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The effects of Antarctic alteration and sample heterogeneity on Sm-Nd and 1 Lu-Hf systematics in H chondrites 2 Ryoga Maeda^{a,b,*}, Steven Goderis^a, Vinciane Debaille^b, Hamed Pourkhorsandi^b, 3 Geneviève Hublet^b, Philippe Claeys^a 4 ^a Analytical-, Environmental-, and Geo-Chemistry, Vrije Universiteit Brussel, Pleinlaan 2, BE-1050 5 6 Brussels, Belgium. 7 ^b Laboratoire G-Time, Université libre de Bruxelles, CP 160/02, 50, Av. F.D. Roosevelt, BE-1050, Brussels, 8 Belgium. 9 * Corresponding author. *E-mail address*: Ryoga.Maeda@vub.be (R. Maeda)

10 Abstract

11 Long-lived radioactive isotope systematics, such as Sm-Nd and Lu-Hf, are useful tools as important 12 chronometers and tracers for chemical differentiation processes. Even though Antarctic meteorites include 13 rare meteorites such as ungrouped, shergottites-nakhlites-chassignites, and lunar meteorites, the effects of 14 Antarctic alteration on the Sm-Nd and Lu-Hf systems in chondrites have not yet been evaluated in detail. 15 Moreover, the heterogeneity of Sm-Nd and Lu-Hf data in bulk chondrites prevents the determinations of 16 precise average Sm-Nd and Lu-Hf values for individual chondrite groups and such robust values of the 17 Chondritic Uniform Reservoir (CHUR). To examine the effects of Antarctic alteration and sample 18 heterogeneity on the Sm-Nd and Lu-Hf isotope systematics, ten Antarctic H chondrites (HCs) and three HCs 19 from hot deserts were characterized for their modal abundances, major and trace elemental concentrations, 20 as well as Sm-Nd and Lu-Hf isotopic compositions. Regardless of the classical weathering index for Antarctic 21 meteorites and the normalized Rb abundance used as a chemical alteration indicator in this study, the modal 22 and elemental abundances in Antarctic HCs appear to be in good agreement with those in non-Antarctic HCs. 23 The Sm-Nd and Lu-Hf isotopic compositions of the characterized H chondrites fall within the range measured 24 for both HC falls and for falls of other chondrite classes, except in the case of the most heavily altered samples. 25 Consequently, the effects of Antarctic alteration processes on the Sm-Nd and Lu-Hf systematics in HCs 26 appear to be limited, except in the case of A 09516 (H6). The latter meteorite exhibits severe mineralogical 27 and chemical alteration, with considerable losses of even the rare earth elements (REEs), which are considered relatively immobile. The ¹⁴⁷Sm/¹⁴⁴Nd, ¹⁴³Nd/¹⁴⁴Nd, ¹⁷⁶Lu/¹⁷⁷Hf, and ¹⁷⁶Hf/¹⁷⁷Hf of bulk HCs 28 29 correlate with their P/Mg and Y/Mg. Furthermore, the Lu-Hf ratios are strongly correlated with their P/Ca 30 and Y/Ca as well as their P/Mg and Y/Mg. Thus, the distribution of the elements between constituent minerals 31 in ordinary chondrites (OCs) may control the heterogeneity observed for the bulk Sm-Nd and Lu-Hf data. In 32 this context, the weight ratio of Ca-phosphates to Ca-pyroxene, or at least that of Ca-phosphates to silicates, 33 may be a key factor leading to the observed elemental and isotopic variations. This observation indicates that 34 the nugget effect of Ca-phosphates in OCs as the result of insufficient homogenization or terrestrial alteration 35 leads to the heterogeneities displayed by the Sm-Nd and Lu-Hf data. Moreover, it also indicates that the use

of equilibrated OCs for the determination of Sm-Nd and Lu-Hf data is affected more by sample heterogeneity, 36 especially with respect to Ca-phosphates, than is the case for unequilibrated OCs, based on the re-distribution 37 of REEs during thermal metamorphism on their parent bodies. This study demonstrates that Antarctic 38 39 meteorites commonly preserve their original Sm-Nd and Lu-Hf isotopic compositions as much as chondrite 40 falls, although exceptions are possible in the case of severe alteration. Similar to previous studies, we 41 recommended the use of unequilibrated chondrites, for which the re-distribution of REEs is less extensive, 42 for the determination of well-constrained average Sm-Nd and Lu-Hf isotopic compositions for individual 43 chondrite groups as well as that of the robust Sm-Nd and Lu-Hf CHUR values.

44 **1 INTRODUCTION**

In 1969, a Japanese expedition found nine meteorites in Antarctica, which in 1973 led to the first 45 46 systematic meteorite recovery expedition on the Southern continent (Yamaguchi et al., 2021), which was 47 followed by expeditions until the present (e.g., Goderis et al., 2021). Today, Antarctic meteorites constitute 48 by number more than half of all known meteorites and include many rare varieties such as ungrouped 49 chondrites and achondrites, shergottites-nakhlites-chassignites, and lunar meteorites (Meteorite Newsletter, NIPR; Antarctic Meteorite Newsletter, NASA). Hence, the Antarctic meteorite collection plays a pivotal role 50 51 in understanding the history of the Solar System. Meteorites recovered after observed fall events are termed 52 "falls"; those that cannot definitely be associated with observed falls, such as Antarctic meteorites, are named 53 "finds." Finds are generally characterized by a much longer terrestrial residence time than falls (roughly of 54 the order of tens of kyr: e.g., Al-Kathri et al., 2015). Many finds become weathered during this long residence 55 time, and this process can take place relatively rapidly, particularly in temperate and tropical zones, as well 56 as in hot deserts (Bland et al., 2006; Pourkhorsandi et al., 2021). On average, Antarctic meteorites exhibit 57 old terrestrial ages, on the order of hundreds of kyr (e.g., Nishiizumi et al., 1989), and these are better 58 protected from terrestrial weathering while buried in the ice compared to finds collected from hot deserts 59 (Whillans and Cassidy, 1983; Cassidy and Whillans, 1990). However, these meteorites were exposed to the 60 extreme Antarctic climate during an extended period of time, leading to significant mineralogical and 61 chemical alteration (e.g., Koebel and Cassidy, 1991; Bland et al., 2006). 62

Rare earth elements (REEs) are refractory lithophile elements that share similar chemical properties due 63 to their electron configurations, implying that they should not have been fractionated by condensation and 64 volatilization processes in the early Solar System or by metal-silicate separation during core formation in 65 differentiated planets. Therefore, REEs are universally used for understanding specific planetary processes, such as silicate differentiation. The REEs include long-lived radioactive isotopes such as ¹⁴⁷Sm and ¹⁷⁶Lu, 66 which decay to ¹⁴³Nd (half-life ~106 Gyr; Lugmair and Marti, 1978) and ¹⁷⁶Hf (half-life ~37.1 Gyr; Patchett 67 68 et al., 2004), respectively. Their isotope systematics form the basis of important chronometers and tracers of 69 chemical differentiation processes. The Sm-Nd and Lu-Hf isotope systematics of chondritic meteorites are 70 particularly important as the Chondritic Uniform Reservoir (CHUR), which constitutes the average reference 71 for undifferentiated planetary material in the Solar System (DePaolo and Wasserburg, 1976). Bouvier et al. 72 (2008) determined the currently most precise CHUR values of the Sm-Nd and Lu-Hf data as well as the

respective isochrons by utilizing unequilibrated carbonaceous, ordinary, and enstatite chondrites of petrologic

types 1-3, mostly from falls, since scatter increases when using equilibrated chondrites. Consequently, the

75 Sm-Nd and Lu-Hf CHUR values are regarded as the equivalents of the composition of Bulk Silicate Earth.

As Bouvier *et al.* (2008) discussed, the heterogeneity of Sm-Nd and Lu-Hf data in bulk chondrites is one of

- the largest problems when willing to determine precise average values (*i.e.*, CHUR values or an average of a
- 78 group of chondrites), as the observed spread leads to larger uncertainties on the mean. For example, such
- heterogeneity occurs even on a duplicate analysis of Lu-Hf values in an H4 chondrite ($^{176}Lu/^{177}Hf = 0.0301$
- 80 \pm 0.0002 and 0.0334 \pm 0.0002, 176 Hf/ 177 Hf = 0.282444 \pm 0.000009 and 0.282761 \pm 0.000016 in Ochansk;
- 81 Patchett *et al.*, 2004).

82 Previous studies regarding Sm-Nd and Lu-Hf systems in bulk chondrites have focused on falls to avoid 83 any possible weathering occurring during long terrestrial residence (e.g., Jacobsen and Wasserburg, 1984; 84 Blichert-Toft and Albarède, 1997, Patchett et al., 2004; Bouvier et al., 2008). Terrestrial weathering can affect 85 REEs, although they are often considered to be relatively immobile (e.g., Shimizu et al., 1983; Crozaz, 2003; 86 Al-Kathri et al., 2015; Pourkhorsandi et al., 2017). The effects of Antarctic alteration on REE abundances 87 were only examined for achondrites, and more specifically for eucrites (Shimizu et al., 1983; Mittlefehldt 88 and Lindstrom, 1991; Crozaz, 2003). Today, few studies exist for chondrites (Tatsumoto et al., 1981; 89 Nishikawa et al., 1990). In particular, the effects of Antarctic alteration on the Sm-Nd and Lu-Hf systems in 90 bulk chondrites remain poorly understood.

91 As such, even though the Antarctic collection includes meteorite groups and clans found only in 92 Antarctica, the effects of Antarctic alteration on Sm-Nd and Lu-Hf systems in chondrites have not been 93 assessed on a systematic way. Therefore, in this study, the results of the Sm-Nd and Lu-Hf isotope systematics 94 in Antarctic HCs are examined in a methodical manner to evaluate weathering effects induced by extended 95 residence times within the Antarctic ice. Moreover, this work also investigates the constituent minerals that 96 control the heterogeneity of these isotope systems in bulk HC by combining the isotopic ratios and elemental 97 abundance ratios, which can aid in determining average Sm-Nd and Lu-Hf isotopic compositions with limited 98 uncertainty (*i.e.*, precise average Sm-Nd and Lu-Hf isotopic compositions) for individual chondrite groups.

99 2 EXPERIMENTAL

100 **2.1 Samples**

101 Ten Antarctic HCs and three HCs collected from hot deserts have been characterized for their modal 102 abundances, and elemental and isotope compositions. Four of the Antarctic HCs, Asuka (A) 09436 (H3), A 103 09387 (H4), A 09618 (H5), A 09516 (H6), and the three hot desert HCs, northwest Africa (NWA) 6752 (H3), NWA 6771 (H4), and Sahara 97035 (H5), were provided by the Royal Belgian Institute of Natural Sciences 104 105 (RBINS), Belgium. Meteorites A-881258 (H3.0), A-880941 (H3.3), Yamato (Y-)793574 (H3.5), Y-790461 106 (H3.7), Allan Hills (ALH) 78084 (H3.9), and Y-790960 (H7) were allocated by the National Institute of Polar 107 Research (NIPR), Japan. Available information on the meteorites is listed in Table 1. To clarify any potential effect of terrestrial weathering on the inner portions of these chondrites - even if the sample visually appears 108 109 fresh -, only the interior portions were used to avoid as much as possible fusion crust or sample fragments

110 directly affected by terrestrial weathering.

111 2.2 In-situ measurement

112 Polished thick sections were prepared for all of the meteorites, except in the case of NWA 6752 (H3) when a polished thin section was used. These polished thick and thin sections were analyzed using a Bruker M4 113 114 Tornado micro-X-ray fluorescence (μ XRF) scanner equipped with a Rh source and two XFlash 430 Silicon 115 Drift detectors at the Vrije Universiteit Brussel (VUB), Belgium. The M4 set up is described in detail in 116 Winter and Claeys (2017), and for each sample, elemental maps of 16 elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Ni, and Zn) were obtained. To produce the element maps, the 25 µm X-ray beam was 117 118 applied under vacuum conditions (20 mbar), with 10 µm step size, 1 ms dwell time per point and maximized 119 source energy settings (50 kV, 600 μ A) without a source filter. Constituent minerals in the samples were 120 identified from the elemental maps and their modal abundances were determined using the ImageJ image 121 analysis program, while confirming the nature of these phases using a JEOL JSM-IT300 scanning electron 122 microscope (SEM) equipped with an Oxford energy dispersive spectrometer (EDS) at the SURF research 123 unit of VUB. Uncertainties for the modal abundances determined using this method are estimated to be better 124 than 10% relative.

125 2.3 Bulk analyses

126 To examine the effects of sample heterogeneity as well as Antarctic weathering on the Sm-Nd and Lu-Hf 127 systems, a relatively small sample volume was preferred in this study to amplify sample heterogeneity caused by sampling. Approximately 200 mg of fragments of each sample were homogenized using an agate mortar 128 129 and pestle dedicated to chondrites. No leaching to remove terrestrial materials was applied, even in the case 130 of the three hot desert HCs. Approximately 100 mg of each powdered sample, which is the smallest possible 131 amount to measure the elemental abundances and the isotope compositions based on their detection limits, 132 was dissolved in Teflon beakers using a mixture of ultrapure concentrated HF-HNO₃ (1:3) at 120 °C overnight. Because undigested residue remained in the beakers, possibly including acid-resistant phases such as oxides, 133 134 the residues were then transferred into Teflon bombs for high-pressure dissolution, using the same acid 135 mixture at 150 °C for 2 days. During the high-pressure dissolution, the supernatants remaining in the first 136 Teflon beaker were evaporated. After the high-pressure dissolution, the solutions in the bombs were added to 137 the Teflon beakers containing the residues of the supernatants. Following evaporation, the residues were 138 dissolved in concentrated HCl and evaporated again. After re-dissolution in 1.5 M HCl with a trace amount 139 of concentrated HF, a ~2% aliquot of each solution was taken for the determination of major and trace elemental abundances. Similarly, a ~5% aliquot of each solution was spiked with mixed ¹⁴⁸Nd-¹⁵⁰Sm and 140 ¹⁷⁶Lu-¹⁷⁹Hf spikes. The spiked aliquots were heated on a hotplate for at least 1 day for spike-sample 141 142 equilibration.

