.06 \pm$
359 0.09). Despite its slightly elevated $\epsilon^{54}\text{Cr}$ value, Khor Temiki has an indistinguishable O
360 isotope composition ($\Delta^{17}\text{O} = 0.01 \pm 0.02$) from other aubrites (Barrat et al., 2016) and
361 lacks other evidence that could be used to distinguish it from other aubrites. As such,
362 based on $\epsilon^{54}\text{Cr}$ values, Khor Temiki may have originated from a distinct parent body
363 than other aubrites or contain foreign, ^{54}Cr -rich clasts but additional data are required to
364 test these hypotheses.

365 Itqiy ($\epsilon^{54}\text{Cr} = -0.27 \pm 0.10$) and MS-MU-19 ($\epsilon^{54}\text{Cr} = -0.25 \pm 0.08$) have the same
366 $\epsilon^{54}\text{Cr}$ values, consistent with their petrological similarities: they are both enstatite-rich,
367 they lack plagioclase and are depleted in Na, K and V (Harries and Bischoff, 2020;
368 Patzer et al., 2001). Although Itqiy has similar O isotope compositions as ECs and
369 aubrites (Patzer et al., 2002), both Itqiy and MS-MU-19 show clearly different $\epsilon^{54}\text{Cr}$
370 values from enstatite chondrites [0.03 ± 0.13 ; 2SD, N = 12; (Mougel et al., 2018; Qin et
371 al., 2010; Trinquier et al., 2007)]. This observation suggests that they do not directly
372 originate from enstatite chondrites and the aubrite (both main-group ones and
373 Shallowater) parent bodies. The same $\epsilon^{54}\text{Cr}$ values of Itqiy and MS-MU-19, and the
374 similar petrology of Itqiy, NWA 2526, MS-MU-19 and MS-MU-36, supports that there

375 may be at least an additional enstatite-dominated asteroid (probably with a size of 500
376 km; Harries and Bischoff, 2020) in the Solar System, i.e. the Itqiy parent body. Thus,
377 combining petrological arguments with the $\epsilon^{54}\text{Cr}$ systematics allows us to distinguish at
378 least three isotopically distinct reservoirs that generated enstatite-rich achondrites: the
379 main-group aubrite parent body with $\epsilon^{54}\text{Cr} = 0.06 \pm 0.12$ (2SD, N = 7), Shallowater
380 parent body with $\epsilon^{54}\text{Cr} = -0.12 \pm 0.04$ and probably, the Itqiy parent body with $\epsilon^{54}\text{Cr} =$
381 -0.26 ± 0.03 (2SD, N = 2). Note that this is not the same situation as for the $\epsilon^{54}\text{Cr}$ data
382 for monomict ureilites, which shows a broad compositional spectrum (i.e. continuous
383 variation, instead of showing three groups of enstatite achondrites), which is interpreted
384 as planetary mantle heterogeneity in a single parent body (Zhu et al., 2020b).

385 Hence, among the enstatite achondrites, only the $\epsilon^{54}\text{Cr}$ values of main-group
386 aubrites are indistinguishable from the Earth [$(0.09 \pm 0.12; 2\text{SD}, N = 17; (\text{Mougel et al.,}$
387 $2018; \text{Trinquier et al., 2007})$] and the Moon [$(0.09 \pm 0.08; (\text{Mougel et al., 2018})$], and
388 enstatite chondrites [$0.02 \pm 0.11; 2\text{SD}, N = 13; (\text{Mougel et al., 2018; Qin et al., 2010;}$
389 $\text{Trinquier et al., 2007; Zhu et al., 2021b})$] and most enstatite chondrite chondrules:
390 -0.11 ± 0.13 (2SD, N = 7) (Zhu et al., 2020a), which may indicate that they originated
391 from a common precursor reservoir. However, the main-group aubrites cannot directly
392 derive from enstatite chondrites, which will be further discussed in section 4.5. Finally,
393 the $\epsilon^{54}\text{Cr}$ sequence for non-carbonaceous achondrites (including the planets and
394 asteroids) can be updated as: Earth = Moon = main-group Aubrites > Shallowater >
395 Itqiy \approx Mars > Angrites > Winonaites \geq Acapulcoite-Lodranite Clan \geq Vesta >

396 Ureilites (Li et al., 2018; Mougél et al., 2018; Qin et al., 2010; Trinquier et al., 2007;
397 Yamakawa et al., 2010; Zhu et al., 2019b; Zhu et al., 2020b).

398 **4.2 Chromium stable isotope fractionation during sulfide mineral** 399 **crystallization**

400 All aubrites and Itqiy (EH7-an; bulk) are enriched in the heavier isotopes of Cr
401 with respect to chondrites and other differentiated parent bodies (Fig. 3). These
402 distinct stable isotopic compositions must be the result of parent body processes since
403 chondrites from different groups have homogeneous $\delta^{53}\text{Cr}$ values (Bonnand et al.,
404 2016b; Schoenberg et al., 2016; Zhu et al., 2021a), which excludes Cr stable isotope
405 fractionation by nebular processes. Chromium isotopes are fractionated during
406 planetary processes, such as partial melting and fractional crystallization as observed
407 for the Earth (Bonnand et al., 2020b; Schoenberg et al., 2008; Shen et al., 2019), the
408 Moon (Bonnand et al., 2016a; Sossi et al., 2018) and Vesta (Zhu et al., 2019c).
409 Moreover, equilibrium isotope fractionation by evaporation processes between
410 oxidized vapor and reduced residue has been documented for lunar and
411 Howardite-Eucrite-Diogenite (HED) samples (Sossi et al., 2018; Zhu et al., 2019c).

412 Aubrites are dominated by silicate minerals [$\approx 99\%$, except for Shallowater;
413 (Watters and Prinz, 1979)], predominantly enstatite and a minor plagioclase, diopside
414 and forsterite. However, these silicate minerals are usually poor in Cr, mostly $< \sim 300$
415 ppm (Watters and Prinz, 1979). Chromium abundances in metals, i.e., kamacite and
416 schreibersite, are even lower, ~ 20 ppm (Casanova et al., 1993). On the contrary, sulfide

417 minerals are usually rich in Cr, e.g., troilite, daubreelite, alabandite, caswellsilverite
418 and heideite can contain between ~0.5 (troilite) to ~37.4 (daubreelite) wt% Cr. The
419 much higher Cr content of sulfides relative to that of silicates and high silicate
420 abundance in main-group aubrites indicate that the bulk contents of Cr are controlled
421 primarily by silicate and sulfide minerals, since metal does not contribute much to the
422 Cr budget (Casanova et al., 1993). Therefore, the variations of the Cr contents and of
423 the $\delta^{53}\text{Cr}$ values possibly result from a two-endmember mixing between isotopically
424 heavy and Cr-poor silicates and isotopically light and Cr-rich sulfides. This is
425 consistent with *ab initio* calculations that predict that CrS and daubr elite (FeCr_2S_4) are
426 isotopically light compared to silicates (Moynier et al., 2011b). However, there is no
427 correlation between $[\delta^{53}\text{Cr}]$ and $[1/\text{Cr}]$ (figure 2c) as would be expected for simple
428 mixing between sulfides (high Cr content and low $\delta^{53}\text{Cr}$ values) and silicates (low Cr
429 content and high $\delta^{53}\text{Cr}$ values). This could be due to secondary processes (i.e. an impact
430 event) that result in the equilibration of the Cr isotopes at the meteorite scale, while the
431 heterogeneous distribution of Cr-rich sulfide would significantly affect the Cr contents
432 of the different chips of a same meteorite. For example, Cumberland Falls shows
433 variable Cr contents, 945 ppm in this study and 549 ppm in Shukolyukov and Lugmair
434 (2004). Thus, it is difficult to determine the representative Cr contents (also the Mn/Cr
435 ratios) in bulk aubrite samples (i.e., different chips from one aubrite can have similar Cr
436 stable isotope compositions but distinct Cr contents), if the sample size is not large

437 enough, e.g., < 600 mg (Stracke et al., 2012). This sample heterogeneity effect is also
438 discussed in section 4.4.

439 This two-endmember (silicate-sulfide) mixing for aubrites is reminiscent of the
440 positive correlation between Fe content and Fe isotopes in aubrites (Wang et al., 2014).
441 This has been interpreted as the mixing of isotopically light Fe from sulfide-rich metal
442 with isotopically heavy, Fe-poor silicates (mantle material). In detail, Wang et al. (2014)
443 argued the silicate in aubrites represent the mantle of aubrite parent body, while the
444 metal is likely some residue in the mantle from incomplete metal-silicate differentiation
445 (Casanova et al., 1993). A common origin for the $\delta^{53}\text{Cr}$ and $\delta^{56}\text{Fe}$ variability of aubrites,
446 ignoring the anomalous Shallowater, is supported by their correlated variability (Figure
447 2d). However, because of its low Cr abundances, the Cr-poor Fe-Ni metal in aubrites
448 (Casanova et al., 1993) cannot account for the Cr isotope variations. Instead, it is more
449 likely that the variable presence of sulfide minerals (e.g., troilite) that are both rich in Fe
450 and Cr (Keil, 2010a; Watters and Prinz, 1979) caused the observed correlated effects
451 between Cr and Fe isotopes. This interpretation is also supported by experimental data
452 that show equilibrium Fe isotope fractionation increases with sulfur content to $\sim 0.4\text{‰}$
453 at 18 wt.% sulfur in metal coexisting with silicate melt (Shahar et al., 2015).