The REEs and Hf were separated from the main fraction by cation exchange chromatography using 2 mL of AG50W-X8 resin (100-200 mesh for the spiked aliquots, 200-400 mesh for the unspiked aliquots). The Hf cut was collected first during the loading of the sample and the following rinsing step using 1.5 M HCl with a trace amount of concentrated HF, while the REE cut was subsequently collected using 6 M HCl. The

REE cuts were loaded onto a column with 2 mL of HDEHP resin to separate Nd for both the unspiked and 147 spiked aliquots, and Sm and Lu for the spiked aliquots. The Hf cuts were further purified from Fe by anion 148 149 exchange chromatography in 6 M HCl using 2 mL of AG1X8 resin (100-200 mesh for the spiked aliquots, 150 200-400 mesh for the unspiked aliquots), and the remaining matrix, especially Ti, was removed on 2 mL of 151 Eichrom LN-Spec resin using 6 M HCl with a trace amount of H₂O₂. Finally, Hf was collected in 4 M HF. 152 The yields of Nd, Sm, Lu, and Hf are estimated to be better than 90%, 95%, 95%, and 90% respectively, for 153 both unspiked and spiked aliquots (Debaille *et al.*, 2007). Total procedural blanks were determined to be \leq 154 26 pg and \leq 10 pg for Nd and Sm, respectively, and \leq 4.5 pg and \leq 20 pg for Lu and Hf, respectively. No 155 blank correction was applied due to these negligible blank levels, even on the spiked aliquots (the blank-to-156 sample rations of more than 1%) because they represent insignificant contributions. The chemical procedures 157 outlined here were performed at the Université libre de Bruxelles (ULB), Belgium, and are based on procedures described in detail in Armytage et al. (2018) and Debaille et al. (2007, 2017). Reference basalt 158 159 BHVO-2 of the United States Geological Survey was subjected to the same procedures and measured in 160 duplicate to validate the analytical accuracy and reproducibility during the following measurements using 161 inductively coupled plasma-optical emission spectrometry (ICP-OES), quadrupole-ICP-mass spectrometry 162 (Q-ICP-MS), and multiple collector-ICP-MS (MC-ICP-MS). Approximately 50 mg of BHVO-2 was weighed 163 and prepared at similar concentrations for measurement to the chondrite samples for both unspiked and spiked

aliquots. Samples A 09436 (H3) and A 09516 (H6) were also analyzed in duplicate. All duplicate analyses

165 were carried out on different powder subsamples.

166 2.3.1 Measurement of major and trace elemental abundances using ICP-OES and Q-ICP-MS

The major elemental abundances (Na, Mg, Al, P, K, Ca, Ti, Cr, Mn, Fe, Co, and Ni) were determined
using a Thermo Scientific iCAP 7000 Plus Series ICP-OES at ULB. Yttrium was used as the internal standard,
and the dilution factors (DFs) for the meteorite samples and the reference basalt were set to be 15,000 and
10,000 respectively, using 5% HNO₃.

171 The trace elemental abundances (Cu, Zn, Rb, Sr, Y, Zr, Nb, Ba, REEs, Hf, Th, and U) were determined 172 using an Agilent 7700 ICP-MS at ULB. Measurement solutions were prepared with 5% HNO3 and all of the 173 measured elements including the high field strength elements were analyzed in 5% HNO₃ without HF. For 174 Q-ICP-MS analysis, In was used as the internal standard, and the DFs for the meteorite samples and the 175 geological reference materials were set to be 2,000 and 6,000, respectively, in order to minimize the influence 176 of non-spectral matrix effects (Ebihara et al., 2020). Although slight analyte loss has been observed during 177 dissolution (Ebihara et al., 2020), no correction for such analyte loss was applied here. Individual single 178 element standards of Ba, Ce, Pr, and Nd were used to correct isobaric interferences from their oxide molecules. The results for the elemental abundances of BHVO-2 (n = 2) are shown in Table 2, together with their 179 180 preferred values (Jochum et al., 2016).

181 2.3.2 Isotope ratio measurement using MC-ICP-MS

All of the unspiked and spiked cuts were analyzed in 0.05 M HNO₃ (with 0.05 M HF for the Hf cuts) using the Nu instruments MC-ICP-MS Nu-Plasma II equipped with an Aridus II desolvating nebulizer at

ULB. For isotope measurements of Nd, the unspiked Nd cuts were diluted to 20 ppb. The isobaric interference 184 of ¹⁴⁴Sm on ¹⁴⁴Nd was monitored on mass ¹⁴⁷Sm, but no Sm was detected. Measured Nd isotope ratios were 185 normalized to 146 Nd/ 144 Nd = 0.7219 using an exponential law following Debaille *et al.* (2007) to correct for 186 instrumental mass fractionation. Measurements of the meteorite samples and BHVO-2 were bracketed by the 187 measurement of a 20 ppb Rennes in-house Nd standard. The measured ¹⁴³Nd/¹⁴⁴Nd values of the standard 188 were re-calculated to the accepted ¹⁴³Nd/¹⁴⁴Nd value of the Rennes in-house Nd standard (0.511961; Chauvel 189 190 and Blichert-Toft, 2001). For isotope measurements of Hf, the unspiked Hf cuts were prepared at 5 ppb. Isobaric interferences for Hf from Yb, Lu, and W were monitored on masses ¹⁷²Yb, ¹⁷⁵Lu, and ¹⁸²W, 191 respectively. No detectable Yb, Lu, or W were found in the unspiked Hf cuts. Measured Hf isotope ratios 192 were normalized to ${}^{179}\text{Hf}/{}^{177}\text{Hf} = 0.7325$ using the exponential law to correct for instrumental mass 193 fractionation. A 5 ppb JMC-475 Hf standard was used to bracket each sample during the runs. The measured 194 195 176 Hf/ 177 Hf values of the Hf standard were re-calculated to the accepted 176 Hf/ 177 Hf value of the JMC-475 Hf 196 standard (0.282163; Blichert-Toft et al., 1997). Internal reproducibilities of Nd and Hf are better than 32 ppm and 31 ppm for all standards, respectively. The means of ¹⁴³Nd/¹⁴⁴Nd values and ¹⁷⁶Hf/¹⁷⁷Hf values in BHVO-197 2 are 0.512971 ± 0.000012 (2SE, n = 4) and 0.283102 ± 0.000011 (2SE, n = 10), as shown in Table 2. These 198 199 values overlap within uncertainty with published reference values (0.512984 ± 0.000011 ; Weis *et al*, 2006 200 and 0.283105 ± 0.000011 ; Weis *et al*, 2007, respectively).

201 For spiked cuts, the mass fractionation for each element was corrected using an exponential law followed 202 by spike stripping using an iterative method (Debaille et al., 2007). The same isotopes as in unspiked Nd and 203 Hf analyses were monitored to check for isobaric interferences during spiked Nd and Hf analyses. No 204 detectable contributions of these isotopes were observed in the spiked Nd and Hf cuts. During Sm measurement, ¹⁴⁷Sm, ¹⁵⁰Sm, ¹⁵²Sm, and ¹⁵⁴Sm were measured. Here, ¹⁵⁰Sm is included due to the use of the 205 ¹⁵⁰Sm enriched spike despite the isobaric interference by ¹⁵⁰Nd. The presence of Nd and Gd in the Sm cuts 206 was monitored on masses ¹⁴⁶Nd and ¹⁵⁶Gd, and the presence of Gd was corrected mathematically. Even 207 though a trace of Nd was observed in the Sm cuts, the contribution was considered negligible. The isobaric 208 interference of ¹⁷⁶Hf was monitored on mass ¹⁷⁷Hf during Lu analysis, but no Hf was detected in the Lu cuts. 209 210 Because Lu possesses only two stable isotopes, a mass fractionation correction for Lu must be applied, for 211 example by using Yb (Blichert-Toft et al., 1997). The uncertainties on the Nd, Sm, Lu, and Hf concentrations 212 are dominated by the uncertainty of the spike calibration, which is estimated to be better than 0.5% (2SE). Therefore, the uncertainties on the concentrations are estimated to be 0.5% as a maximum and are presented 213 in Tables 2 and 4. The obtained 147 Sm/ 144 Nd and 176 Lu/ 177 Hf values for BHVO-2 are 0.1515 ± 0.0011 (2SE) 214 and 0.0088 ± 0.0001 (2SE), shown in Table 2, which are in good agreement with their preferred values of 215 0.1501 ± 0.0010 and 0.00878 ± 0.00006 , respectively (Jochum *et al.*, 2016). 216

217 **3 RESULTS**

218 **3.1 Modal abundances**

Table 3 summarizes the modal abundances in all samples obtained using the μ XRF, including the mean values of unequilibrated H chondrites (UHCs; n = 7) and equilibrated H chondrites (EHCs; n = 6), together

with the published modal abundances of EHCs (Van Schmus, 1969; McSween et al., 1991). According to the 221 mineral observation using SEM-EDS in this study, forsterite, silica, and a few accessory minerals (Table 3) 222 223 can be found in a limited number of samples. Also, H chondrites generally contain ilmenite (~0.2 wt.%: e.g., 224 McSween et al., 1991). However, the mapping using µXRF is not sufficiently quantitative and the minimum spot size (25 µm) used in this study is larger than the size of some mineral phases (e.g., silica and ilmenite), 225 226 which does not allow distinguishing forsterite from low-Ca pyroxene (especially enstatite), silica from 227 feldspar, or ilmenite from chromite. Therefore, the modal abundances of low-Ca pyroxene, feldspar, and 228 chromite obtained in this study include the abundances of forsterite, silica, and ilmenite, respectively. 229 Moreover, grains smaller than $\sim 5 \,\mu m$, such as tiny Ca-phosphates especially present in UHCs, might have 230 been missed because of this limitation. However, forsterite and silica are only observed in some UHCs (for 231 forsterite: A-881258, Y-793574, Y-790461, and NWA 6752; for silica: Y-790461 and A 09436) and A 09387 232 (H4), and these were confirmed to be present in the HCs in trace abundances only. Based on this work, there 233 is no resolvable difference in the modal abundances between Antarctic HCs and hot desert HCs, thus the 234 mean values of UHCs and EHCs are calculated from the combined data for Antarctic and hot desert HCs 235 (Fig. 1). Most of the mean modal abundances in UHCs and EHCs are in good agreement with each other 236 (Table 3 and Fig. 1). These mean modal abundances also compare well with the published values listed in Table 3. The mean modal abundance of total Ca-phosphates in EHCs is 0.70 ± 0.10 wt. % at the 95% 237 238 confidence interval (95% CI) and agrees within uncertainty with previously published values. In addition, 239 the mean modal abundances of chlorapatite and merrillite in UHCs and EHCs overlap within uncertainty 240 $(0.10 \pm 0.07 \text{ wt. }\% \text{ vs. } 0.17 \pm 0.14 \text{ wt. }\% \text{ and } 0.29 \pm 0.08 \text{ wt. }\% \text{ vs. } 0.53 \pm 0.24 \text{ wt. }\%, \text{ respectively at the}$ 95% CI; Table 3). However, the mean modal abundance of total Ca-phosphates in UHCs is 0.39 ± 0.09 wt. % 241 242 (95% CI), which is significantly lower than the abundance in EHCs (Fig. 1).

243 **3.2 Elemental abundances**

244 The major and trace elemental abundances in bulk samples measured using ICP-OES and ICP-MS are 245 summarized in Table 4, with mean values for the Antarctic UHCs (n = 7), Antarctic EHCs (n = 3), Antarctic 246 HCs (n = 10) and hot desert HCs (n = 3), and literature values for non-Antarctic HCs (Jarosewich, 1990 for 247 Na. Mg, Al, P, K, Ca, Ti, Cr, Mn, Fe, and Ni; Wasson and Kallmeyn, 1988 for Cu, Zn, Rb, Sr, Y, Zr, Nb, Ba, and Hf; Nakamura, 1974 for REEs; Tatsumoto et al, 1973 for Th and U). The data of A 09516 (H6) are 248 249 excluded from the mean values of Antarctic EHCs and Antarctic HCs because of the anomalous depletions 250 in various elements, including the REEs, displayed by this sample as the result of chemical alteration in 251 Antarctica (e.g., Lipschutz and Samuels, 1991). This sample also exhibits a positive Eu anomaly, as often 252 observed in heavily weathered Antarctic chondrites (e.g., Y-74014: Ebihara, 1986; Y-74492: Nishikawa et al., 1990). The characteristics of this sample are described in detail in subsection 4.1.2. 253

254 *3.2.1 Major elements*

The abundances of major elements for individual samples obtained using ICP-OES were determined with $\sim 2\%$ relative standard deviation (RSD; 1SD) on average, while the individual RSDs ranged from 0.05% to 8%. Although most of the mean values for major elemental abundances in Antarctic HCs are in good

- agreement with the literature values, the mean values of K, Ti, Fe, and Ni are ~15% lower than those reported
- in literature. However, the mean Ti abundance in Antarctic HCs is in excellent agreement with the published
- 260 Ti abundance in HCs determined using the isotope dilution method (0.0549 ± 0.0015 wt. % at the 95% CI;

calculated from Shima, 1979). According to these results, there is almost no difference in the abundances of

- 262 major elements between Antarctic and hot desert HCs, although the K and Ca abundances in hot desert HCs
- are ~20% higher than those in Antarctic HCs possibly due to either mobilization of these elements by
- 264 chemical alteration in Antarctica (Velbel *et al.* 1991) or terrestrial contamination in hot deserts (Al-Kathiri *et*
- 265 *al.*, 2005; Pourkhorsandi *et al.*, 2019).
- 266 *3.2.2 Trace elements*

267 The trace elemental abundances for individual samples obtained using Q-ICP-MS are reproducible within ~3% RSD (1SD) on average. The abundances of Lu and U in some samples display larger uncertainties, due 268 to their lower absolute abundances, on the order of ~10% RSD. The CI-normalized REE, Th, and U 269 270 abundances in each sample are shown in Fig. 2, and their mean REE, Th and U abundances in Antarctic 271 UHCs, EHCs, and HCs are compared with the literature values in Fig. 3 (CI values are from Anders and 272 Grevesse, 1989). No resolvable difference is observed between the trace elemental abundances of bulk 273 Antarctic UHCs and EHCs, which is consistent with the conclusions of Huss et al. (2006) and Hublet et al. 274 (2019). Most of the mean trace elemental abundances in Antarctic HCs, especially for REEs, Th, and U, are 275 in good agreement with the literature values for non-Antarctic HC listed in Table 4. The mean values for Sr 276 and Ba in Antarctic HCs deviate from the literature values (8.82 ± 0.31 ppm vs. 10 ppm and 3.22 ± 0.15 ppm vs. 4.20 ppm at the 95% CI, respectively). However, Sr and Ba are strongly affected by terrestrial weathering 277 278 as their abundances in the hot desert HCs characterized in this study are high (16.9-70.9 ppm and 17.8-166 279 ppm, respectively). Moreover, the mean values for Sr and Ba in Antarctic HCs are in agreement within 280 uncertainty with their published values for Antarctic HCs determined by the isotope dilution method (9.29 \pm 281 0.93 ppm and 2.69 ± 0.75 ppm at the 95% CI; Nishikawa *et al.*, 1990). Thus, the observed deviations may 282 only reflect a difference between Antarctic and non-Antarctic meteorites. For hot desert HCs, the mean values 283 of Cu, Zn, Rb, and Y agree within uncertainty with those for Antarctic HCs in this study and with the literature 284 values for non-Antarctic HC, with the exception of Rb. However, other trace elements are enriched compared with the values of Antarctic HCs and non-Antarctic HCs, especially in the case of Sr, Ba, the light REEs 285 (LREEs), Th, and U. In addition, negative Eu anomalies are observed in the CI-normalized REE patterns of 286 hot desert HCs (Fig. 2). 287

3.3 Isotope compositions

The Nd, Sm, Lu, and Hf abundances determined by the isotope dilution method, and Sm-Nd and Lu-Hf isotopic compositions in the bulk samples measured using MC-ICP-MS are summarized in Table 2 (for BHVO-2) and Table 4 (for meteorites) as well as in Figs. 4 and 5. The ¹⁴³Nd/¹⁴⁴Nd and ¹⁷⁶Hf/¹⁷⁷Hf values in most of the samples were determined within 20 ppm relative standard error (RSE) and 60 ppm RSE (2SE), respectively. This relatively large standard error for ¹⁷⁶Hf/¹⁷⁷Hf values compared with that for ¹⁴³Nd/¹⁴⁴Nd values clearly results from the difference in the adjusted concentrations in the measurement solutions (see subsection 2.3.2). Furthermore, the actual Hf concentration in the measurement solutions for most of the meteorite samples was less than 5 ppb, leading to larger standard errors for the ¹⁷⁶Hf/¹⁷⁷Hf values. On the other hand, the differences in these elemental abundances between those determined using the isotope dilution method (MC-ICP-MS) and calibration line method (Q-ICP-MS) are less than 5% on average, which is in excellent agreement.

300 The mean values of the elemental abundances determined using the isotope dilution method in Antarctic 301 HCs are in excellent agreement with those in HC falls (Table 4). Most of the Sm-Nd and Lu-Hf data for 302 Antarctic HCs are within the range of chondrite fall data and several of them plot on the respective chondrite 303 isochrons within uncertainty (for Sm-Nd data: Jacobsen and Wasserburg, 1980, 1984; Patchett et al., 2004; 304 Boyet and Carlson, 2005; Carlson et al., 2007; Bouvier et al., 2008, Gannoun et al., 2011; Burkhardt et al., 2016; Fukai and Yokoyama, 2017; for Lu-Hf data: Blichert-Toft and Albarède, 1997; Bizzarro et al., 2003; 305 Patchett et al., 2004; Bouvier et al., 2008; Dauphas and Pourmand, 2011; for chondrite isochrons: Bouvier 306 et al., 2008). The mean values of ¹⁴⁷Sm/¹⁴⁴Nd, ¹⁴³Nd/¹⁴⁴Nd, ¹⁷⁶Lu/¹⁷⁷Hf, and ¹⁷⁶Hf/¹⁷⁷Hf for Antarctic HCs 307 overlap with those of HC falls within uncertainties (Table 4). There are no resolvable differences in the mean 308 values of Antarctic UHC and EHC. In the case of hot desert HCs, the ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁴³Nd/¹⁴⁴Nd values 309 (0.1578-0.1866 and 0.512312-0.512540, respectively) are significantly lower than the mean values reported 310 311 in literature for HC falls (0.1953 ± 0.0006 and 0.512623 ± 0.000009 at 2SE, respectively; Table 4) and the mean values determined here for Antarctic HCs (0.1971 ± 0.0016 and 0.512633 ± 0.000044 at the 95% CI, 312 respectively; Table 4), plotting as far from the isochron as falling out of the range of chondrite fall data (Fig. 313 4). In contrast, it seems that the ¹⁷⁶Lu/¹⁷⁷Hf and ¹⁷⁶Hf/¹⁷⁷Hf values of hot desert HCs fall within the ranges 314 determined in literature and measured for Antarctic HCs (Table 4 and Fig. 5), except in the case of NWA 315 6771 (H4), which is characterized by an anomalously high Hf abundance (269 ppb). 316

317 4 DISCUSSION

318 4.1 Assessment of weathering effects on the Sm-Nd and Lu-Hf systems in Antarctic HCs

319 For the assessment of weathering effects on Antarctic meteorites, the classical ABC index system is 320 commonly used: A—minor rustiness (e.g., rust stains along fractures are minor); B—moderate rustiness (e.g., 7.5 to 35% of metal particles are weathered to limonite); C-severe rustiness (e.g., most metal particles are 321 322 weathered to limonite); and e-evaporite minerals are visible to the naked eye (Antarctic Meteorite 323 Newsletter, NASA; Meteorite Newsletter, NIPR). However, this index may not be sufficient as an indicator 324 especially in terms of chemical alteration (Koebel and Cassidy, 1991), as it is determined based on mineralogy. 325 Velbel (1988) and Nishikawa et al. (1990) found that bulk Rb abundances in some Antarctic ordinary 326 chondrites (OCs) and heavily weathered non-Antarctic HCs are anomalously low and do not co-vary with 327 the weathering index. Velbel (1988) concluded that Rb loss could be due to chemical leaching during the 328 residence of the meteorite in Antarctica, indicating that relative loss of Rb may be a better indicator of 329 chemical alteration than the weathering index. The results presented here display remarkable deviations in 330 the Rb abundances of both Antarctic and hot desert HCs relative to those reported in literature (Table 4). The 331 mean Rb abundance of Antarctic HCs is significantly lower than the literature value $(2.26 \pm 0.34 \text{ ppm vs}, 2.9 \text{ pm vs}, 2.9 \text{ pm vs})$

332 ppm at the 95% CI) with extremely low Rb abundances in A-880941 (H3.3) and A 09516 (H6), implying a 333 loss of Rb during Antarctic alteration. However, these lower Rb abundances may just reflect heterogeneity 334 in the amount of feldspar in the samples, which may be induced as the result of a sampling bias (*i.e.*, sampling 335 heterogeneity), as Rb is mostly distributed in feldspar (Shima and Honda, 1968; Mason and Graham, 1970). The average Na and Al abundances in the bulk samples are used as an indicator for the amount of feldspar in 336 337 the sample, and the respective Rb abundances normalized to the indicator are listed in Table 5. Even 338 considering the heterogeneous distribution of feldspar based on these normalized Rb abundances, the samples 339 displaying extremely low Rb abundances also show low normalized Rb abundances. Thus, any possible effect 340 on the bulk Rb abundance by sampling biases alone can be ruled out. The normalized Rb abundances can 341 also be compared with the normalized abundances of Sr, which is especially concentrated in feldspar (Shima 342 and Honda, 1968; Mason and Graham, 1970) but generally considered less mobile than Rb. The data from 343 this study are compared with calculated values for non-Antarctic HC (Table 4) and Antarctic HCs from 344 Nishikawa et al. (1990), in Fig. 6. The normalized Sr abundances in most of the samples are in good 345 agreement with the literature values, but in various samples the normalized Rb abundances are lower than 346 those reported in literature and display a wider range than the Sr abundances. A similar trend emerges when comparing the normalized abundance of Rb and that of Ba, which is concentrated in feldspar as well (Table 347 348 5). The low normalized Rb abundances (< 2.7) in some Antarctic HCs co-vary to some degree with their weathering grades, although exceptions exist. A-880941 (H3.3) is categorized as A/B but contains a low 349 350 normalized Rb abundance (1.77 ± 0.09) , which may reflect a difference in the weathering degree implied by 351 elemental ratios versus that inferred based on petrographic observation. In any case, the low Rb abundances 352 clearly reflect the loss of Rb during the terrestrial residence of the meteorites in Antarctica or elsewhere in 353 the world (Nishikawa et al., 1990), possibly because of chemical leaching. Therefore, the normalized Rb 354 abundances are taken here as a potential indicator for chemical alteration in the following discussions.

355 4.1.1 Weathering effects on bulk Sm-Nd and Lu-Hf systems

356 The Sm-Nd and Lu-Hf data of a few Antarctic HCs (e.g., A 09516) plot away from the respective chondrite 357 isochrons (Figs. 4 and 5). This may reflect an effect of weathering on the Sm-Nd and Lu-Hf systems in bulk chondrites in Antarctica. However, it remains unclear whether the parent isotopic ratios ¹⁴⁷Sm/¹⁴⁴Nd and 358 ¹⁷⁶Lu/¹⁷⁷Hf or the daughter isotopic ratios ¹⁴³Nd/¹⁴⁴Nd and ¹⁷⁶Hf/¹⁷⁷Hf have been affected by Antarctic 359 weathering. Alternatively, it is also possible that both ratios were disturbed. Therefore, we need to consider 360 361 one by one which of the following processes affected the Sm-Nd and Lu-Hf systems: i) sample heterogeneity, 362 ii) thermal metamorphism on the parent body(ies), iii) contamination with terrestrial material, and iv) chemical alteration during residence on the Earth. 363

i) Based on the results of this study and previous works, REEs in bulk chondrites are largely unfractionated (*e.g.*, Huss *et al.*, 2006). On the other hand, REEs in individual constituent minerals are highly fractionated depending on their partition coefficients (*e.g.*, Curtis and Schmitt, 1979). This fractionation of REEs between the constituent minerals affects not only their abundances but also their radiogenic isotope systems (*e.g.*, Jacobsen and Wasserburg, 1983). Thus, a loss of constituent mineral(s) due to terrestrial weathering, as observed in Antarctic eucrites (Mittlefehldt and Lindstrom, 1991), or a non-representative 370 sampling bias may induce minor variation in the elemental compositions and radiogenic isotope data of bulk

chondrites. According to Amelin and Rotenberg (2004), Amelin (2005), and Debaille *et al.* (2017), the Sm-

372 Nd and Lu-Hf data in individual minerals are indeed different from those in bulk chondrites. However, both

data of individual minerals and bulk chondrites plot on the same chondrite isochron when considering the uncertainties. Therefore, the heterogeneous distribution of constituent mineral(s) due to a sampling bias or terrestrial weathering, *i.e.*, sample heterogeneity may result in heterogeneity of the Sm-Nd and Lu-Hf data of bulk chondrites, but the isotopic data will always plot along the isochrons.

- 377 ii) Thermal metamorphism can affect the Sm-Nd and Lu-Hf systems at the bulk scale as a redistribution 378 due to crystallization of REE-rich minerals such as Ca-phosphates, can take place during metamorphism 379 (Bouvier et al., 2008; Martin et al., 2013; Debaille et al., 2017; Bloch et al., 2017, 2018). The nugget effect, 380 in which the REE-rich Ca-phosphates are heterogeneously distributed, can lead to sampling biases due to 381 non-representative sample volumes, while such Ca-phosphate phases may also be preferentially weathered 382 during the terrestrial residence of the meteorite of interest. These processes may produce wider ranges in the 383 Sm-Nd and Lu-Hf data in bulk equilibrated chondrites than in bulk unequilibrated chondrites (Bouvier et al., 384 2008; Dauphas and Pourmand, 2011, 2015). However, as mentioned above, unequilibrated and equilibrated 385 chondrites do not show any fractionation or change in the bulk REE abundances in most of the cases. If 386 unequilibrated and equilibrated chondrites had originally the exact same bulk elemental composition, the 387 bulk Sm-Nd and Lu-Hf data in these chondrites should exhibit the same values. This is consistent with the 388 observation that the mean Sm-Nd and Lu-Hf values in bulk unequilibrated and equilibrated chondrites are in 389 agreement with each other, both based on this study and literature values (Jacobsen and Wasserburg, 1980, 1984; Blichert-Toft and Albarède, 1997; Bizzarro et al., 2003; Patchett et al., 2004; Boyet and Carlson, 2005; 390 391 Carlson et al., 2007; Bouvier et al., 2008, Dauphas and Pourmand, 2011; Gannoun et al., 2011; Burkhardt et 392 al., 2016; Fukai and Yokoyama, 2017). Therefore, thermal metamorphism emphasizes the heterogeneity of 393 the Sm-Nd and Lu-Hf data in bulk chondrites at the scale of the individual sample. Theoretically, the isotopic 394 data in bulk unequilibrated and equilibrated chondrites should have the same values as long as these have the 395 same elemental compositions.
- iii) Considering the higher Nd, Sm, Lu, and Hf abundances and lower ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁷⁶Lu/¹⁷⁷Hf in the 396 397 upper continental crust (26 ppm, 4.5 ppm, 0.32 ppm, 5.8 ppm, 0.10, and 0.0079, respectively; Taylor and 398 McLennan, 1995) as potential contaminating material relative to the lower REE concentrations in chondrites, 399 the Sm-Nd and Lu-Hf data in bulk chondrites display lower values in most of the cases when terrestrial 400 contamination occurred (Bast et al., 2017a, b; Pourkhorsandi et al., 2017, 2021). As the hot desert HCs used 401 in this study exhibit enrichments in Sr, Ba, LREEs, Th, and U and negative Eu anomalies (Table 4 and Fig. 402 2), these enrichments must result from terrestrial contamination (e.g., Al-Kathiri et al., 2005; Pourkhorsandi et al., 2017). Thus, the low ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁴³Nd/¹⁴⁴Nd values in the hot desert HCs (Table 4 and Fig. 4) 403 404 must also be explained by mixing with terrestrial material. However, the absolute Lu abundance in the upper 405 continental crust is only ~ 10 times higher than that in HCs and Hf is highly immobile during terrestrial 406 alteration, while the absolute abundances of Nd and Sm in the upper continental crust are ~45 and ~25 times higher than those in HCs, respectively (Taylor and McLennan, 1995), and these elements are also considered 407 408 to be more mobile than Hf (Middelburg et al., 1988). In this way, because it is more difficult for the Lu-Hf