454 **4.3 Isotopically heavy Cr in the mantle of aubrite parent bodies** 455 **induced by core formation**

456 In order to use Cr stable isotopes to understand the formation and differentiation
457 of the aubrite parent bodies (main-group and Shallowater), it is necessary to estimate

458 the $\delta^{53}\text{Cr}$ composition of their mantles. The depletion of HSE (van Acken et al., 2012a;
459 van Acken et al., 2012b) in aubrites compared to chondrites is evidence that their parent
460 body underwent core formation. However, given the high Cr content of sulfides
461 compared to silicates and their heterogeneous distribution within aubrites, obtaining a
462 pure silicate mantle composition requires a sulfide-free sample. Also, troilite, as the
463 major sulfide minerals in aubrites, contains Fe (~60%; (Watters and Prinz, 1979),
464 which given their chemical composition are usually interpreted as reflecting leftover
465 core material after incomplete core-mantle differentiation of the aubrite parent body
466 (Casanova et al., 1993). As explained above, Norton County has the highest Mg# (99.9)
467 and a bulk Cr content (180 ppm) comparable to the Cr content of typical silicate
468 minerals [$<0.04\%$, (Keil, 2010a; Watters and Prinz, 1979)]. As such, the Cr isotope
469 composition of Norton County, $\delta^{53}\text{Cr} = 0.24 \pm 0.03 \text{‰}$, is taken as the best estimation of
470 the mantle Cr isotope composition. Considering the silicate-sulfide mixing model for
471 Cr stable isotope variation, Bustee with heavier $\delta^{53}\text{Cr}$ value ($0.30 \pm 0.03 \text{‰}$) may also
472 indicate the silicate compositions and bulk silicate main-group aubrite parent body. The
473 elevated Cr contents and lower Mg# for Bustee can be also caused by sample
474 heterogeneity. However, the $\delta^{53}\text{Cr}$ values for both Norton County and Bustee are
475 resolvably heavier than those of chondrites, Shallowater and Itqiy. Given that
476 Shallowater and Itqiy are unique samples, their bulk $\delta^{53}\text{Cr}$ values of $0.10 \pm 0.03 \text{‰}$ and
477 $-0.03 \pm 0.03 \text{‰}$ are our best approximation for their parent bodies' mantle. The mantle
478 $\delta^{53}\text{Cr}$ difference between the three parent bodies (Figure 3), consistent with their $\epsilon^{54}\text{Cr}$

479 signatures (Figure 1b and 2a), confirms the existence of at least three enstatite
480 achondrite parent bodies in the Solar System. This is also consistent with the difference
481 in the mantle Mg# between three parent bodies (Figure 2b).

482 Chondrites have homogeneous Cr stable isotope compositions, with $\delta^{53}\text{Cr} = -0.12$
483 $\pm 0.04 \text{‰}$ (2SD, N = 42) (Bonnand et al., 2016b; Schoenberg et al., 2016; Zhu et al.,
484 2021a). The $\delta^{53}\text{Cr}$ values of all aubrites, including main-group, Shallowater and Itqiy
485 are heavier than bulk chondrites as well as the mantles of other planetary bodies: Earth
486 [$-0.12 \pm 0.04 \text{‰}$; (Jerram et al., 2020; Schoenberg et al., 2008; Sossi et al., 2018)],
487 Moon [$-0.21 \pm 0.03 \text{‰}$; (Bonnand et al., 2016a; Sossi et al., 2018)] and Vesta [$-0.22 \pm$
488 0.03‰ ; (Zhu et al., 2019c)] (Figure 3). These isotopic offsets: $\Delta_{\text{main-group aubrites-Chondrites}}$
489 $= 0.36 \pm 0.05 \text{‰}$, $\Delta_{\text{Shallowater-Chondrites}} = 0.22 \pm 0.05 \text{‰}$ and $\Delta_{\text{Itqiy-Chondrites}} = 0.09 \pm 0.05 \text{‰}$,
490 combined with significant depletion in Cr between aubrites and chondrites (including
491 enstatite chondrites) may provide significant insights into the origin of the bulk silicate
492 aubrite parent bodies.

493 Several mechanisms may have caused the Cr stable isotope difference between
494 chondrites and the bulk silicate aubrite parent bodies: 1) igneous processes, such as
495 partial melting (e.g., Zhu et al., 2019c), 2) volatilization processes (e.g., Sossi et al.,
496 2018; Zhu et al., 2019c) or 3) core formation (e.g., Bonnand et al., 2016b; Moynier et
497 al., 2011b). Partial melting is a possible way to fractionate Cr stable isotopes, because
498 of the potential exchange between “heavy” Cr^{3+} and “light” Cr^{2+} in residue and melts
499 (e.g., Shen et al., 2019; Zhu et al., 2019c). This is also consistent with the fact that the

500 aubrites are depleted in basaltic components (Wilson and Keil, 1991). However, given
501 the highly reduced conditions during aubrite differentiation (IW-2—IW-6), both the
502 melt and residue are dominated by Cr²⁺ (Bell et al., 2020; Berry et al., 2006), and
503 therefore partial melting is unlikely to fractionate Cr stable isotopes. Moreover, for
504 the Earth's mantle, which is more oxidized than aubrites and contains both Cr²⁺ and
505 Cr³⁺, the extent of Cr isotopic fractionation is less than ~0.15 ‰ (based on the $\delta^{53}\text{Cr}$
506 difference between terrestrial chromite-free peridotites and chromites (Shen et al.,
507 2015). This difference is much lower than what would be required to explain the
508 difference between chondrites and main-group aubrite parent body (0.36 ± 0.05 ‰).
509 Hence, partial melting cannot result in the observed Cr stable isotopic variations of
510 the aubrites.

511 The volatility of Cr decreases together with $f\text{O}_2$, e.g., at $f\text{O}_2$ below IW = -2 and
512 atmospheric pressure Cr becomes more refractory than Mg (Sossi et al., 2019). Thus,
513 under reduced conditions volatile loss of Cr from aubrites should have resulted in
514 concomitant losses of Mg which is not observed (Sedaghatpour and Teng, 2016).
515 However, it is unknown whether the sulfur content affect the volatility of Cr, so we
516 cannot totally exclude that the isotopically heavy Cr in aubrites is the result of
517 evaporation (e.g., if sulfur increase the Cr volatility), considering that aubrite sulfides
518 are rich in Cr (Watters and Prinz, 1979). Given the distinct Cr content and
519 metal-sulfide abundance between bulk silicate aubrite parent body and ECs (Keil,
520 1968), and that Cr is not likely to be lost by volatilization, stable isotope fractionation

521 associated with core formation may be the most plausible mechanism to account for
522 the heavier Cr in aubrites compared to chondrites.

523 The size of the aubrite parent body is not known, but is hypothesized to be
524 probably <100 km based on models of the solubility of volatiles in basaltic melts of
525 the aubrite parent body (Wilson and Keil, 1991). Chromium is mostly chalcophile, not
526 siderophile, under the prevalent very low fO_2 of enstatite meteorites and low pressure
527 conditions (Wood et al., 2008) and, thus, there should also be little Cr dissolved in the
528 metallic core of the aubrite parent bodies. However, the partition coefficient of Cr into
529 metal increases with the increasing sulfur content (Wood et al., 2014), which is also
530 consistent with the high Cr content in the sulfide minerals in aubrites (Keil, 2010a;
531 Watters and Prinz, 1979). Previous experiments have predicted that there should be
532 very limited Cr stable isotope fractionation during metal-silicate segregation
533 (Bonnand et al., 2016b). However, the experiments did not consider the composition
534 effect of S, so it does not necessarily apply to aubrites, because the extremely reduced
535 conditions during differentiation of aubrite parent body would likely form a core with
536 abundant S. At present, it is difficult to provide a quantitative model to test the
537 hypothesis that Cr isotope fractionation occurred during core formation. The core of
538 the aubrite parent body may have contained both Fe-Ni-Si metal and sulfide.
539 Unfortunately, even if the Cr contents in metal and sulfide were known, the
540 proportions of metal and sulfide in the core cannot be estimated reliably, and thereby,
541 the Cr contents in the core remain unknown.

542 The three enstatite achondrite parent bodies possess higher $\delta^{53}\text{Cr}$ values relative
543 to other planets/asteroids, including Earth, Moon and Vesta (Figure 3). Earth possesses
544 similar $\delta^{53}\text{Cr}$ values as chondrites (Bonnand et al., 2016b; Jerram et al., 2020;
545 Schoenberg et al., 2016; Schoenberg et al., 2008; Sossi et al., 2018), suggesting that
546 core formation did not fractionate Cr stable isotopes, which is consistent with the high
547 temperature of Earth's core formation and further confirmed by high-pressure and
548 high-temperature experiments (Bonnand et al., 2016b). Due to the relatively low
549 temperature of the lunar core-mantle boundary (Cr is only siderophile at high
550 temperature; (Corgne et al., 2008), only ~1% of the Moon's Cr would enter its core
551 (Sossi et al., 2018). Therefore, the isotopically light Cr in lunar rocks is not due to core
552 formation, but rather to a volatile loss during the magma ocean stage (Sossi et al., 2018).
553 Also, Vesta, with a Cr-poor core, has lower $\delta^{53}\text{Cr}$ values than chondrites and enstatite
554 achondrites, and its isotopically light Cr is interpreted to be caused by low-temperature
555 volatile process rather than core formation (Zhu et al., 2019c). The Cr stable isotope
556 composition of the Martian mantle is not well constrained, and in the future the physical
557 conditions for the formation of the Martian core can be potentially traced with Cr
558 isotopes.