system to be affected by terrestrial contamination than the Sm-Nd system, the ¹⁷⁶Lu/¹⁷⁷Hf and ¹⁷⁶Hf/¹⁷⁷Hf 409 values in the hot desert HCs used in this study do not deviate as far from the isochron as observed for the 410 ¹⁴⁷Sm/¹⁴⁴Nd, ¹⁴³Nd/¹⁴⁴Nd values (Table 4 and Fig. 5). This is consistent with the observation of Al-Kathiri et 411 al. (2005) who showed that Hf abundances in HCs collected from hot deserts do not deviate from the literature 412 413 value listed in Table 4 (0.15-0.18 ppm vs. 0.18 ppm), while their corresponding Nd and Sm abundances are 414 \sim 10-30% higher than the literature values (0.66-0.75 ppm vs. 0.59 ppm and 0.21-0.24 ppm vs. 0.19 ppm, 415 respectively). Therefore, although the extent of terrestrial contamination on chondrites strongly depends on 416 where the sample was collected (Pourkhorsandi et al., 2017, 2021), this process can produce deviations from 417 the Sm-Nd and Lu-Hf isochrons in the case of chondrites (Bast et al., 2017b). These deviations should be 418 accompanied by additional indications of terrestrial contamination, such as higher abundances of Sr, Ba, 419 LREEs, Th, and U.

420 iv) Chemical alteration during terrestrial residence, such as leaching, occurs anywhere on Earth in the 421 presence of water, either in the form of rain or humidity. Even in Antarctica, melting of ice can trigger sample 422 alteration (Mittlefehldt and Lindstrom, 1991). Chemical alteration is commonly divided into two main types: 423 the first type encompasses alteration affecting minerals, such as the loss of Ca-phosphates (Mittlefehldt and 424 Lindstrom, 1991), while the second type reflects alteration that depends on the mobility of the element, as in 425 the case of Rb loss (Velbel, 1988; Nishikawa et al., 1990). For the former type of alteration, the effect on the 426 Sm-Nd and Lu-Hf systems is similar to the effect of sample heterogeneity. Thus, even if this alteration occurs, 427 the affected Sm-Nd and Lu-Hf data would plot on their respective isochrons assuming no prior disturbance. 428 The latter type of alteration can disturb these systems because the parent nuclide and the daughter nuclide 429 may have different chemical characteristics, which is consistent with the observations of Nishikawa et al. 430 (1990) although based on the Rb-Sr system. Thus, if this alteration occurred during the residence of the meteorite at the Earth's surface, the ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁷⁶Lu/¹⁷⁷Hf values would be disturbed producing 431 deviations from the Sm-Nd and Lu-Hf isochrons. This type of alteration does not influence the ¹⁴³Nd/¹⁴⁴Nd 432 and ¹⁷⁶Hf/¹⁷⁷Hf values at the bulk scale. 433

In summary, thermal metamorphism amplifies the nugget effect of REE-rich minerals such as Ca-434 435 phosphates on the Sm-Nd and Lu-Hf isotope systems. Sample heterogeneity also affects the Sm-Nd and Lu-436 Hf data in bulk chondrites, but the measured data continue to plot on the respective chondrite isochrons. In addition, ¹⁴³Nd/¹⁴⁴Nd and ¹⁷⁶Hf/¹⁷⁷Hf values are not affected by chemical alteration at the bulk scale or at 437 least such isotopic fractionation is not yet resolvable. Thus, the ¹⁴³Nd/¹⁴⁴Nd and ¹⁷⁶Hf/¹⁷⁷Hf values obtained 438 likely reflect the original values of the bulk HC sample, unless this sample was contaminated by terrestrial 439 440 material. Because the Asuka, Yamato, and Allan Hill samples used in this study are collected from blue ice fields, it is unlikely that the samples were contaminated during their residence in Antarctica. Therefore, the 441 deviations from the isochrons on the Sm-Nd and Lu-Hf data that are apparent in a small set of samples (Figs. 442 4 and 5) have likely been produced by disturbance of the ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁷⁶Lu/¹⁷⁷Hf values during the 443 444 chemical alteration that depends on the elemental mobility.

Based on the assumption above, to evaluate the effects of Antarctic weathering on the Sm-Nd and Lu-Hf data in bulk chondrites, the differences in ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁷⁶Lu/¹⁷⁷Hf values between the measured values and the respective chondrite isochrons are expressed as δ^{147} Sm/¹⁴⁴Nd (‰) and δ^{176} Lu/¹⁷⁷Hf (‰) respectively, 448 calculated following the equations (in the case of the Sm-Nd system):

449
$$\delta \frac{{}^{147}Sm}{{}^{144}Nd} = \left[\left(\frac{{}^{147}Sm}{{}^{144}Nd} \right)_{measured} - \left(\frac{{}^{147}Sm}{{}^{144}Nd} \right)_{ideal} \right] \times 1,000 \ [\%_0]$$

450 and

451
$$\left(\frac{^{147}Sm}{^{144}Nd}\right)_{ideal} = \frac{\left(\frac{^{143}Nd}{^{144}Nd}\right)_{measured} - b}{a}$$

where a is the slope of the isochron and b is the intercept of the isochron, which is the initial 143 Nd/ 144 Nd 452 value. The values for the isochrons and the initial isotope ratios in Sm-Nd and Lu-Hf systems are from 453 Bouvier et al. (2008). These calculated differences for Antarctic HCs reflect the extent of the disturbance on 454 the ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁷⁶Lu/¹⁷⁷Hf values by Antarctic alteration. The calculated differences are shown in Fig. 455 7, with a range of differences in chondrite falls since the Sm-Nd and Lu-Hf data in chondrite falls also show 456 a deviation from the isochron (Figs. 4 and 5). According to Fig. 7, the δ^{147} Sm/¹⁴⁴Nd values of Antarctic HCs, 457 except for A 09436 (H3)-2, ALH 78084 (H3.9), and A 09516 (H6), fall within the range of HC falls taking 458 into account the associated uncertainty. The δ^{176} Lu/¹⁷⁷Hf of Antarctic HCs, except for Y-793574 (H3.5), Y-459 790461 (H3.7), A 09387 (H4), and A 09618 (H5), also fall within the range of HC falls. Although the 460 δ^{147} Sm/¹⁴⁴Nd or δ^{176} Lu/¹⁷⁷Hf values of the samples excluded above do not fall within the range of HC falls, 461 the δ^{147} Sm/¹⁴⁴Nd and δ^{176} Lu/¹⁷⁷Hf values of all Antarctic HCs, except for A 09516 (H6)-1, do plot within the 462 range of other chondrite falls. The values of Antarctic HCs that do not fall within the range of HC falls could 463 indicate disturbance of the systems by chemical alteration in Antarctica. However, this disturbance can be 464 considered minor as most of the values fall within the range of other chondrite falls. The δ^{147} Sm/¹⁴⁴Nd and 465 δ^{176} Lu/¹⁷⁷Hf values in A-880941 (H3.3), which displays the lowest weathering index but the second highest 466 Rb loss in this study, fall close to 0. Similar δ^{147} Sm/¹⁴⁴Nd and δ^{176} Lu/¹⁷⁷Hf values are observed for heavily 467 weathered (sub)samples A 09618 (H5) and A 09436 (H3)-2 respectively, and for A-881258 (H3.0) that 468 indicates no Rb loss. Furthermore, δ^{147} Sm/¹⁴⁴Nd and δ^{176} Lu/¹⁷⁷Hf values of all published data for Antarctic 469 chondrites (Figs. 4 and 5), except for ALHA 77295 (EH3) with a high δ^{147} Sm/¹⁴⁴Nd (5.66‰), range from -470 471 1.96 to 1.94‰ and from -0.02 to 0.18‰, respectively, falling within the range of chondrite falls also 472 regardless of their weathering index. Therefore, it can be concluded that Antarctic chondrites display limited effects of weathering on their isotopic values, and the extent of the disturbance on ¹⁴⁷Sm/¹⁴⁴Nd and 473 ¹⁷⁶Lu/¹⁷⁷Hf in Antarctic chondrites would be of a level similar to that measured for chondrite falls even if 474 Antarctic weathering occurred. In addition, there seems to be no correlation between the δ^{147} Sm/¹⁴⁴Nd and 475 δ^{176} Lu/¹⁷⁷Hf values and the weathering indicators, suggesting that both the Sm-Nd and Lu-Hf systems are 476 477 fairly robust against Antarctic weathering relative to the Rb-Sr system (Nishikawa et al., 1990). However, 478 the most weathered sample in this study, as indicated by the weathering index C and the lowest normalized 479 Rb abundance (Table 5), A 09516 (H6), shows the largest absolute δ^{147} Sm/¹⁴⁴Nd values in this study and an atypical CI-normalized REE signature that is characterized by subchondritic values and a positive Eu 480 481 anomaly (Figs. 2 and 7). Therefore, the Sm-Nd and Lu-Hf systems are generally unaffected by Antarctic weathering except in the case of anomalous CI-normalized REE abundances, as all samples except A 09516 482 (H6) display normal chondritic REE patterns and low δ^{147} Sm/¹⁴⁴Nd and δ^{176} Lu/¹⁷⁷Hf values. 483

484 *4.1.2 Loss of REEs in A 09516 (H6)*

As described above, A 09516 clearly underwent a significant degree of alteration, both mineralogically and chemically. Since this sample was analyzed in duplicate and the low elemental abundances are observed for this sample twice, significant sample losses during acid decomposition can be ruled out. Moreover, no other sample analyzed in this study exhibits such low abundances. Given that the weathering mechanism that affected A 09516 appears considerably more complicated than that of any other sample, this H6 chondrite is excluded from the previous discussions and is discussed here separately.

491 The low P and REE abundances and the P/Mg of A 09516 relative to the other HCs (Tables 4 and 5) may 492 imply a loss of Ca-phosphates, known to take place during the alteration of eucrites in Antarctica (Mittlefehldt 493 and Lindstrom, 1991). However, the characteristic LREE depletion and Ce anomaly typical for alteration of 494 Antarctic eucrites are not observed A 09516 (Fig. 2). Considering the CI-normalized REE abundance in Ca-495 phosphates of HCs (Ebihara and Honda, 1983; Ward et al., 2017), HCs that have lost Ca-phosphates should 496 display lower CI-normalized LREE abundances relative to the heavy REE (HREE) abundances, which 497 indicates that the CI-normalized REE abundances in A 09516 cannot be explained only by a loss of Ca-498 phosphates, as is the case for the weathered Antarctic eucrites (Mittlefehldt and Lindstrom, 1991). Normal 499 chondritic Th and U abundances in A 09516 (Table 4 and Fig. 2) support that this sample did not lose 500 considerable Ca-phosphate, simply because Th and U are also concentrated in Ca-phosphates of OCs (Pellas 501 and Störzer, 1975; Crozaz, 1979). Generally, OCs, including HCs, contain chlorapatite and merrillite as Ca-502 phosphates (e.g., Van Schmus, 1969). The CI-normalized REE abundances in merrillite are relatively flat, 503 while those in chlorapatite are more enriched in LREEs than in HREEs in OCs (Crozaz et al., 1989; Ward et 504 al., 2017). In addition, the Th and U abundances in merrillite are considerably lower (~3 ppm and ~0.3 ppm, 505 respectively) than those in chlorapatite (~11 ppm and ~7.5 ppm, respectively) for OCs (Crozaz, 1974, 1979), 506 implying that a loss of merrillite from bulk OCs affects the bulk Th and U abundances significantly less than 507 a loss of chlorapatite. Thus, the REE abundances in A 09516 might be explained by the selective or more 508 rapid dissolution of merrillite rather than chlorapatite during Antarctic alteration. However, the solubilities 509 of merrillite and chlorapatite are comparable under acidic conditions (Adcock et al., 2013), and selective or 510 rapid dissolution of merrillite is hence unlikely. Therefore, a simple loss of Ca-phosphates by alteration 511 cannot explain the elemental composition of A 09516.

512 Alternatively, chemical leaching depending on the solubility of an element, for example taking place at 513 grain boundaries, could explain the elemental losses observed for A 09516. It is assumed that this mechanism 514 does not strongly affect the mineralogy of the OCs, as highly mobile elements are leached more during this 515 type of alteration. According to studies dealing with the mobility of REEs during weathering (Nesbitt, 1979; 516 Middelburg et al., 1988; van der Weijden and van der Weijden, 1995; Hannigan and Sholkovitz, 2001; Su et 517 al., 2017), the middle REEs (MREEs) and HREEs can more strongly be leached than the LREEs by alteration depending on the redox conditions, which could produce the observed REE abundances in A 09516. This can 518 519 leave immobile elements such as Hf and Th in this sample. Moreover, the solubility difference explains the 520 large deviation from the isochron on the Sm-Nd plots for A 09516 (Figs. 4 and 7) because any weathering 521 mechanism affecting the modal abundance of the constituent minerals, such as a loss of Ca-phosphates, could

- 522 not induce this divergence. However, the positive Eu anomaly in A 09516 cannot be produced by this leaching
- 523 mechanism because the sample would exhibit no or rather a negative Eu anomaly if leaching occurred
- 524 following their solubilities only. In addition, the Lu/Hf data in A 09516 display only a limited deviation
- 525 ($\delta Lu/Hf = -1.32 \pm 0.23$ ‰; Figs. 5 and 7) from the isochron while the Sm-Nd data show a larger deviation
- 526 $(\delta \text{Sm/Nd} = -5.58 \pm 1.40 \text{ }\% \text{ and } -10.5 \pm 1.4 \text{ }\%; \text{ Figs. 4 and 7}), \text{ which is inconsistent with their relative mobility.}$
- 527 Therefore, the chemical leaching depending on the solubility of an element can also not fully explain the
- 528 characteristics of A 09516.
- 529 Lastly, two speculative possibilities remain to explain the REE abundances in A 09516: either some Caphosphates were partially leached during Antarctic alteration, or some Ca-phosphates were completely 530 531 leached during Antarctic alteration, but the sample was at a later stage contaminated by terrestrial material. 532 Similar scenarios have been proposed by Bast et al. (2017b) to explain the observed discrepancies from the 533 reference isochron for bulk Lu-Hf data in eucrites and angrites. In the case of the former scenario, Ca-534 phosphates are fractured to some extent by an interruption in the ionic bonding between cations, especially impurity cations, and (PO₄)²⁻ in the Ca-phosphates as the result of Antarctic alteration, but the Ca-phosphates 535 are not completely broken due to insufficiencies in time, in the amount of fluid, or in the pH of the fluid 536 during the leaching. This leads to selective leaching of more soluble trace elements from Ca-phosphates. In 537 538 this case, the losses of MREEs and HREEs are more remarkable than those of LREEs, while the positive Eu 539 anomaly and the normal Th and U abundances in A 09516 remain. Moreover, this incomplete dissolution of 540 Ca-phosphates can produce the disturbance on the Sm-Nd plots for A 09516. However, the observation of a 541 lack of disturbance for the Lu/Hf in A 09516 still remains to be explained. In the case of the latter scenario, 542 contamination by terrestrial material during residence in Antarctica (Delisle et al., 1989; Welten et al., 2001) 543 or during the experimental procedure such as sample crushing cannot be ruled out completely, although the 544 contamination during Antarctic residence is unlikely because other samples, collected in the vicinity of A 545 09516, show no traces of contamination based on the results in this study. In both cases of contamination, 546 some Ca-phosphates are removed by leaching during Antarctic alteration entirely, but the abundances of 547 LREEs, Th, and U in A 09516 remain elevated because of terrestrial contamination. This can also explain the 548 disturbance of Sm/Nd, the presence of the positive Eu anomaly and even the absence of any disturbance of 549 the Lu/Hf because of less pronounced effects from terrestrial contamination on the Lu and Hf abundances. 550 However, the observation that the Th and U abundances are still chondritic limits the possible admixture of 551 terrestrial material. In any case, both possibilities including the loss of Ca-phosphates are inconsistent with a 552 high modal abundance of Ca-phosphates in this sample (0.75 wt. %; Table 3), although their mass scales need 553 to be considered. Therefore, A 09516 may have lost some amount of Ca-phosphates, but the way this HC lost 554 Ca-phosphates appears to be different from how Antarctic eucrites are affected (Mittlefehldt and Lindstrom, 555 1991), indicating that Antarctic alteration may be significantly more complicated than what is expected based 556 on heavily altered samples.

557 4.2 Control of heterogeneity on bulk Sm-Nd and Lu-Hf data by Ca-phosphates and Ca-pyroxene

558 The isotopic heterogeneity mostly depends on the sample heterogeneity unless the sample is affected by 559 terrestrial weathering, as discussed above. Thus, it is important to constrain which minerals mainly affect the

Sm-Nd and Lu-Hf data of bulk chondrites. To do this, elemental abundance ratios in Antarctic HCs were 560 561 compared with their isotope ratios, and their correlation coefficients r are shown in Table 6. If a constituent 562 mineral controls the heterogeneity in bulk Sm-Nd and Lu-Hf data, elements accommodated in the mineral 563 should display co-variation with the isotope data, if no selective removal or addition of elements occurred 564 during weathering. According to the correlation coefficients for all samples, P/Mg and Y/Mg display strong positive correlations with 147 Sm/ 144 Nd, 176 Lu/ 177 Hf, and 176 Hf/ 177 Hf (r = 0.5-0.8) and strong negative 565 correlations with ¹⁴³Nd/¹⁴⁴Nd (r = -0.6 and -0.7, respectively). If A 09516 (H6) is excluded, because its 566 ¹⁴⁷Sm/¹⁴⁴Nd is the most disturbed by Antarctic weathering among the samples studied in this work, the P/Mg 567 displays a strong negative correlation with 147 Sm/ 144 Nd instead of a positive correlation (r = -0.6; Table 6). 568 In the case of OCs, Y is distributed in Ca-phosphates (e.g., Mason, 1970), and Ca-phosphates have lower 569 ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁴³Nd/¹⁴⁴Nd values and higher ¹⁷⁶Lu/¹⁷⁷Hf and ¹⁷⁶Hf/¹⁷⁷Hf values compared to bulk samples 570 571 (Amelin and Rotenberg, 2004; Amelin, 2005; Debaille et al., 2017). Thus, these correlations indicate that the 572 isotope ratios in bulk samples highly depend on the Ca-phosphates-to-silicates weight ratio in the sample, 573 because the P/Mg and Y/Mg can be considered as indicators of the Ca-phosphates/silicates.

574 The correlation coefficients excluding A 09516 (H6) are shown in Table 6, not only because of the reasons 575 outlined above but also because this sample is characterized by an abnormally low abundance of Caphosphates as discussed in the subsection 4.1.2. When A 09516 (H6) is excluded, the P/Mg no longer 576 correlates with the ¹⁷⁶Lu/¹⁷⁷Hf and ¹⁷⁶Hf/¹⁷⁷Hf. Because the Y/Mg still correlates with these ratios and P is in 577 part distributed in metals especially in unequilibrated chondrites (Murrell and Burnett, 1983; Zanda et al., 578 579 1994), this may be caused by a disturbance of the bulk P abundance through the accommodation of P in metal. On the other hand, Al/Mg, Ca/Mg, and Ti/Mg display negative correlations with ¹⁷⁶Lu/¹⁷⁷Hf and ¹⁷⁶Hf/¹⁷⁷Hf 580 (r = -0.8 to -0.4), although the correlations with Al/Mg are relatively weak (r = -0.4 and -0.5, respectively). 581 582 According to *in-situ* analyses and Hf partition coefficients of minerals (Alexander, 1994; Righter and Shearer, 583 2003; Martin et al., 2013), Hf is distributed in chondrule glass and Ca-pyroxene in unequilibrated chondrites 584 and in Ca-pyroxene in equilibrated chondrites. Aluminum, Ca, and Ti are distributed in these silicate minerals 585 as well, although Al is mostly distributed in feldspar in EOCs (Curtis and Schmitt, 1979; Alexander, 1994). 586 Considering the REE distribution in EOCs, Lu is not only distributed in Ca-phosphates but also partitioned 587 to a large degree into Ca-pyroxene, while Sm and Nd are mostly distributed in Ca-phosphates (e.g., Curtis and Schmitt, 1979). In the case of UOCs, Lu is distributed in chondrule glass and Ca-pyroxene (Alexander, 588 589 1994; Shinotsuka, 1997). In addition, chondrule glass has the lowest Lu/Hf among silicates and Ca-590 phosphates for unequilibrated chondrites and Ca-pyroxene has the lowest Lu/Hf among these minerals for 591 equilibrated chondrites (Alexander, 1994; Martin et al., 2013). Therefore, the negative correlations of Al/Mg, Ca/Mg, and Ti/Mg with ¹⁷⁶Lu/¹⁷⁷Hf and ¹⁷⁶Hf/¹⁷⁷Hf indicate that the amount of Ca-pyroxene, which is a major 592 593 host phase of Hf and a partial carrier of Lu, affects the heterogeneity of bulk Lu-Hf data. Moreover, the 594 negative correlations are also produced by the heterogeneity in the amount of chondrule glass, which is a 595 major host phase of Lu and Hf in UHCs. The negative correlation between Ti/Mg and Lu-Hf data may also 596 be affected by the amount of ilmenite, which can have lower Lu/Hf than that in Ca-pyroxene according to 597 their partition coefficients (Green, 1994; Kleine et al., 2008). However, considering the budget of Hf between 598 constituent minerals in the whole rock, the correlation mostly depends on the weight ratio of Ca-pyroxene to

599 silicates (or Ca-pyroxene and chondrule glass to silicates).

- According to Table 6, P/Ca and Y/Ca exhibit negative correlations with Sm-Nd data and positive 600 correlations with Lu-Hf data, both when including and excluding A 09516 (H6), although ¹⁴⁷Sm/¹⁴⁴Nd still 601 displays positive correlations possibly because of the disturbance by weathering. Most of these correlations 602 603 are stronger than those for the P/Mg and Y/Mg. For OCs, REEs including Y are mostly distributed among 604 the two phases of Ca-phosphate and Ca-pyroxene (Ehihara and Honda, 1984). Among silicates and Ca-605 phosphates in chondrites, Ca-phosphates have the highest P/Ca, Y/Ca, and Lu/Hf and the lowest Sm/Nd (e.g., 606 Curtis and Schmitt, 1979; Amelin, 2005). In contrast, Ca-pyroxene has the lowest P/Ca, Lu/Hf, and Y/Ca and 607 the highest or second highest Sm/Nd (Curtis and Schmitt, 1979; Alexander, 1994; Martin et al., 2013). Thus, 608 Ca-phosphates and Ca-pyroxene are considered to be the endmembers of the bulk P/Ca, Y/Ca, Sm/Nd, and 609 Lu/Hf, indicating that the correlations of the P/Ca and Y/Ca reflect the Ca-phosphates-to-Ca-pyroxene weight 610 ratio. Therefore, the heterogeneity of Sm-Nd and Lu-Hf data in bulk OCs is mainly controlled by the weight
- 611 ratio of Ca-phosphates to Ca-pyroxene in the sample. For UOCs, the amount of chondrule glass also affects
- 612 the overall REE budget and isotopic values.

4.3 Relationship between the distributions of REEs and Hf and the heterogeneities of bulk Sm-Nd and Lu-Hf data

615 According to the result of the *in-situ* measurement in this study, the modal abundance of Ca-phosphates 616 in UHCs is significantly lower than that in EHCs (Fig. 1). This increase in the modal abundances of Ca-617 phosphates from UHCs to EHCs must result from thermal metamorphism on their parent body(ies) (Huss et 618 al., 2006). On the other hand, the bulk REE and P abundances overlap within uncertainty between UHCs and 619 EHCs, which is consistent with previous works (e.g., Jarosewich, 1990). Murrell and Burnett (1983) reported 620 that Ca-phosphate grains in UOCs are very fine (< 10 µm). Perron et al. (1992) found phosphate, chromite, 621 silica, and silica inclusions as secondary minerals in some UOCs. However, Semarkona (LL3.00), which is 622 one of the least metamorphosed chondrites, does not contain such grains. Based on these observations, 623 combining the bulk P abundances and the modal abundances of Ca-phosphates between UHCs and EHCs 624 used in this study, Ca-phosphates must have formed and grown in a closed system during thermal 625 metamorphism on the parent body or bodies. Ebihara (1989) and Shinotsuka (1997) analyzed REE 626 abundances in acid residues of OCs with lower and higher degrees of metamorphism, which were leached 627 using HCl or HNO₃. Because REEs are enriched in chondrule glass in UOCs (Alexander, 1994), Shinotsuka (1997) suggested that the REEs are re-distributed mainly from chondrule glass to Ca-phosphates during 628 629 thermal metamorphism. Since the bulk REE abundances are the same between UOCs and EOCs, the re-630 distribution of REEs may have occurred in a closed system with the growth of Ca-phosphates. This REE re-631 distribution in a closed system can explain the observation that the Sm-Nd and Lu-Hf data of each constituent 632 mineral in chondrites are plotted on the respective chondrite isochrons (Amelin and Rotenberg, 2004; Amelin, 633 2005; Debaille et al., 2017). In this way, because REEs are more distributed to Ca-phosphates in EOCs than 634 UOCs, the nugget effect of Ca-phosphates for EOCs produces larger isotopic variabilities of Sm-Nd and Lu-Hf systematics than that for UOCs, which reinforces the conclusions by Bouvier et al. (2008) and Dauphas 635 636 and Pourmand (2011, 2015). Moreover, sampling effects on bulk Lu-Hf data due to a heterogeneous