559 **4.4 Timescale of evolution of the main-group aubrite parent body**

560 Aubrites are the product of magmatic differentiation on their parent body, which
561 can potentially be dated using the ^{53}Mn - ^{53}Cr chronometer (e.g., Shukolyukov and
562 Lugmair, 2004). The $^{55}\text{Mn}/^{52}\text{Cr}$ ratio of Cumberland Falls reported by Shukolyukov

563 and Lugmair (2004), corresponding to 1.99, is different from the one determined here
564 (8.40). However, the two portions of Cumberland Falls have the same $\epsilon^{53}\text{Cr}$ values. A
565 similar observation has been made for leachates with variable $^{55}\text{Mn}/^{52}\text{Cr}$ ratios (0.04 –
566 9.58) for Bishopville (Shukolyukov and Lugmair, 2004) and two portions of LAR
567 04316 in this study, with $^{55}\text{Mn}/^{52}\text{Cr}$ ratios of 4.55 and 7.46, that all return similar $\epsilon^{53}\text{Cr}$
568 values, suggesting that internal Mn/Cr redistribution post-dates ^{53}Mn decay and lack of
569 internal ^{53}Mn - ^{53}Cr isochrons in aubrites. This situation does not mimic the leachates or
570 minerals (with variable $^{55}\text{Mn}/^{52}\text{Cr}$ ratios) in some individual angrites (Glavin et al.,
571 2004; Lugmair and Shukolyukov, 1998), eucrites (Lugmair and Shukolyukov, 1998)
572 and ureilites (Yamakawa et al., 2010), which show variable $\epsilon^{53}\text{Cr}$ values. Considering
573 the half-life of ^{53}Mn [(3.7 Myrs; (Holden, 1990)] combined with the Mn/Cr variability
574 in LAR 04316, Cumberland Falls and Bishopville and the precision of the $\epsilon^{53}\text{Cr}$ data,
575 the redistribution of Mn in these aubrites likely occurred after the extinction of ^{53}Mn ,
576 i.e. >20 Myrs (eight half-life of ^{53}Mn ; Figure 5) after Solar System formation.
577 Metamorphic redistribution of Mn and Cr on the meteorite scale might have occurred
578 by impact processes and is also supported by the fractionated trace elements and by
579 Ar-Ar ages (Biswas et al., 1980; Bogard et al., 2010). On the other hand, the
580 heterogeneous distribution of Mn/Cr ratios in aubrites, mainly caused by Mn- or
581 Cr-rich sulfide minerals, suggest that it will be difficult to determine the true $^{55}\text{Mn}/^{52}\text{Cr}$
582 ratios of bulk aubrites. In other words, the $\epsilon^{53}\text{Cr}$ of each meteorite sample could be
583 representative of the bulk meteorite, while the $^{55}\text{Mn}/^{52}\text{Cr}$ ratio may not be, since

584 different sample fractions with variable Mn/Cr ratios have the same $\epsilon^{53}\text{Cr}$ values.

585 Aubrites do not all fall on the same line in a $\epsilon^{53}\text{Cr}$ against Mn/Cr ratio diagram
586 (Figure 4a), including the three data points from Shukolyukov and Lugmair (2004).
587 To limit nugget effects of Cr-rich sulfides in aubrites (that would result in a variation
588 of Mn/Cr ratios of bulk aubrites and would prohibit obtaining the “true” Mn/Cr ratios
589 of bulk aubrites), we focus on the largest samples that should be more representative
590 of the bulk sample composition. For this reason, we only consider the largest LAR
591 04316 sample (Table 1). Similarly, for Cumberland Falls, we use the data reported by
592 Shukolyukov and Lugmair (2004) (Cumberland Falls-SL; Table 1) that is obtained
593 from a large sample (905 mg) compared to ours (42 mg). Excluding also Bustee,
594 which has a very different Mn/Cr ratio in our study (1.42) compared to the literature
595 (6.08, Watters and Prinz, 1970) and Shallowater that is likely originating from a
596 different parent body, we obtain a correlation line (Figure 4a) for the main-group
597 aubrites. Using a model 3 (due to $\text{MSWD} \gg 1$) regression of *Isoplot R*, this
598 ^{53}Mn - ^{53}Cr correlation line for the main-group aubrites has an intercept (i.e., initial
599 $\epsilon^{53}\text{Cr}$ value) of 0.50 ± 0.16 and a slope of 0.225 ± 0.046 ($\text{MSWD} = 9.8$, $N = 8$). This
600 slope corresponding to a $^{53}\text{Mn}/^{55}\text{Mn}$ ratio of $(2.55 \pm 0.52) \times 10^{-6}$ can be translated to an
601 absolute date of 4562.5 ± 1.1 Ma, anchored to the U isotope corrected U-Pb age of
602 D’Orbigny angrite (Amelin, 2008; Brennecka and Wadhwa, 2012; Glavin et al., 2004).
603 This date is similar to that reported in Shukolyukov and Lugmair (2004), but in view of
604 our new stable isotope data, we interpret it was controlled by mixing of silicate and

605 sulfide minerals, and the time significance needs to be discussed.

606 Most aubrites are breccias (Keil, 2010a) and, as such, many are mixtures of rocks
607 with a different history. Hence, these rocks do not have simple igneous history,
608 which limits the use of the ^{53}Mn - ^{53}Cr system to obtain reliable age information such
609 as previously done for eucrites and diogenites (Trinquier et al., 2008b) and angrites
610 (Zhu et al., 2019b). This mixing history is also supported by the stable Cr isotope data
611 (section 3.2 and 3.3), suggesting that the $\delta^{53}\text{Cr}$ variation in aubrites is mainly
612 controlled by the relative abundance of sulfides and silicates. Finally, the $\delta^{53}\text{Cr}$ values
613 of aubrites positively correlate with their $\epsilon^{53}\text{Cr}$ values (Figure 4b). Their $\epsilon^{53}\text{Cr}$ values
614 mainly reflect the true $^{55}\text{Mn}/^{52}\text{Cr}$ ratios, because the Mn/Cr heterogeneity and
615 secondary processes for aubrites make it difficult to determine the representative
616 Mn/Cr ratios for bulk aubrite samples. This correlation indicates that the Cr in
617 aubrites are mixtures of Cr-rich sulfide (low Mn/Cr, low $\epsilon^{53}\text{Cr}$ and low $\delta^{53}\text{Cr}$) and
618 Cr-poor silicates (high Mn/Cr, high $\epsilon^{53}\text{Cr}$ and high $\delta^{53}\text{Cr}$).

619 Since aubrites are igneous rocks that experienced magmatic processes, both
620 silicate and sulfide should crystallize from the melt and derive from a common initial
621 reservoir. Considering this, the conditions for silicate and sulfide to define an isochron
622 are: 1) to be formed at the same time (crystallization from the same magma), 2) to
623 originate from the same reservoir (the melt) and 3) that possible later impact
624 metamorphism would not change the Mn/Cr ratios and Cr isotope compositions of the
625 bulk aubrites (closed system; but this could reset and flatten the internal isochron for

626 individual aubrites). In this respect, an external ^{53}Mn - ^{53}Cr isochron age for
627 main-group aubrites of 4562.5 ± 1.1 Ma would be valid and with time significance.

628 As suggested before for Mn-Cr whole rock isochrons of other planetary bodies
629 (Lugmair and Shukolyukov, 1998; Trinquier et al., 2008b; Zhu et al., 2019b; Zhu et al.,
630 2020b), the age from the bulk aubrite isochron likely records the timing of the
631 differentiation of the main-group aubrite parent body, similar to those of Vesta, the
632 angrite and ureilite parent bodies (Trinquier et al., 2008b; Zhu et al., 2019b; Zhu et al.,
633 2020b). Mineral crystallization would also fractionate Mn/Cr ratios between aubrites.
634 Enstatite in aubrites is typically characterized by low Mn and Cr contents (< 400 ppm
635 for Mn and < 200 ppm for Cr) (Watters and Prinz, 1979). As such, the Mn/Cr
636 fractionation between different main-group aubrites reflects variable incorporation of
637 Mn-rich and Cr-rich minerals (e.g. oldhamite, daubreelite, caswellsilverite, heideite
638 and niningerite) under reduced conditions during their crystallization. This age is very
639 similar to the rock-forming age of Shallowater 4562.4 ± 0.2 Ma (Pravdivtseva et al.,
640 2017), suggesting that the magmatic process for the enstatite achondrite parent bodies
641 occurred at the same time. On the other hand, the differentiation time of the enstatite
642 achondrite parent bodies is delayed compared to the ureilite parent body with
643 ^{53}Mn - ^{53}Cr age of 4566.7 ± 1.5 Ma (Zhu et al., 2020b), Vesta with ^{53}Mn - ^{53}Cr age of
644 4564.8 ± 0.6 Ma (Trinquier et al., 2008b) and the angrite parent body with ^{53}Mn - ^{53}Cr
645 age of 4563.2 ± 0.3 Ma (Zhu et al., 2019b) when anchored to D'Orbigny.