distribution of Ca-pyroxene may be larger in EOCs than in UOCs because Lu is hosted in Ca-pyroxene as 637 638 well as Ca-phosphates, while Hf seems to be re-distributed from chondrule glass to Ca-pyroxene based on 639 their initial distribution and their re-distribution after metamorphism (Curtis and Schmitt, 1979; Alexander, 640 1994; Righter and Shearer, 2003; Martin et al., 2013), although no data exist on the Hf distribution in OCs 641 and the mobility of Hf during thermal metamorphism. These larger contributions to bulk Sm-Nd and Lu-Hf 642 data by the sampling heterogeneities of Ca-phosphates and Ca-pyroxene in EOCs in terms of the distributions 643 of REE and Hf are complementary to the correlations between the elemental abundance ratios and the isotopic 644 ratios obtained in this study (Table 6). Therefore, unequilibrated chondrites in which the re-distributions have 645 not proceeded in the case of OCs should be used for the determination of precise average Sm-Nd and Lu-Hf 646 isotopic compositions to limit the effects of the sampling heterogeneities, reinforcing the conclusions by 647 Bouvier et al. (2008) and Debaille et al. (2017). Even in this case, the sampling heterogeneity of chondrule glass can still lead to heterogeneity in the bulk Sm-Nd and Lu-Hf data although the effect on the isotope data 648 649 by the heterogeneous distribution of chondrule glass appears to be less than that by the heterogeneous distribution of Ca-phosphates. Theoretically, equilibrated chondrites could also be useful for the 650 651 determination of precise average isotopic compositions as aimed for the CHUR values if a sufficiently large 652 amount of the sample is homogenized (speculatively more than a few grams at least), because the 653 heterogeneity of bulk Sm-Nd and Lu-Hf data only results from the sample heterogeneity, provided that re-654 distributions occurred in a closed system. Based on Patchett et al. (2004) and Bouvier et al. (2008), the mean values and uncertainties of bulk Sm-Nd data in equilibrated chondrites are almost the same as those in 655 unequilibrated chondrites when a few grams of equilibrated chondrite are homogenized $(^{147}Sm/^{144}Nd =$ 656 0.1960 ± 0.0006 and 0.1960 ± 0.0004 ; ¹⁴³Nd/¹⁴⁴Nd = 0.512634 ± 0.000012 and 0.512630 ± 0.000011 , 657 658 respectively) On the other hand, the homogenization with even a few grams of equilibrated chondrite seems 659 to be insufficient for the determination of bulk average Lu-Hf isotopic compositions for the purpose of CHUR determination ($^{176}Lu/^{177}Hf = 0.0324 \pm 0.0009$ and 0.0336 ± 0.0001 ; $^{176}Hf/^{177}Hf = 0.282693 \pm 0.000078$ and 660 661 0.282785 ± 0.000011 , the same order as above), as also concluded by Patchett *et al.* (2004). Thus, equilibrated 662 chondrites may provide representative Sm-Nd isotopic compositions in individual samples as long as more 663 than a few grams of the sample is homogenized to remove the nugget effect, and such data would be useful 664 for the determination of precise average Sm-Nd isotopic compositions for individual chondrite groups. However, because of the uncertainty related to the essential mass to be actually representative, only 665 unequilibrated chondrites should be used for the determination of the precise Lu-Hf CHUR values, probably 666 667 applied to the determination of precise average Sm-Nd isotopic compositions for individual chondrite groups 668 as well. This difference in sufficient sample amounts for homogenization may be because the variation in Sm-Nd data largely depends on the sampling heterogeneity of Ca-phosphates, while the variation in Lu-Hf 669 670 data depends not only on the sampling heterogeneity of Ca-phosphates but also on that of Ca-pyroxene and 671 chondrule glass.

The distributions of REEs and Hf and the redistribution of these elements during parent body processes differ between the various classes of chondrites as the major hosts of those elements and their parent body processes vary, as shown in Table 7, although the distribution and the mobility of Hf remain poorly understood. Sampling biases involving these major hosts are largely responsible for the observed variations

in Sm-Nd and Lu-Hf isotopic ratios. For example, CV and CO chondrites contain abundant Ca, Al-rich 676 inclusions (CAIs; 2.98 vol.% and 0.99 vol.%, respectively; Hezel et al., 2008), producing variable REE 677 abundances and probably variable Hf abundance as well, as the result of a heterogeneous distribution of CAIs 678 679 in samples of these meteorites. However, CI-normalized abundances of REEs in bulk CAIs are only $\sim 20 \times CI$ (e.g., El Goresy et al., 2002), while bulk oldhamite in enstatite chondrites and bulk Ca-phosphates in R 680 681 chondrites contain ~100×CI and ~200×CI of REEs, respectively (Laimer and Ganapathy, 1987; Maeda et al., 682 2017a). Moreover, other refractory components such as amoeboid olivine aggregates contain much less REE 683 and Hf abundances (Krot et al., 2004). Thus, based on mass balance considerations, the nugget effects of 684 CAIs or other refractory components on the Sm-Nd and Lu-Hf isotopic heterogeneities are not as much as 685 those of Ca-phosphates or oldhamite. In the cases of enstatite and Rumuruti chondrites, the modal abundances 686 of the REE host phases are only less than 2 wt. % and about 0.4 wt. %, respectively (Crozaz and Lundberg, 687 1995; Bischoff et al., 2011). Moreover, Barrat et al. (2014) and Maeda et al. (2017b) concluded that the REEs 688 of both chondrite types were re-distributed between the respective host phases during thermal metamorphism. 689 Thus, similar to OCs, the unequilibrated counterparts of these other chondrite classes should be preferred to 690 determine the precise average Sm-Nd and Lu-Hf isotopic compositions for enstatite and Rumuruti chondrites 691 to avoid strong nugget effects of the respective host phases. This may also apply to CK chondrites because 692 these experienced thermal metamorphism and contain < 1 vol. % of Ca-phosphates as one of the hosts of 693 REEs (Martin et al., 2013). Kakangari, CV, and CO chondrites underwent aqueous alteration but 694 simultaneously also underwent a limited degree of thermal metamorphism on their parent bodies. However, 695 their thermal metamorphism occurred within the unequilibrated range, implying the effect of elemental re-696 distribution by thermal metamorphism is almost negligible. For many groups of carbonaceous chondrites that 697 underwent aqueous alteration, it is unclear whether heavily altered samples can be used for the determination 698 of the precise average isotopic compositions because the redistribution of REEs and Hf during aqueous 699 alteration remains poorly constrained. According to Bouvier et al. (2008), aqueously altered samples produce 700 fewer heterogeneities of bulk Sm-Nd and Lu-Hf data than metamorphosed samples. Therefore, the use of 701 unequilibrated chondrites not only for OCs but also for most other classes of chondrites can reduce the effect 702 of variability in the isotope data stemming from the sample heterogeneity, which is required for the 703 determination of precise average Sm-Nd and Lu-Hf isotopic compositions for individual chondrite groups.

704 5 CONCLUSIONS

In this study, we analyzed ten Antarctic HCs and three HCs from hot deserts in a systematic manner to determine their modal mineral abundances, major and trace elemental abundances, and Sm-Nd and Lu-Hf isotopic compositions. The following conclusions can be drawn.

Firstly, by calculating the Rb abundances normalized to the average Na and Al abundances for each sample to evaluate Antarctic chemical alteration, our results indicate that not all weathered samples categorized as C according to the weathering index display a loss of Rb. Conversely, samples categorized as A/B may exhibit a high Rb loss. Thus, the normalized Rb abundance can be a good indicator for the assessment of Antarctic alteration chemically but only indicates the relative intensity of the chemical 713 weathering in Antarctica.

714 Secondly, the effects of Antarctic alteration on the Sm-Nd and Lu-Hf systems in bulk HCs are evaluated using the δ^{147} Sm/¹⁴³Nd and δ^{176} Lu/¹⁷⁷Hf notations. These values do not correlate with the weathering index 715 or the Rb indicator, except in the case of a single heavily weathered sample A 09516 (H6), which displays 716 717 various element losses, including for the REEs. This result suggests that the Sm-Nd and Lu-Hf systems 718 preserve their original compositions during Antarctic alteration. However, when a sample is heavily affected 719 by Antarctic weathering and has lost many elements, the isotope systems may also be disturbed. Such high 720 elemental losses might reflect partial dissolution of Ca-phosphates during alteration. Therefore, unless the 721 sample is clearly heavily weathered, most Antarctic meteorites, including rare meteorites such as ungrouped, 722 shergottites-nakhlites-chassignites, and lunar meteorites can be used for the investigation of their Sm-Nd and 723 Lu-Hf isotopic compositions.

724 Finally, we confirm that the Sm-Nd and Lu-Hf data in bulk HCs are correlated with the weight ratio of 725 Ca-phosphates to silicates. The weight ratio of Ca-phosphates to Ca-pyroxene may correlate stronger with the isotopic ratios, especially in the case of the Lu-Hf system. Sample heterogeneity of Ca-phosphates in HCs 726 727 may thus lead to heterogeneities in the bulk Sm-Nd and Lu-Hf data. Since REEs in OCs are re-distributed to 728 Ca-phosphates in a closed system during thermal metamorphism on the parent body(ies), the nugget effect 729 of Ca-phosphates on the heterogeneities of bulk Sm-Nd and Lu-Hf isotope data is larger for EOCs than for 730 UOCs. Therefore, the use of UOCs should be preferred for the determination of well-constrained average 731 Sm-Nd and Lu-Hf isotopic compositions such as CHUR values unless at least a few grams of EOCs are used, 732 which reinforces the conclusion by Bouvier et al. (2008). The preferred use of unequilibrated specimens may 733 also be extended to the other classes of chondrites, but the optimal sample amount for homogenization to 734 obtain representative Sm-Nd and Lu-Hf isotopic compositions in a sample is expected to differ for each class 735 because the host phases of REEs and Hf and their remobilization during parent body processes are distinct. 736 For the analysis of bulk isotope data, major and trace elemental abundances should be determined together 737 with the isotope data in order to evaluate the sample heterogeneity and to assess the effects of weathering. A better understanding of the Hf distribution on the bulk meteorite scale, the Hf mobilization during thermal 738 739 metamorphism, and the redistribution of REEs and Hf during aqueous alteration on meteorite parent bodies 740 may help to determine the sample sizes required to overcome sample heterogeneity and to measure 741 representative Sm-Nd and Lu-Hf isotopic compositions in individual samples, leading to precise average Sm-742 Nd and Lu-Hf isotopic compositions for individual chondrite groups.

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1003 TABLE CAPTIONS

- 1004 **Table 1.** List of H chondrites analyzed in this study.
- 1005 * The shock stages are estimated from descriptions of the shock degree in Meteorite Newsletter, NIPR,
- 1006 based on Stöffler *et al.* (1991).

Mataouita	Sectio	on Number	Type ^{a, b}	Weathering ^{a, b}	Shock	Common
Meteorite	PTS	Powder	Type	index	stage	Sources
A-881258	71 - A	82	H3.0	В	?	NIPR
A-880941	51-A	48	H3.3	A/B	S1-2*	NIPR
Y-793574	51-A	56	H3.5	?	?	NIPR
Y-790461	91-A	100	H3.7	В	S3*	NIPR
ALH 78084	81 - A	89	H3.9	B/Ce	?	NIPR
A 09436	-	-	H3	С	?	RBINS
A 09387	-	-	H4	B/C	?	RBINS
A 09618	-	-	H5	С	?	RBINS
A 09516	-	-	H6	С	?	RBINS
Y-790960	63-A	62	H7	В	S3*	NIPR
NWA 6752	-	-	H3	W2	S 1	RBINS
NWA 6771	-	-	H4	W1	S 1	RBINS
Sahara 97035	-	-	H5	W2	S1	RBINS

^a Meteorite Newsletter, NIPR. ^b Antarctic Meteorite Newsletter, NASA.

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- 1010 analyzed).
- 1011 The uncertainties of elemental abundances (1SD), ¹⁴³Nd/¹⁴⁴Nd, and ¹⁷⁶Hf/¹⁷⁷Hf (2SE) reflect internal
- 1012 reproducibilities during the measurements by ICP-OES, Q-ICP-MS, and MC-ICP-MS. The uncertainties of
- 1013 the elemental abundances (ID), ¹⁴⁷Sm/¹⁴⁴Nd, and ¹⁷⁶Lu/¹⁷⁷Hf (2SE) reflect the maximum uncertainties due to
- 1014 spike calibrations (see subsection 2.3.2).
- 1015 * The uncertainty of the ¹⁷⁶Hf/¹⁷⁷Hf value (2SE) reflects external reproducibilities during the measurements
 1016 of MC-ICP-MS.
- ^a Jochum *et al.* (2016) for elemental abundances including those determined by isotope dilution, ¹⁴⁷Sm/¹⁴⁴Nd,
- 1018 and ¹⁷⁶Lu/¹⁷⁷Hf. ^b Weis *et al.* (2006) for ¹⁴³Nd/¹⁴⁴Nd. ^c Weis *et al.* (2007) for ¹⁷⁶Hf/¹⁷⁷Hf.

¹⁰⁰⁹ Table 2. Elemental abundances and isotope compositions in BHVO-2 (ID: isotope dilution, n.a.: not

	Th	is work	Preferred values ^{a-c}		
(%)					
Na	1.79 ± 0.00	1.59 ± 0.02	1.646 ± 0.036		
Mg	4.74 ± 0.03	4.44 ± 0.02	4.376 ± 0.025		
Al	7.79 ± 0.05	6.87 ± 0.03	7.113 ± 0.032		
Р	0.115 ± 0.002	0.114 ± 0.003	0.1172 ± 0.0022		
K	0.439 ± 0.005	0.416 ± 0.001	0.4259 ± 0.0031		
Ca	8.15 ± 0.02	7.66 ± 0.03	8.148 ± 0.043		
Ti	1.68 ± 0.01	1.57 ± 0.00	1.637 ± 0.011		
Cr	0.0310 ± 0.0008	0.0335 ± 0.0004	0.02872 ± 0.00031		
Mn	0.129 ± 0.002	0.130 ± 0.002	0.1309 ± 0.0015		
Fe	8.73 ± 0.01	8.30 ± 0.01	8.666 ± 0.063		
(ppm)					
Ni	127 ± 31	127 ± 4	119.8 ± 1.2		
Co	80.8 ± 1.7	76.5 ± 1.9	44.89 ± 0.32		
Cu	123 ± 1	119 ± 1	129.3 ± 1.4		
Zn	97.9 ± 0.5	123 ± 1	103.9 ± 1.0		
Rb	9.91 ± 0.10	8.24 ± 0.19	9.261 ± 0.096		
Sr	370 ± 2	354 ± 5	394.1 ± 1.7		
Y	25.7 ± 0.1	24.3 ± 0.2	25.91 ± 0.28		
Zr	165 ± 1	157 ± 1	171.2 ± 1.3		
Nb	16.9 ± 0.1	16.4 ± 0.1	18.10 ± 0.20		
Ba	126 ± 1	121 ± 2	130.9 ± 1.0		
La	15.7 ± 0.2	15.2 ± 0.2	15.20 ± 0.08		
Ce	37.7 ± 0.4	36.5 ± 0.5	37.53 ± 0.19		
Pr	5.20 ± 0.03	5.08 ± 0.08	5.339 ± 0.028		
Nd	24.7 ± 0.2	23.7 ± 0.4	24.27 ± 0.25		
Sm	6.01 ± 0.13	5.75 ± 0.1	6.023 ± 0.057		
Eu	2.02 ± 0.03	1.98 ± 0.03	2.043 ± 0.012		
Gd	6.47 ± 0.11	6.18 ± 0.12	$\boldsymbol{6.207 \pm 0.038}$		
Tb	0.914 ± 0.012	0.876 ± 0.020	0.9392 ± 0.0053		
Dy	5.29 ± 0.05	5.11 ± 0.08	5.280 ± 0.028		
Но	0.980 ± 0.015	0.940 ± 0.020	0.9887 ± 0.0053		
Er	2.57 ± 0.03	2.43 ± 0.04	2.511 ± 0.014		
Tm	0.335 ± 0.004	0.315 ± 0.012	0.3349 ± 0.0031		
Yb	2.00 ± 0.06	1.93 ± 0.01	1.994 ± 0.027		
Lu	0.278 ± 0.005	0.256 ± 0.008	0.2754 ± 0.0024		
Hf	4.40 ± 0.04	4.06 ± 0.12	4.470 ± 0.025		
Th	1.38 ± 0.06	1.14 ± 0.05	1.224 ± 0.016		
U	0.412 ± 0.007	0.377 ± 0.012	0.412 ± 0.035		
Nd (ID)		22.5 ± 0.1	24.27 ± 0.18		
Sm (ID)		5.63 ± 0.03	6.023 ± 0.038		
Lu (ID)	n.a.	0.249 ± 0.001	0.2754 ± 0.0018		
Hf (ID)		4.04 ± 0.02	4.470 ± 0.020		

1020 **Table 3.** Modal abundances in H chondrites: vol. % at left; wt. % at right (UHC: unequilibrated H chondrite, EHC: equilibrated H chondrite).

- 1021 Wt. % is calculated from vol. % using the following estimated mineral specific gravities and normalized to 100%: olivine (3.48), low-Ca pyroxene (3.2). Ca-pyroxene (3.4),
- 1022 feldspar (2.62), kamacite (7.9), taenite (8.01), troilite (4.61), chlorapatite (3.16), merrillite (3.12), chromite (4.6).
- 1023 The uncertainties for the mean values from this study are 95% confidence interval, and those for McSween *et al.*, (1991) are 2SE calculated from their data.
- 1024 * The modal abundances of low-Ca pyroxene, feldspar, and chromite obtained in this study may include forsterite, silica, and ilmenite, respectively (see section 3.1).

Name	A-88	31258	A-88	30941	Y-79	3574	Y-79	0461	ALH	78084	A 09	9436	A 09	9387	A 09	9618	A 09	9516	Y-79	0960
Area (mm ²)	71.	294	52.	706	61.	594	104	.907	81.	524	190	.921	89.	756	152	.775	177	.284	34.	607
Olivine	40.7	38.9	36.8	33.4	34.4	32.2	38.0	35.9	35.8	33.5	35.4	33.1	36.8	35.4	38.0	35.6	36.8	33.4	38.7	36.0
Low-Ca pyroxene*	30.7	27	24.1	20	27.4	24	26.8	23	29.7	26	32.8	28	29.2	26	30.6	26	30.8	26	29.0	25
Ca-pyroxene	1.15	1.1	6.26	5.6	7.88	7.2	5.53	5.1	5.51	5.0	4.52	4.1	4.86	4.6	2.26	2.1	3.94	3.5	5.02	4.6
Feldspar*	14.8	10.7	14.5	9.91	14.6	10.3	14.7	10.5	14.1	9.91	13.1	9.22	14.3	10.3	13.6	9.58	10.9	7.45	14.0	9.77
Kamacite	6.01	13	3.52	7.2	5.76	12	6.05	13	6.64	14	6.09	13	4.81	10	7.23	15	7.35	15	6.46	14
Taenite	1.59	3.49	8.85	18.5	3.49	7.51	2.67	5.82	2.86	6.17	3.15	6.76	2.91	6.44	2.06	4.45	4.40	9.19	2.59	5.54
Troilite	3.59	4.55	3.12	3.75	4.33	5.36	3.93	4.92	3.47	4.30	3.74	4.62	4.04	5.15	3.97	4.94	3.50	4.20	3.79	4.67
Chlorapatite	0.05	0.04	0.09	0.07	0.11	0.10	0.05	0.04	0.29	0.25	0.16	0.14	0.03	0.02	0.30	0.26	0.14	0.11	0.35	0.30
Merrillite	0.47	0.41	0.37	0.30	0.38	0.32	0.41	0.35	0.32	0.27	0.36	0.30	0.90	0.78	0.33	0.28	0.78	0.64	0.43	0.36
Total Ca-phosphates	0.52	0.45	0.46	0.38	0.49	0.41	0.46	0.39	0.61	0.51	0.52	0.44	0.93	0.80	0.63	0.53	0.92	0.75	0.78	0.66
Chromite*	0.59	0.74	0.97	1.2	1.05	1.3	0.81	1.0	0.72	0.89	0.60	0.74	0.77	0.97	0.79	0.98	0.63	0.76	0.28	0.35
Iron oxide		-	tra	ace	tra	ice		-	tra	ace	tra	ice		-		-		-		-
Silica		-		-		-	tra	ice		-		-	tra	ice		-		-		-
(Mg, Fe)-spinel		-	tra	ace		-	tra	ace		-		-		-		-		-		-
Hibonite		-		-		-	tra	ace		-		-		-		-		-		-
(Zn, Fe)S		-		-		-		-		-	tra	ace	tra	ice		-		-		-

Total	99.6	98.6	99.3	99.0	99.3	100.0	98.6	99.0	99.3	100.7	
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1026 (continued)

Name	NWA	6752	NWA	6771	Sahara	97035	UHC mean	(this study)	EHC mean	(this study)	EHC mean (1	literature data; in wt. %)
Area (mm ²)/ n / Ref.	496	.441	94.	315	100	.041	n =	= 7	n =	= 6	Van Schmus (1969)	McSween <i>et al.</i> (1991), n = 25
Olivine	38.0	34.5	39.3	36.4	35.3	33.7	37.0 ± 1.9	34.5 ± 2.4	37.5 ± 1.5	35.1 ± 1.3	33 - 37	35.7 ± 1.5
Low-Ca pyroxene*	31.2	26	26.4	22	30.1	26	29.0 ± 2.8	25 ± 3	29.4 ± 1.7	25 ±2	23 - 27	26.3 ± 1.2
Ca-pyroxene	2.02	1.8	5.55	5.0	5.50	5.1	4.70 ± 2.19	4.3 ± 2.0	4.52 ± 1.31	4.1 ± 1.2	4 - 5	4.1 ± 0.2
Feldspar*	9.00	6.15	12.1	8.40	13.8	9.89	13.5 ± 1.9	9.52 ± 0.58	13.1 ± 1.4	9.24 ± 1.14	9 - 10	9.6 ± 0.2
Kamacite	7.30	15	7.21	15	6.88	15	5.91 ± 1.09	13 ± 3	6.65 ± 1.01	14 ± 2	15 - 17	19.2 + 0.6
Taenite	4.54	9.48	2.88	6.13	1.51	3.32	3.88 ± 2.19	8.25 ± 4.64	2.72 ± 1.03	5.84 ± 2.10	2 - 3	18.2 ± 0.6
Troilite	4.88	5.86	3.95	4.85	4.06	5.14	3.86 ± 0.54	4.77 ± 0.47	3.89 ± 0.22	4.82 ± 0.37	5 - 6	5.5 ± 0.2
Chlorapatite	0.06	0.05	0.42	0.35	tra	ace	0.12 ± 0.08	0.10 ± 0.07	0.21 ± 0.17	0.17 ± 0.14	-	-
Merrillite	0.14	0.12	0.41	0.34	0.89	0.76	0.35 ± 0.10	0.29 ± 0.08	0.62 ± 0.27	0.53 ± 0.24	-	-
Total Ca-phosphates	0.20	0.17	0.83	0.69	0.89	0.76	0.47 ± 0.12	0.39 ± 0.09	0.83 ± 0.12	0.70 ± 0.10	0.6	0.65 ± 0.03
Chromite*	0.83	0.99	0.80	0.98	0.48	0.60	0.79 ± 0.16	0.98 ± 0.20	0.63 ± 0.22	0.77 ± 0.27	0.5	0.76 ± 0.02
Iron oxide	tra	ice		-		-						
Silica		-		-		-						
(Mg, Fe)-spinel		-		-		-						
Hibonite		-		-		-						
(Zn, Fe)S		-		-		-						
Total	98.0		98.9		98.5							

- Table 4. Elemental abundances and isotope compositions in H chondrites (UHC: unequilibrated H chondrite,
 EHC: equilibrated H chondrite, ID: isotope dilution, n.d.: not determined).
- 1030 The uncertainties for each sample reflect the same as those in Table 2 and the number of digits is based on
- 1031 the reproducibility of the reference material. The uncertainties for the mean values are 95% confidence
- 1032 interval. The elemental abundances (ID) and the isotope ratios for non-Antarctic HC are mean values of H
- 1033 chondrite falls from the following literatures (see Supplementary data).
- 1034 * The data of A 09516 (H6) are excluded from the means (see section 3.2).
- ^a Jarosewich (1990) for Na-Ni. ^b Wasson and Kallemeyn (1988) for Cu-Ba and Hf. ^c Nakamura (1974) for
- 1036 REEs. ^d Tatsumoto *et al.* (1973) for Th and U. ^e Jacobsen and Wasserburg (1980, 1984), Patchett *et al.*
- 1037 (2004), Boyet and Carlson (2005), Carlson et al. (2007), Bouvier et al. (2008), Gannoun et al. (2011),
- 1038 Burkhardt et al. (2016), and Fukai and Yokoyama (2017) for Nd and Sm isotope compositions. ^f Blichert-
- 1039 Toft and Albarède (1997), Bizzarro et al. (2003), Patchett et al. (2004), Bouvier et al. (2008), and Dauphas
- and Pourmand (2011) for Lu and Hf isotope compositions.
- 1041

Name	A-881258	A-880941	Y-793574	Y-790461	ALH 78084
(%)					
Na	0.56 ± 0.03	0.54 ± 0.04	0.68 ± 0.04	0.61 ± 0.01	0.62 ± 0.03
Mg	14 ± 0	13 ± 0	15 ± 0	14 ± 0	14 ± 0
Al	0.99 ± 0.00	0.92 ± 0.00	1.1 ± 0.0	1.1 ± 0.0	1.0 ± 0.00
Р	0.11 ± 0.00	0.11 ± 0.00	0.13 ± 0.00	0.12 ± 0.00	0.10 ± 0.00
K	0.065 ± 0.001	0.050 ± 0.001	0.071 ± 0.002	0.065 ± 0.002	0.065 ± 0.001
Ca	0.98 ± 0.00	0.94 ± 0.01	1.2 ± 0.0	1.0 ± 0.0	1.0 ± 0.0
Ti	0.057 ± 0.01	0.050 ± 0.002	0.061 ± 0.002	0.062 ± 0.001	0.055 ± 0.000
Cr	0.38 ± 0.00	0.34 ± 0.00	0.41 ± 0.00	0.40 ± 0.00	0.35 ± 0.00
Mn	0.23 ± 0.00	0.22 ± 0.00	0.25 ± 0.00	0.23 ± 0.00	0.23 ± 0.00
Fe	23 ± 1	22 ± 0	27 ± 0	23 ± 0	22 ± 0
Ni	1.6 ± 0.0	1.6 ± 0.0	1.6 ± 0.1	1.5 ± 0	1.5 ± 0.0
Co	0.064 ± 0.001	0.066 ± 0.000	0.085 ± 0.001	0.060 ± 0.000	0.065 ± 0.01
(ppm)					
Cu	88.0 ± 0.6	80.0 ± 1.6	78.0 ± 1.4	90.5 ± 1.8	75.2 ± 1.7
Zn	63.9 ± 0.8	52.1 ± 1.3	61.4 ± 0.9	61.2 ± 0.7	54.8 ± 1.3
Rb	2.45 ± 0.08	1.29 ± 0.05	2.29 ± 0.05	2.67 ± 0.07	2.26 ± 0.08
Sr	8.77 ± 0.13	8.76 ± 0.14	8.53 ± 0.14	9.42 ± 0.12	8.66 ± 0.23
Y	1.90 ± 0.02	1.97 ± 0.06	1.87 ± 0.04	1.98 ± 0.05	2.08 ± 0.05
Zr	4.78 ± 0.08	4.79 ± 0.10	4.69 ± 0.08	5.28 ± 0.09	5.31 ± 0.02
Nb	0.358 ± 0.014	0.356 ± 0.008	0.375 ± 0.007	0.370 ± 0.015	0.382 ± 0.008
Ba	3.19 ± 0.07	3.17 ± 0.07	3.06 ± 0.01	3.38 ± 0.02	3.18 ± 0.02