646 **4.5 The relationship between enstatite chondrites and aubrites and**
647 **core formation based on the initial $\epsilon^{53}\text{Cr}$ values**

648 The initial $\epsilon^{53}\text{Cr}$ value of the aubrite precursors defined by the intercept of the
649 Mn-Cr isochron with the ordinate axis provides additional insight into the origin of
650 aubrites and their potential genetic relationship to enstatite chondrites. The initial $\epsilon^{53}\text{Cr}$
651 value for the main-group aubrites (0.50 ± 0.16 , 2σ) is significantly higher than the
652 average $\epsilon^{53}\text{Cr}$ value of bulk enstatite (0.16 ± 0.06 , 2SD , $N = 12$), Rumuruti (0.23 ± 0.05 ,
653 2SD , $N = 12$), carbonaceous ($-0.04 \sim 0.27$) and ordinary (0.19 ± 0.10 , 2SD , $N = 23$)
654 chondrites (Mougel et al., 2018; Qin et al., 2010; Trinquier et al., 2008b; Zhu et al.,
655 2021a; Zhu et al., 2021b). This high initial $\epsilon^{53}\text{Cr}$ value is also consistent with the
656 intercept of the three-point isochron from Shukolyukov and Lugmair (2004), but its
657 implications have never been fully discussed. Moreover, whole-rock Mn-Cr isochrons
658 for Vesta, angrite and ureilite parent bodies, which record Mn-Cr ages of ~ 2.5 Ma, ~ 4.1
659 Ma and ~ 0.5 Ma after CAIs, respectively, record lower initial $\epsilon^{53}\text{Cr}$ values than their
660 parental melts of -0.12 ± 0.05 (Trinquier et al., 2008b), -0.10 ± 0.06 (Zhu et al., 2019b)
661 and -0.24 ± 0.10 (Zhu et al., 2020b). It should also be noted that the initial $\epsilon^{53}\text{Cr}$ value of
662 main-group aubrites is much higher than that of EC chondrules, -0.13 ± 0.07 (Zhu et al.,
663 2020a), which are potential building blocks of enstatite achondrite parent bodies (e.g.,
664 Johansen et al., 2015). The higher initial $\epsilon^{53}\text{Cr}$ value as recorded by the aubrite whole
665 rock isochron implies that the parent magma of the aubrites had a supra-chondritic
666 Mn/Cr ratio. Shallowater is characterized by a low $\epsilon^{53}\text{Cr}$ value (0.00 ± 0.02) compared

667 to other aubrites, indicating that it evolved from another source with a resolvably lower
668 initial $\epsilon^{53}\text{Cr}$ value than those of the main-group aubrites. Combined with the $\epsilon^{54}\text{Cr}$
669 systematics, the Cr isotope evidence strongly suggests that the main-group and
670 Shallowater aubrites should have distinct genetic origins.

671 The elevated initial $\epsilon^{53}\text{Cr}$ value of main-group aubrites compared to that of other
672 asteroids (e.g., Vesta, angrite and ureilite parent bodies) in the Solar System, indicates
673 that their parent melt had a high, non-chondritic, Mn/Cr ratio. A supra-chondritic
674 Mn/Cr ratio of the parent magma of main-group aubrite parent body can occur either if
675 the aubrite parent body precursor was characterized by a Mn/Cr ratio higher than
676 chondrites or if the Mn/Cr ratio of primitive melt for aubrites changed during the
677 early evolution of the parent body. Note that the partition coefficient between FeS and
678 silicate melt, i.e. $D^{\text{FeS/silicate melt}}$, of Cr is higher than that of Mn (Berthet et al., 2009), so
679 the S-rich core formation process can efficiently increase the Mn/Cr ratio and $\epsilon^{53}\text{Cr}$
680 value of the silicate melt of main-group aubrite parent body. This scenario is supported
681 by the S-rich core formation triggering large Cr stable isotope fractionation of aubrites
682 (see section 4.3). This is also consistent with that other S-poor and early formed rocky
683 planets, i.e. Vesta, angrite and ureilite parent body, have sub-chondritic initial $\epsilon^{53}\text{Cr}$
684 values (Trinquier et al., 2008b; Zhu et al., 2019b; Zhu et al., 2020b).

685 Given the otherwise close kinship between aubrites and enstatite chondrites
686 expressed by their similarly reduced conditions, mineralogy and isotope compositions
687 (including $\Delta^{17}\text{O}$, $\epsilon^{54}\text{Cr}$ and $\epsilon^{50}\text{Ti}$ values), it is more likely that the initial Mn/Cr ratio of

688 the aubrite parent body was close to chondritic. Therefore, aubrites may be the product
689 of post-core formation melting on an enstatite chondrite-like parent body that accreted
690 (Zhu et al., 2020a) and melted very early (e.g., < 2 Myr after Solar System formation) to
691 generate ^{53}Cr ingrowth prior to aubrite formation (Figure 5). Such early melting is
692 typically associated with large scale differentiation on other asteroids such as Vesta
693 (Schiller et al., 2011; Trinquier et al., 2008b) or the angrite parent body (Schiller et al.,
694 2015; Zhu et al., 2019b) due to the abundance of ^{26}Al at this time.
695

696 5. Conclusions

697 This work reported both the mass-independent and mass-dependent Cr isotope
698 data for bulk enstatite achondrites, providing insights into the formation of their
699 parent bodies in three ways, nucleosynthetic ($\epsilon^{54}\text{Cr}$; origin), stable ($\delta^{53}\text{Cr}$; process)
700 and radiogenic ($\epsilon^{53}\text{Cr}$; timing) isotope fractionations:

701 1. The $\epsilon^{54}\text{Cr}$ systematics define three groups of enstatite achondrites that likely
702 represent three parent bodies, the main-group aubrite parent body with $\epsilon^{54}\text{Cr}$ of $0.06 \pm$
703 0.12 (2SD), Shallowater parent body with $\epsilon^{54}\text{Cr} = -0.12 \pm 0.04$ and Itqiy parent body
704 with $\epsilon^{54}\text{Cr} = -0.26 \pm 0.03$ (2SD, N =2). This is consistent with their different $\delta^{53}\text{Cr}$
705 compositions: 0.24 ± 0.03 ‰, 0.10 ± 0.03 ‰ and -0.03 ± 0.03 ‰, respectively.

706 2. Mixing of different proportions of sulfides and silicates possibly result in the
707 Cr stable isotope variation in aubrites. The globally heavy Cr in aubrites relative to
708 that in chondrites could reflect large-scale Cr stable isotope equilibrium fractionation
709 between sulfide (light) and silicate (heavy) followed by segregation of sulfide during
710 core formation, resulting in isotopically heavy mantle.

711 3. The aubrite samples record heterogeneous distribution of Mn and Cr. The
712 ^{53}Mn - ^{53}Cr correlation for the main-group aubrites (except Bustee) is a mixing line but
713 also an isochron that holds a time significance of the crystallization of silicate and
714 sulfide minerals and return an absolute age of 4562.5 ± 1.1 Ma (i.e. 4.8 ± 1.1 Ma after
715 CAIs). This most likely represents the age of the silicate differentiation on the

716 main-group aubrite body. The absence of internal Mn-Cr isochrons for individual
717 aubrites implies that they underwent metamorphic redistribution of Mn and Cr after
718 ^{53}Mn extinction, maybe following a disruption event of the parent body.

719 4. The initial $\epsilon^{53}\text{Cr}$ value for main-group aubrites are higher than the $\epsilon^{53}\text{Cr}$ values
720 for enstatite chondrites, suggesting the main-group aubrites did not directly originate
721 from enstatite chondrites. Instead, the high initial $\epsilon^{53}\text{Cr}$ values of main-group aubrites
722 are potentially caused by an early sulfur-rich core formation that efficiently increased
723 the Mn/Cr ratio of the silicate fraction.

724

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747

748 **Appendix**

749 **Individual data for TIMS total-evaporation measurements**

Sample	Itqiy		MS-MU-19		Allende		DTS-1	
N	$\epsilon^{53}\text{Cr}$	$\epsilon^{54}\text{Cr}$	$\epsilon^{53}\text{Cr}$	$\epsilon^{54}\text{Cr}$	$\epsilon^{53}\text{Cr}$	$\epsilon^{54}\text{Cr}$	$\epsilon^{53}\text{Cr}$	$\epsilon^{54}\text{Cr}$
1	0.50	-0.33	1.87	-0.15	0.19	1.01	0.19	0.31
2	0.56	0.06	1.78	-0.26	0.11	0.75	0.11	0.30
3	0.60	-0.23	1.78	-0.10	0.11	0.90	0.05	0.18
4	0.48	-0.55	1.71	-0.50	-0.04	0.67	0.05	0.41
5	0.44	-0.59	1.87	-0.29	0.12	1.01	-0.04	0.05
6	0.55	-0.22	1.65	-0.45	0.08	0.82	-0.01	0.08
7	0.43	-0.44	1.76	-0.33	0.05	0.84	-0.01	0.10
8	0.65	-0.20	1.72	-0.11	0.02	0.72	0.05	0.22
9	0.55	-0.16	1.77	-0.29	0.03	0.85	-0.10	-0.11
10	0.59	-0.17	1.66	-0.15	0.01	0.55	0.11	0.28
11	0.52	-0.07	1.80	-0.15			-0.04	0.01
12	0.46	-0.32					0.00	0.01
13	0.50	-0.21						
14								
Average	0.52	-0.27	1.76	-0.25	0.07	0.81	0.03	0.15
2SD	0.13	0.37	0.14	0.27	0.13	0.29	0.16	0.31
2SE	0.04	0.10	0.04	0.08	0.04	0.09	0.05	0.09

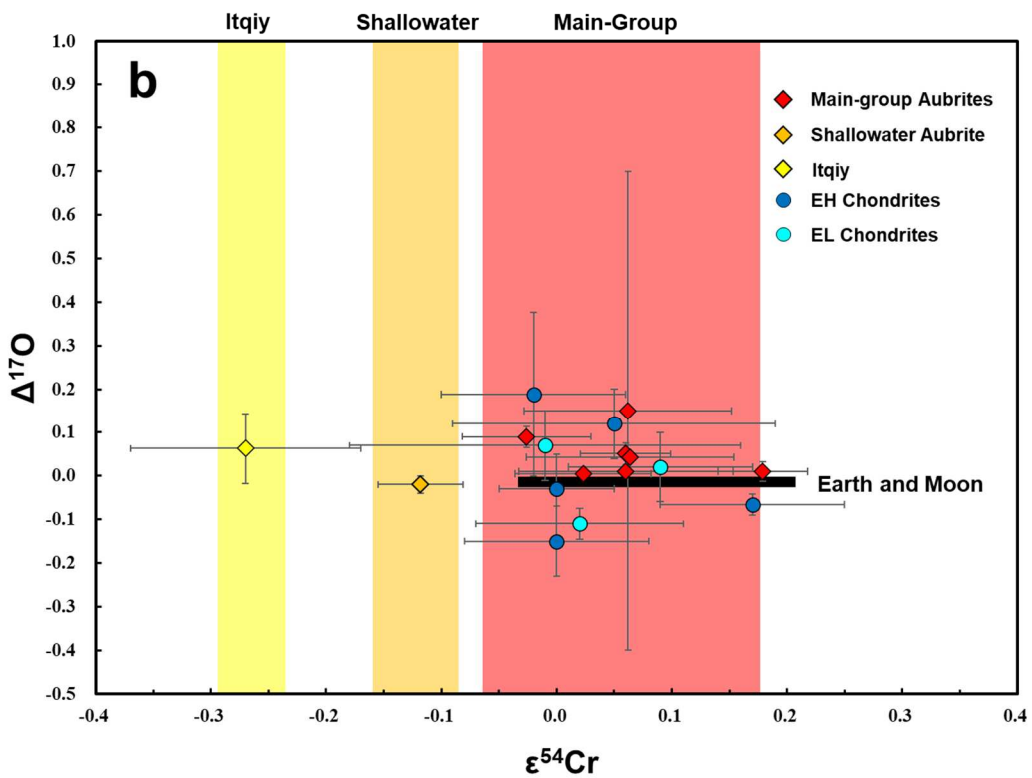
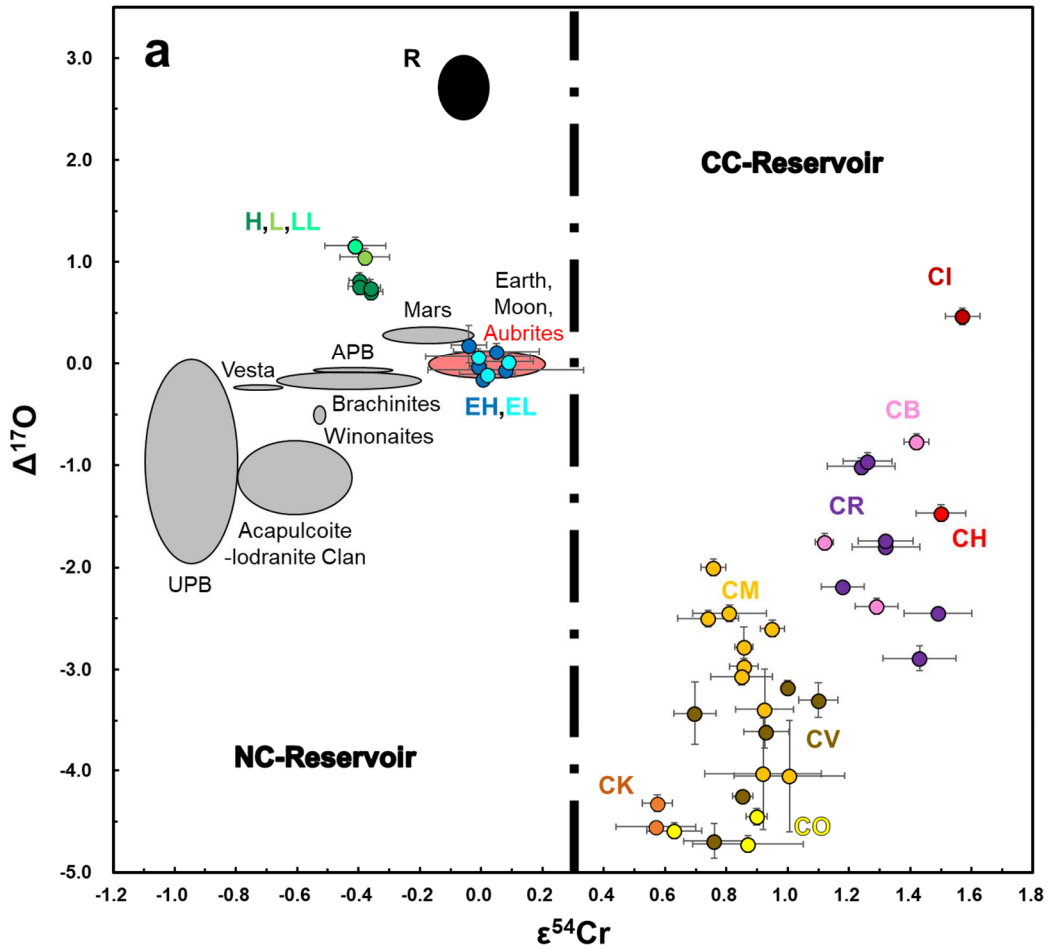
750

Table 1 Cr (both mass-independent and mass-dependent) and O isotopes data for aubrites.

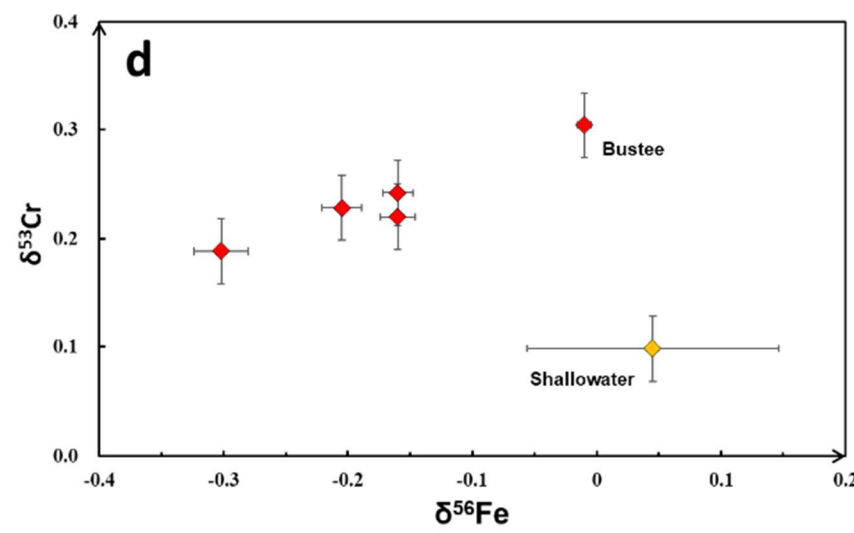
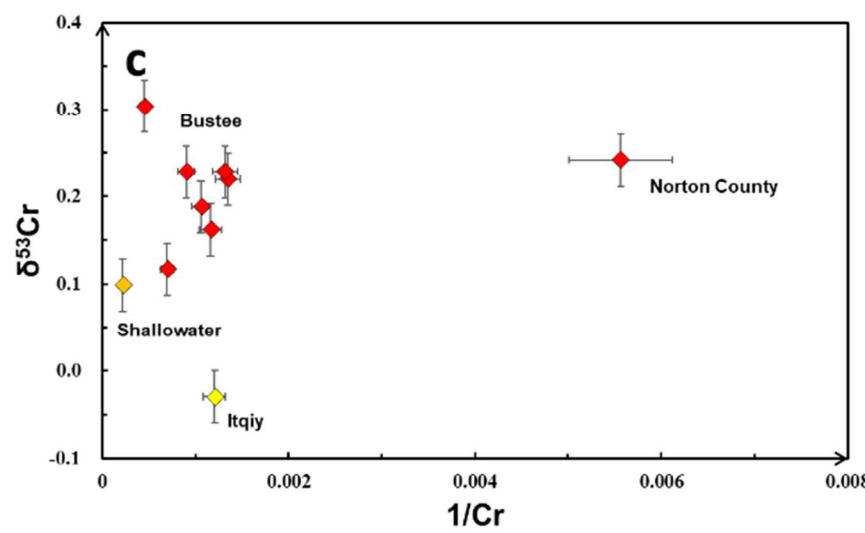
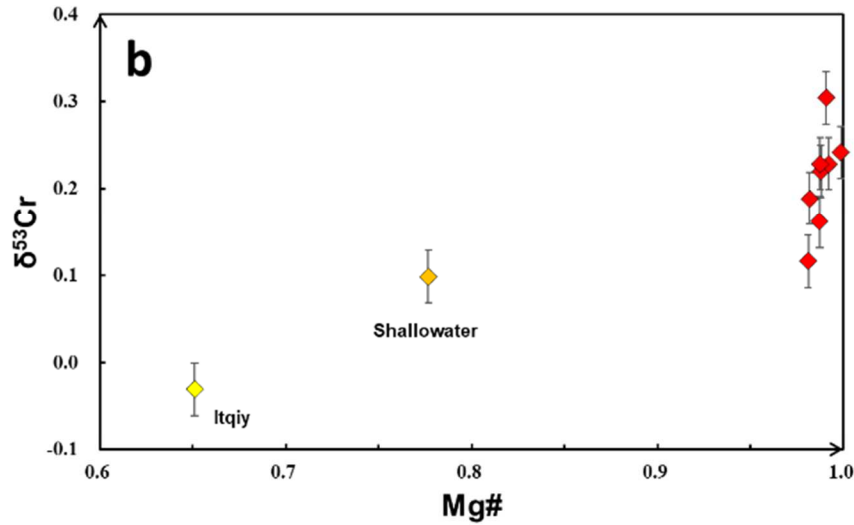
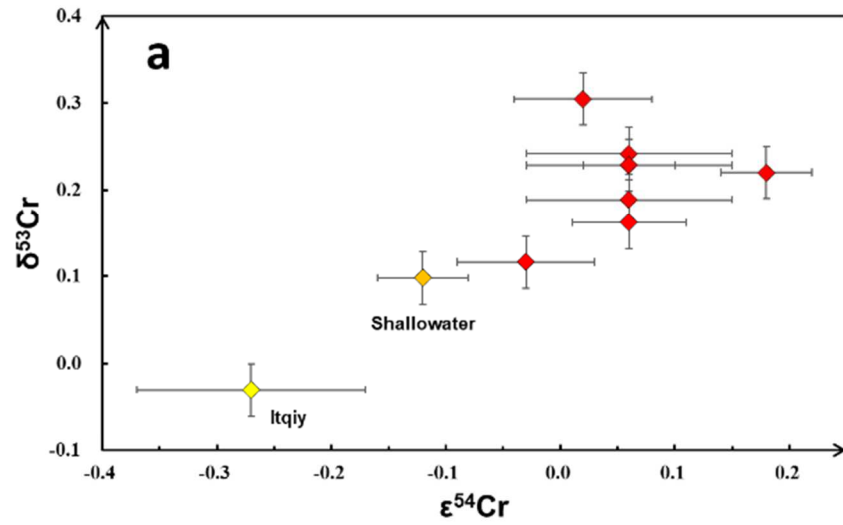
Name	Fall/Find	Texture/Type	Mass (mg)	Cr content (ppm)	Mg#	Fe/Cr	⁵⁵ Mn/ ⁵² Cr	ε ⁵³ Cr	2SE	ε ⁵⁴ Cr	2SE	N/Ref.	Δ ¹⁷ O	Error	Ref.	δ ⁵³ Cr	2SD	N
Khor Temiki	Fall	Fragmental breccia	39.8	742	98.8	14.6	3.77	1.39	0.01	0.18	0.04	5	0.01	0.02	[2]	0.22	0.03	2
Bustee	Fall	Regolith breccia	33.7	2221	99.1	3.5	1.42	2.41	0.03	0.02	0.06	5	0.01	0.00	[2]	0.30	0.03	2
Norton County	Fall	Fragmental breccia	40.0	180	99.9	6.5	4.37	1.53	0.06	0.06	0.09	5	0.01	0.01	[2]	0.24	0.03	1
ALH 78113	Find	Fragmental breccia	43.5	1439	98.1	11.9	2.58	0.97	0.02	-0.03	0.06	5	0.09	0.02	[3]	0.12	0.03	2
ALH 84007	Find	Fragmental breccia	50.7	758	99.2	9.1	2.34	1.08	0.01	0.06	0.04	5	0.05	0.03	[3]	0.23	0.03	2
LAR 04316 (powder)	Find	Fragmental breccia	46.0	1109	98.7	10.0	4.55	1.48	0.02	0.06	0.09	5	0.04	0.01	[2]	0.23	0.03	2
LAR 04316 (pieces)			26.4	861	98.7	13.1	7.46	1.49	0.00*	0.06	0.05	5				0.16	0.03	2
Shallowater	Find	Unbrecciated	30.9	4597	77.7	42.2	0.33	0.00	0.02	-0.12	0.04	5	-0.02	0.02	[3]	0.10	0.03	2
Peña Blanca Spring	Fall	Fragmental breccia	898	394			3.01	1.16	0.05			15/[1]	0.01	0.01	[2]			
Bishopville	Fall	Fragmental breccia	911	273			3.79	1.34	0.08			17/[1]	-0.05	0.06	[3]			
Cumberland Falls	Fall	Fragmental breccia	42.0	945	98.2	17.2	8.40	1.11	0.05	0.06	0.09	5	0.15	0.55	[3,4,5]	0.19	0.03	2
Cumberland Falls-SL			905	549			1.99	1.00	0.09			14/[1]						
Chondritic Clast in Cumberland Falls		Clast														0.65	0.02	[2]
Itqiy#	Find	Unbrecciated-EH7		830	65.1	25.7		0.52	0.04	-0.27	0.10	13	0.06	0.08	[6]	-0.03	0.03	3
MS-MU-19#	Fall	Clast						1.76	0.04	-0.25	0.08	11						
Allende#	Fall	CV chondrite						0.07	0.04	0.81	0.09	10						
DTS-1#		Terrestrial dunite						0.03	0.05	0.15	0.09	12						

Note: References: [1] (Shukolyukov and Lugmair, 2004), [2] (Barrat et al., 2016), [3] (Newton et al., 2000), [4] (Clayton et al., 1984), [5] (Miura et al., 2007) and [6] (Patzner et al., 2002). The δ⁵³Cr and elemental data for Itqiy are from a bulk sample, while the ε⁵³Cr and ε⁵⁴Cr data for Itqiy and MS-MU-19 are from leached

samples with mostly silicate (the metal and sulfide were removed the by HCl). The Cr content, Mg# ($\text{Mg}/(\text{Mg}+\text{Fe})\times 100\%$, atom ratios) and Fe/Cr (atom ratios) are measured by ICP-MS, with an error of 5-10% (2se), except for the data for bulk Itqiy that are from (Patzner et al., 2001). N is then number of runs. The data marked * indicates the 2SE uncertainty is less than 0.004. The data for sample marked # are measured by total evaporation method on Triton TIMS housed at Freie Universität Berlin.

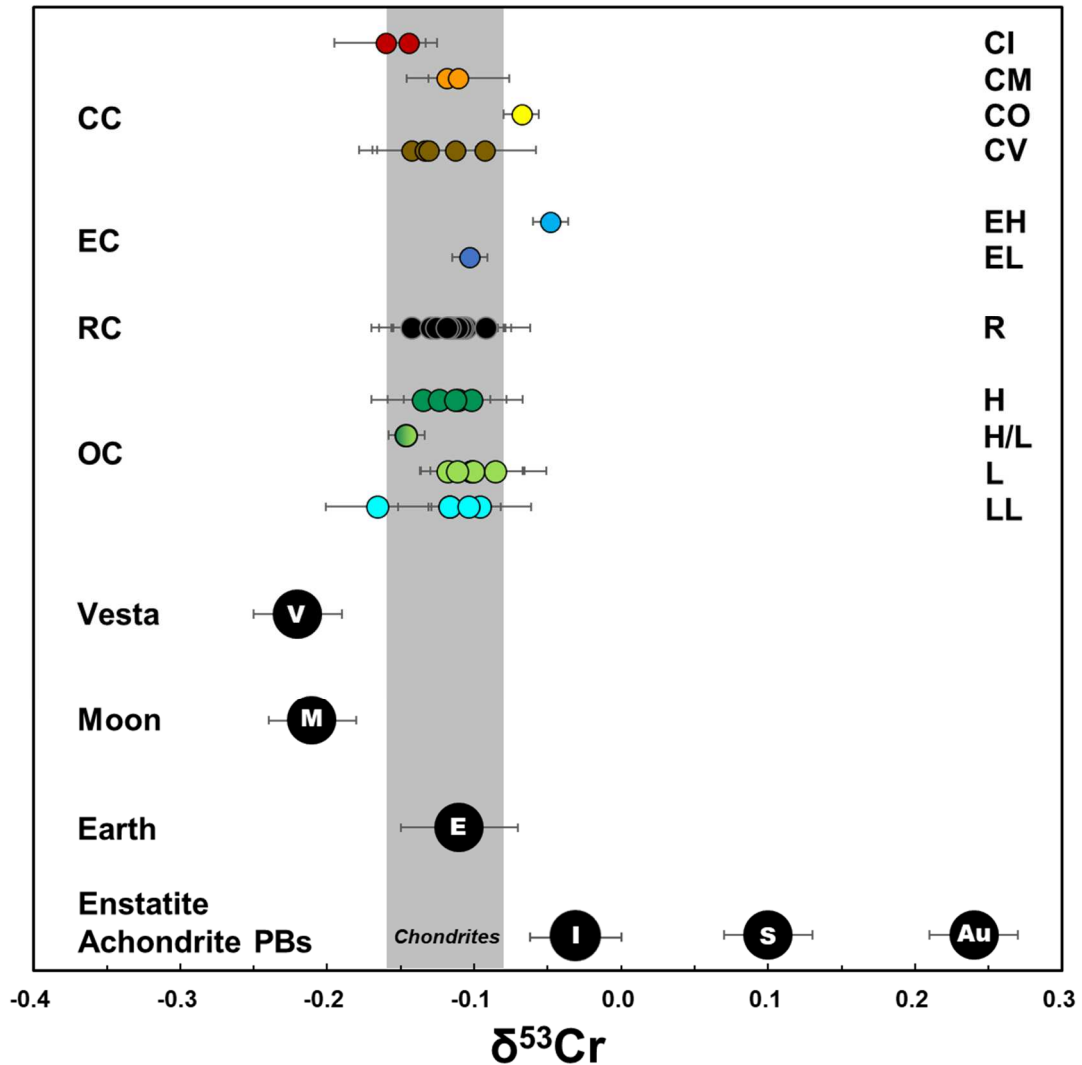


2 **Figure 1a.** The $\epsilon^{54}\text{Cr} - \Delta^{17}\text{O}$ diagram for aubrites, enstatite chondrites and other Solar System
3 materials. The area of enstatite meteorites (in red) overlaps with that of Earth-Moon system.
4 The template and dataset (all the data cited are from published literature and no conference
5 abstracts) are from Zhu et al. (2021b), and the data sources are mainly from Schiller et al.
6 (2014), Trinquier et al. (2007), Yamashita et al. (2010), Qin et al. (2010), Göpel et al. (2015),
7 Pedersen et al. (2019), Mougél et al. (2018), Clayton and Mayeda (1999), Clayton and
8 Mayeda (1984), Greenwood et al. (2010), Clayton et al. (1991), Clayton et al. (1984),
9 Greenwood et al. (2017), Newton et al. (2000), Schrader et al. (2011), Petitat et al. (2011), Zhu
10 et al. (2020b), Zhu et al. (2020a), Zhu et al. (2019b), Li et al. (2018), Zhu et al. (2020b),
11 Yamakawa et al. (2010), Greenwood et al. (2017), Zhu et al. (2020b), Ireland et al. (2020), Zhu
12 et al. (2021a) and Bischoff et al. (2011). The colored circles are chondrites, while the grey
13 shade areas indicate planets/asteroids (i.e., achondrite parent bodies). **Figure 1b.** The
14 zoomed figure for the aubrite area of figure 1a. The enstatite achondrites are diamonds, and
15 the error for $\epsilon^{54}\text{Cr}$ data are reported as the 2SE, while the error for grouping $\epsilon^{54}\text{Cr}$ (shades) of
16 main-group aubrites and Itqiy is the 2SD. The $\epsilon^{54}\text{Cr}$ and $\Delta^{17}\text{O}$ for main-group aubrites overlap
17 those the enstatite chondrite and Earth-Moon system (black bar), but Shallowater has a
18 different $\epsilon^{54}\text{Cr}$ value compared to other aubrites, enstatite chondrites and Earth-Moon system.
19 The red-orange-yellow shades indicate the three enstatite achondrite parent bodies inferred
20 from $\epsilon^{54}\text{Cr}$ systematics.



22 **Figure 2a.** $\epsilon^{54}\text{Cr}$ - $\delta^{53}\text{Cr}$ values for enstatite achondrites, with the red diamonds for main-group aubrites, the orange diamond for Shallowater and the yellow
23 diamond for Itqiy. There is no correlation between $\epsilon^{54}\text{Cr}$ and $\delta^{53}\text{Cr}$ values of main-group aubrites, suggesting the mass-independent data are not influenced by
24 the mass-dependent fractionation. The different $\delta^{53}\text{Cr}$ values between the three groups of enstatite achondrites suggests they formed from different processes,
25 which is consistent with their different $\epsilon^{54}\text{Cr}$ values suggesting at least three enstatite chondrite parent bodies. **Figure 2b.** Mg#- $\delta^{53}\text{Cr}$ values for aubrites. Except
26 Shallowater and Itqiy, all main-group aubrites have high Mg# (98-100%). The three enstatite achondrite parent bodies have correlated Mg# and $\delta^{53}\text{Cr}$ values.
27 **Figure 2c.** $1/\text{Cr}$ - $\delta^{53}\text{Cr}$ values of aubrites. The uncertainty of Cr contents is estimated as 5% (2σ). There is no correlation between $1/\text{Cr}$ and $\delta^{53}\text{Cr}$. The reason
28 for the lack of mixing correlation here could be the sample heterogeneity and secondary processes (most likely impact). **Figure 2d.** $\delta^{56}\text{Fe}$ - $\delta^{53}\text{Cr}$ values for
29 aubrites. The $\delta^{56}\text{Fe}$ isotope data are from Wang et al., (2011), and the data with multiple measurements are averaged with 2SD uncertainty. The correlation
30 between $\delta^{56}\text{Fe}$ and $\delta^{53}\text{Cr}$ values for main-group aubrites should reflect a mixing between sulfide-metal and silicates. The sulfide-metal are rich in both
31 isotopically light Cr and Fe, while the silicates possess isotopically heavy Cr and Fe.

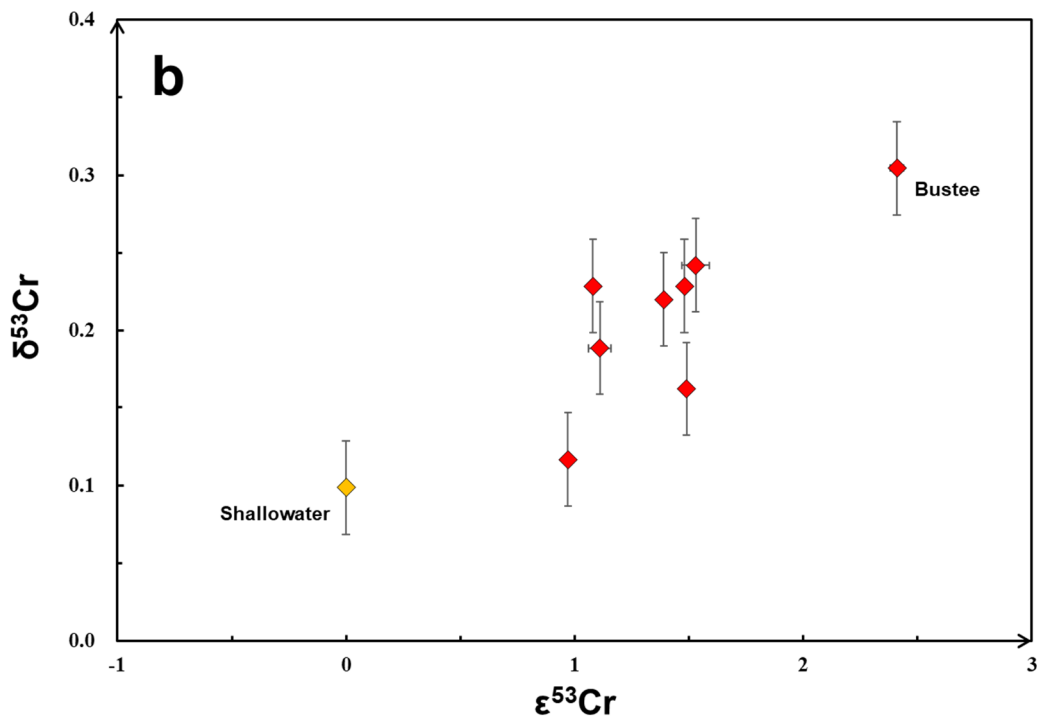
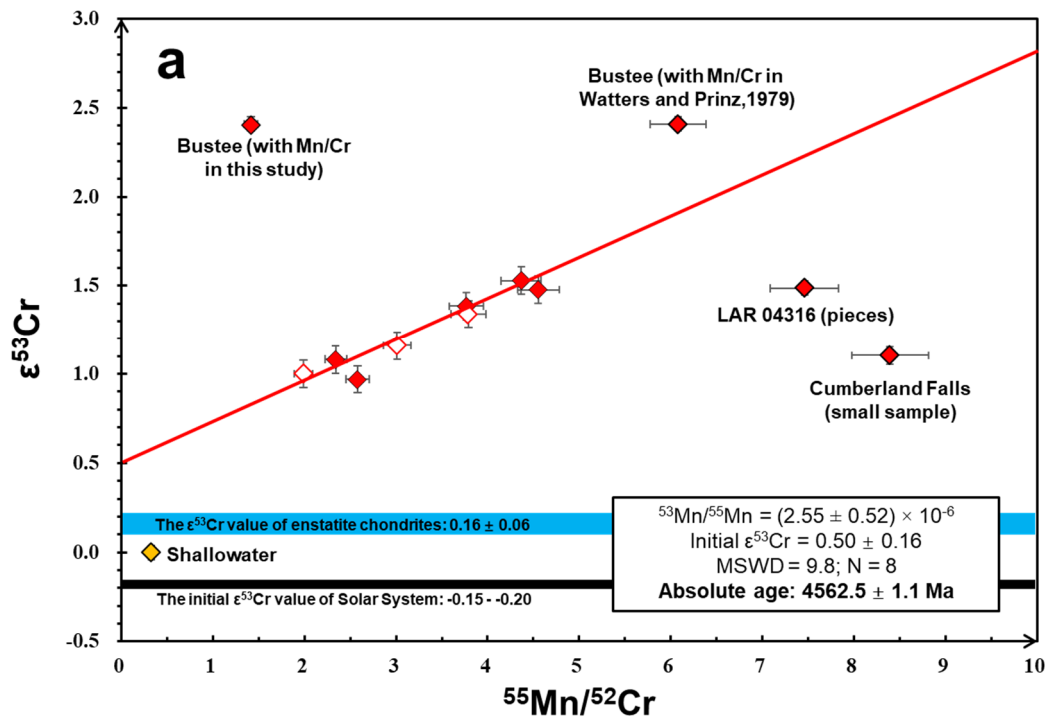
The $\delta^{53}\text{Cr}$ Variation in Chondrites and Planets



32

33 **Figure 3.** Comparison of the $\delta^{53}\text{Cr}$ variations amongst chondrites, achondrites and the
 34 Earth-Moon system. The small colorful circles are chondrites, while the big black circles
 35 represent the Earth, Moon, Vesta and enstatite achondrite parent bodies. The gray bar defines
 36 the average $\delta^{53}\text{Cr}$ values (-0.12 ± 0.04 ; 2SD, $N = 42$) of all the chondrites, and the three
 37 enstatite achondrite parent bodies have distinguished mass-dependent Cr isotope
 38 compositions ($\delta^{53}\text{Cr}$ values): 0.24 ± 0.03 ‰, 0.10 ± 0.03 ‰ and -0.03 ± 0.03 ‰ for main-group
 39 aubrite (Au), Shallowater (S) and Itqiy (I) parent bodies respectively, which is consistent their
 40 $\epsilon^{54}\text{Cr}$ characteristics. The $\delta^{53}\text{Cr}$ difference between enstatite achondrite parent bodies and
 41 chondrites result from a sulfur-rich core formation. Itqiy and Shallowater parent bodies have
 42 lower $\delta^{53}\text{Cr}$ values than main-group aubrites, which may result from more sulfides-metal
 43 contents (lower Mg#) in Shallowater and Itqiy. This is consistent with the mixing models in
 44 figure 2d. Literature data sources: chondrites (Bonnand et al., 2016b; Schoenberg et al., 2016),
 45 Earth (Jerram et al., 2020; Schoenberg et al., 2008; Sossi et al., 2018), Moon (Bonnand et al.,
 46 2016a; Sossi et al., 2018) and HEDs-Vesta (Zhu et al., 2019c).

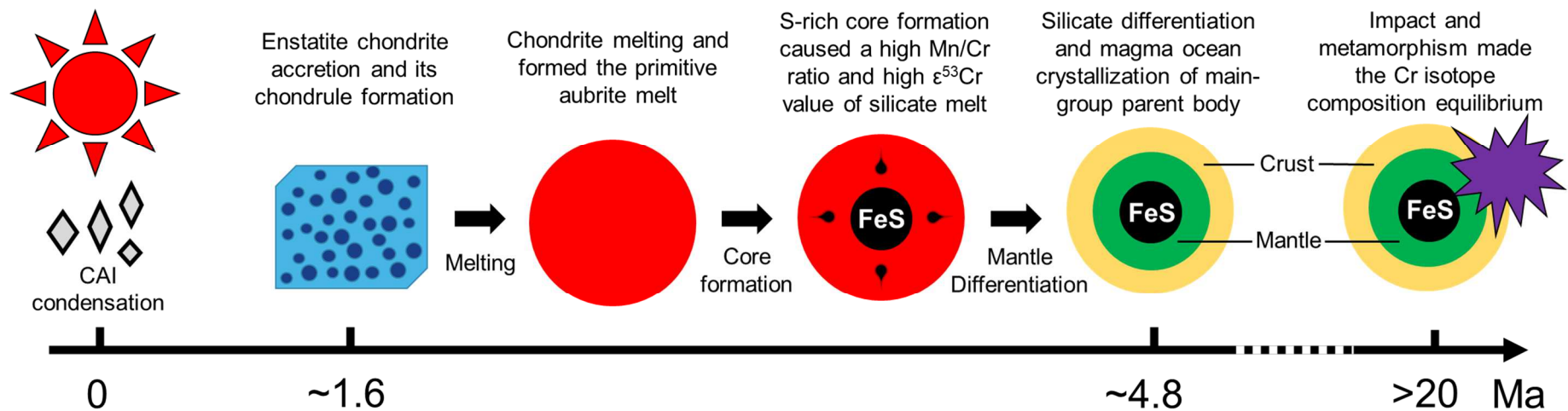
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48

49 **Figure 4a.** The ^{53}Mn - ^{53}Cr isochron for bulk aubrites, the open diamonds are the Cr isotope
 50 data from Shukolyukov and Lugmair (2004). This red correlation line does not include
 51 Shallowater, Bustee, LAR 04316 (pieces) and Cumberland Falls (small sample) from this study,
 52 due to the different parent bodies and sample heterogeneity. The blue and black bars are the
 53 $\epsilon^{53}\text{Cr}$ value of enstatite chondrites and the initial $\epsilon^{53}\text{Cr}$ value of Solar System that is estimated
 54 by D'Orbigny angrite, CI chondrite and chondrule precursors (Göpel et al., 2015; Trinquier et

55 al., 2008b; Zhu et al., 2019a). The ^{53}Mn - ^{53}Cr correlation line is controlled by co-genetic sulfide
56 and silicates, which also holds the time significance reflecting the differentiation of main-group
57 parent body and crystallization of sulfide and silicates. More importantly, the elevated initial
58 $\epsilon^{53}\text{Cr}$ value (0.50 ± 0.16 , 2σ ; higher than that of the Solar System and enstatite chondrites),
59 expressed by the intercept of y axis, could be caused by the early S-rich core formation
60 process. **Figure 4b.** The $\delta^{53}\text{Cr}$ values correlate with the $\epsilon^{53}\text{Cr}$ values that mainly reflect the
61 true Mn/Cr ratios (since there is a large Mn/Cr heterogeneity in aubrites). This correlation
62 indicates that the aubrites reflect mixing of lithologies with more abundant Cr-rich sulfide (low
63 Mn/Cr, low $\epsilon^{53}\text{Cr}$ and low $\delta^{53}\text{Cr}$) and lithologies with Cr-poor silicates (high Mn/Cr, high $\epsilon^{53}\text{Cr}$
64 and high $\delta^{53}\text{Cr}$).



65
 66 **Figure 5** The timeline of evolution of the main-group aubrite parent body. The Ca-Al-rich inclusion condensation marked the formation of the Solar System (t_0 ,
 67 4567.30 ± 0.16 Ma) (Connelly et al., 2012). Then, enstatite chondrite chondrules formed at 4565.7 ± 0.7 Ma (precursor age), i.e. ~ 1.6 Ma after t_0 (Zhu et al.,
 68 2020a), and the accretion of enstatite chondrites is estimated as ~ 1.8 Ma after t_0 (Alexander et al., 2018). These two ages can potentially represent the
 69 accretion time of enstatite chondrites. After that, enstatite chondrites experienced a melting event and formed the primitive aubrite melt. At this stage, this melt
 70 has chondritic Mn/Cr ratio. Then, the sulfur-rich core formation process changed the composition of the primitive melt, making the core rich in Cr and poor in
 71 Mn (Berthet et al., 2009), while the silicate melt possessing high Mn/Cr ratio. In this way, after the decay of ^{53}Mn , the silicate melt has a high $\epsilon^{53}\text{Cr}$ value that is
 72 the source of main-group aubrites. It furthermore experienced a silicate differentiation and mineral crystallization that fractionates the Mn/Cr ratios in
 73 main-group aubrites. The ^{53}Mn - ^{53}Cr system recorded the age of this event, 4562.5 ± 1.1 Ma (i.e. 4.8 ± 1.1 Ma after CAIs), which indicates the differentiation
 74 time of main-group aubrite parent body. Finally, after extinction of ^{53}Mn (around eight times of the half-life of ^{53}Mn), i.e. ~ 20 Ma after t_0 , there should be a
 75 metamorphosed process, mostly caused by impact, redistributed the Mn and Cr in individual aubrites and reset the internal ^{53}Mn - ^{53}Cr isochrons.

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