(ppb)					
La	299 ± 10	330 ± 8	324 ± 4	333 ± 10	317 ± 15
Ce	788 ± 19	833 ± 24	830 ± 21	850 ± 22	820 ± 40
Pr	116 ± 4	121 ± 8	119 ± 2	125 ± 4	113 ± 2
Nd	549 ± 19	602 ± 53	618 ± 14	581 ± 42	615 ± 60
Sm	177 ± 15	216 ± 12	181 ± 6	208 ± 12	188 ± 18
Eu	72.9 ± 3.4	73.8 ± 4.8	74.3 ± 3.0	76.6 ± 5.4	65.3 ± 4.8
Gd	257 ± 15	283 ± 17	292 ± 8	289 ± 19	285 ± 17
Tb	45.1 ± 2.7	47.2 ± 3.2	49.8 ± 0.9	46.9 ± 2.6	47.5 ± 2.5
Dy	324 ± 8	335 ± 11	337 ± 17	338 ± 20	365 ± 15
Но	67.5 ± 3.0	75.0 ± 2.7	75.8 ± 1.5	74.3 ± 2.6	77.6 ± 2.3
Er	206 ± 3	215 ± 9	226 ± 7	232 ± 3	229 ± 4
Tm	30.2 ± 0.7	32.1 ± 1.7	34.6 ± 2.0	32.3 ± 1.4	31.5 ± 1.6
Yb	210 ± 12	210 ± 5	226 ± 8	211 ± 13	220 ± 9
Lu	30.1 ± 3.1	31.7 ± 4.3	34.3 ± 2.1	32.8 ± 2.7	30.0 ± 0.6
Hf	123 ± 9	135 ± 3	140 ± 10	147 ± 14	149 ± 7
Th	42.7 ± 1.4	38.1 ± 0.9	42.4 ± 0.6	43.0 ± 2.5	39.7 ± 1.9
U	11.4 ± 1.1	9.99 ± 0.64	12.4 ± 0.4	10.8 ± 0.5	12.0 ± 0.7
Nd (ID)	585 ± 3	609 ± 3	527 ± 3	604 ± 3	543 ± 3
Sm (ID)	190 ± 1	196 ± 1	172 ± 1	197 ± 1	178 ± 1
Lu (ID)	28.9 ± 0.1	29.7 ± 0.1	32.4 ± 0.2	29.2 ± 0.1	31.0 ± 0.2
Hf (ID)	123 ± 1	125 ± 1	152 ± 1	136 ± 1	140 ± 1
¹⁴⁷ Sm/ ¹⁴⁴ Nd	0.1958 ± 0.0014	0.1949 ± 0.0014	0.1977 ± 0.0014	0.1971 ± 0.0014	0.1986 ± 0.0014
¹⁴³ Nd/ ¹⁴⁴ Nd	0.512604 ± 0.000007	0.512570 ± 0.000014	0.512712 ± 0.000009	$0.512585 \pm$	$0.512619 \pm$
ivu/ ivu	0.512004 ± 0.000007	0.512570 ± 0.000014	0.512/12 ± 0.000000	0.000008	0.000008
¹⁷⁶ Lu/ ¹⁷⁷ Hf	0.0336 ± 0.0002	0.0338 ± 0.0002	0.0304 ± 0.0002	0.0306 ± 0.0002	0.0315 ± 0.0002
¹⁷⁶ Hf/ ¹⁷⁷ Hf	0 282825 - 0 000015	0 202060 - 0 000016	0 282825 + 0 000029	$0.282689 \pm$	$0.282702 \pm$
	0.282825 ± 0.000015	0.282868 ± 0.000016	0.282825 ± 0.000038	0.000019	0.000013

1043 **Table 5.** Elemental abundance ratios in H chondrites (UHC: unequilibrated H chondrite, EHC: equilibrated

- 1044 H chondrite).
- 1045 The uncertainties are derived from the uncertainties of the respective elemental abundances. The
- 1046 uncertainties for the mean values are 95% confidence interval. The mean values of non-Antarctic HC and
- 1047 Antarctic HC are calculated from the following literatures.
- 1048 * The data of A 09516 (H6) are excluded from the means (see section 3.2).
- ^a Jarosewich (1990) for Na and Al. ^b Wasson and Kallemeyn (1988) for Rb, Sr, and Ba. ^c Nishikawa *et al.*,
- 1050 (1990) for Rb, Sr, and Ba.

	Rb	Sr	Ва
Name	(Na + Al)/2	(Na + Al)/2	(Na + Al)/2
A-881258	3.16 ± 0.12	11.3 ± 0.3	4.11 ± 0.14
A-880941	1.77 ± 0.09	12.0 ± 0.5	4.35 ± 0.18
Y-793574	2.56 ± 0.09	9.55 ± 0.32	3.43 ± 0.15
Y-790461	3.19 ± 0.09	11.3 ± 0.2	4.04 ± 0.24
ALH 78084	2.80 ± 0.13	10.7 ± 0.4	3.94 ± 0.24
A 09436-1	2.32 ± 0.05	10.7 ± 0.2	4.04 ± 0.15
A 09436-2	2.38 ± 0.07	11.1 ± 0.2	4.23 ± 0.11
A 09387	3.77 ± 0.11	10.7 ± 0.3	3.88 ± 0.11
A 09618	2.65 ± 0.06	11.0 ± 0.2	3.96 ± 0.08
A 09516-1	0.895 ± 0.049	8.71 ± 0.35	3.19 ± 0.15
A 09516-2	1.29 ± 0.07	12.3 ± 0.5	5.24 ± 0.31
Y-790960	3.11 ± 0.22	10.4 ± 0.3	3.65 ± 0.25
NWA 6752	2.51 ± 0.09	26.5 ± 0.7	202 ± 6
NWA 6771	3.21 ± 0.08	21.8 ± 0.5	22.9 ± 0.5
Sahara 97035	3.96 ± 0.13	123 ± 3	51.4 ± 1.3
Antarctic UHC mean $(n = 7)$	2.60 ± 0.50	11.0 ± 0.8	4.02 ± 0.29
Antarctic EHC mean* $(n = 3)$	3.17 ± 0.56	10.7 ± 0.3	3.83 ± 0.16
Antarctic HC mean* (n = 10)	2.77 ± 0.56	10.9 ± 0.7	3.96 ± 0.27
Hot desert HC mean $(n = 3)$	3.23 ± 3.23	57.2 ± 57.4	92.1 ± 96.3
Non-Antarctic HC mean ^{a, b}	3.28 ± 0.09	11.3 ± 0.3	4.74 ± 0.13
Antarctic HC mean ^{a, c}	3.24 ± 0.48	10.5 ± 1.1	3.04 ± 0.85

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1058 1059 1060 **Table 6.** Correlation coefficients (*r*) between elemental ratios and isotopic ratios in Antarctic HCs.

1061 Strong and no correlations are defined as |r| > 0.6 and 0.4 > |r|, and are shown in bolded and underlined,

1062 respectively.

	Corr	elation coeffici	ients for all san	nples	Correlation coefficients excluding A 09516 (H6)							
	147Sm/144Nd	143Nd/144Nd	¹⁷⁶ Lu/ ¹⁷⁷ Hf	¹⁷⁶ Hf/ ¹⁷⁷ Hf	147Sm/144Nd	143Nd/144Nd	¹⁷⁶ Lu/ ¹⁷⁷ Hf	¹⁷⁶ Hf/ ¹⁷⁷ Hf				
Al/Mg	-0.50	0.29	-0.42	-0.12	-0.21	-0.47	-0.36	-0.51				
P/Mg	0.48	-0.57	0.64	0.62	-0.60	-0.45	-0.04	0.09				
Ca/Mg	0.19	0.40	-0.24	-0.01	0.14	0.45	-0.74	-0.54				
Ti/Mg	-0.42	0.28	-0.63	-0.43	-0.23	<u>-0.01</u>	-0.79	-0.73				
Y/Mg	0.65	-0.65	0.78	0.66	0.04	-0.48	0.56	0.40				
P/Ca	<u>0.33</u>	-0.74	0.74	0.60	-0.47	-0.63	0.58	0.50				
Y/Ca	0.41	-0.71	0.75	0.49	<u>-0.06</u>	-0.55	0.80	0.58				

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1067 **Table 7.** Major hosts of REEs and Hf, and alteration types for chondrites.

^a Curtis and Schmitt (1979) and Ebihara and Honda (1983). ^b Larimer and Ganapathy (1987) and Ebihara

1069 (1988). ^c Martin *et al.* (2013). ^d Ebihara and Honda (1987). ^e Maeda *et al.* (2017a, 2017b). ^f Alexander

1070 (1994) and Mason and Graham (1970). ^g Barrat *et al.* (2014). ^h Krot *et al.* (2003) and Huss *et al.* (2006). ⁱ

1071 Weisberg *et al.* (1996) and Barosch *et al.* (2020).

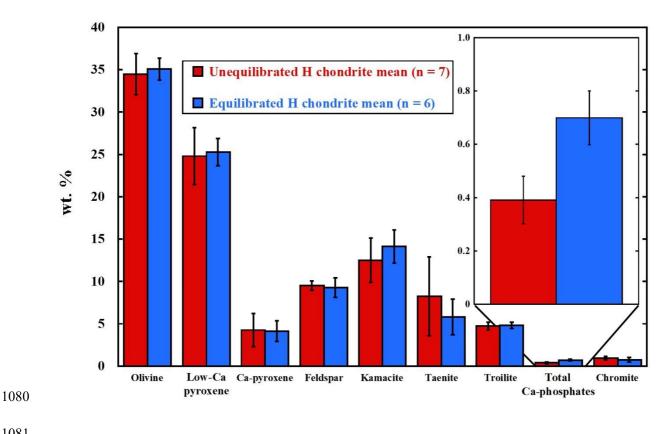
Chondrite	Major hosts of REEs	Major hosts of Hf	Alteration type
H, L, LL	Ca-phosphates ^a	Ca-pyroxene (silicates) ^f	
EH, EL	Oldhamite (CaS) ^b	Sulfides and silicates ^g	Thermal metamorphism ^h
CK	Ca-phosphates and silicates ^c	Silicates ^c	
CV	Ca, Al-rich inclusions (CAIs) ^d	?	Aqueous alteration and
СО	?	?	weak thermal metamorphism ^h
CI, CM, CR,	0	0	A teach
CH, CB	?	?	Aqueous alteration ^h
Rumuruti	Ca-phosphates ^e	?	Thermal metamorphism ^h
77 1	0	0	Aqueous alteration and
Kakangari	?	?	weak thermal metamorphism ⁱ

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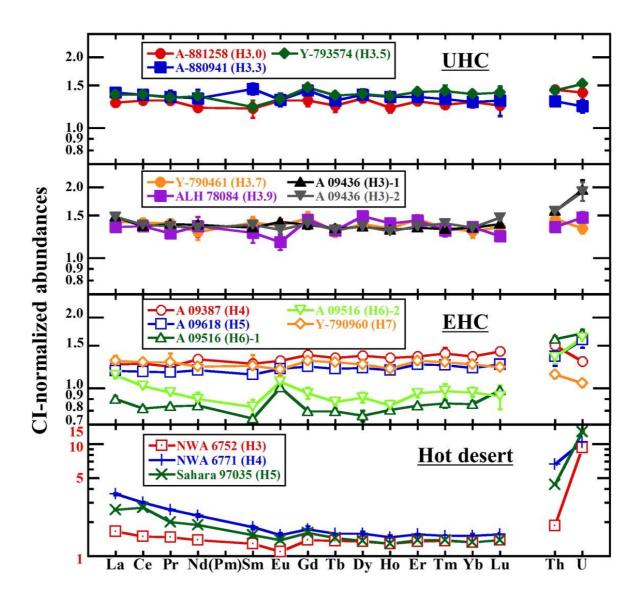
1073

FIGURE CAPTIONS

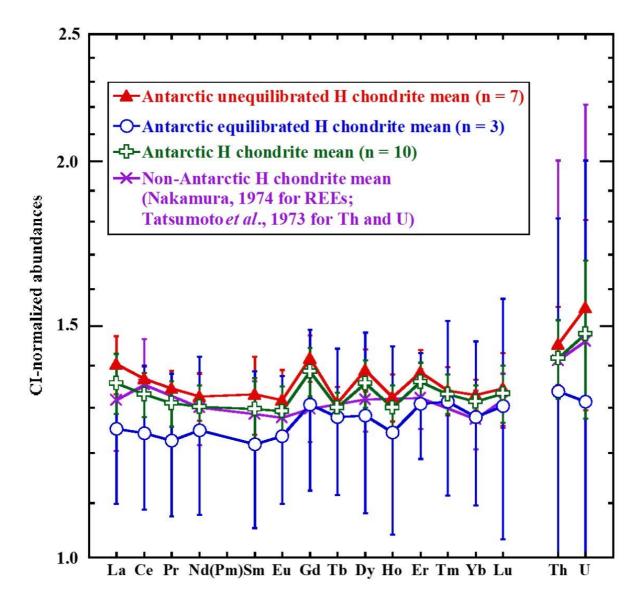
Fig. 1. The mean modal abundances (wt. %) in UHC and EHC obtained in this study (n = 7 and 6, respectively). The mean modal abundances of low-Ca pyroxene, feldspar, and chromite may include forsterite, silica, and ilmenite, respectively (see section 3.1). The uncertainties represent 95% confidence interval of the mean values.

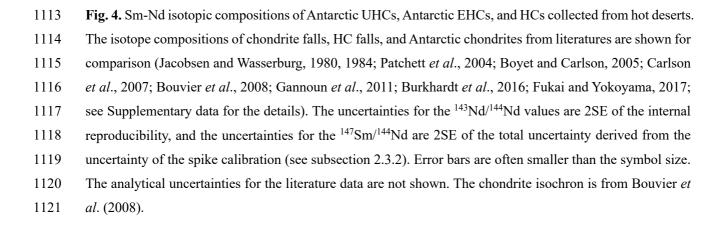


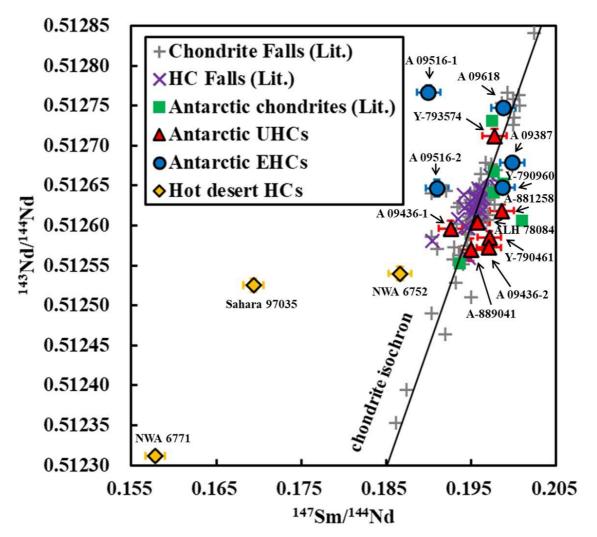
1091 Fig. 2. CI-normalized REE, Th, and U abundances in each Antarctic and hot desert HC. The uncertainties 1092 represent 1SD of the measurements by Q-ICP-MS. Error bars are often smaller than the symbol size. Note 1093 that the scale of y-axis for hot desert is different from the other. CI values are from Anders and Grevesse 1094 (1989).



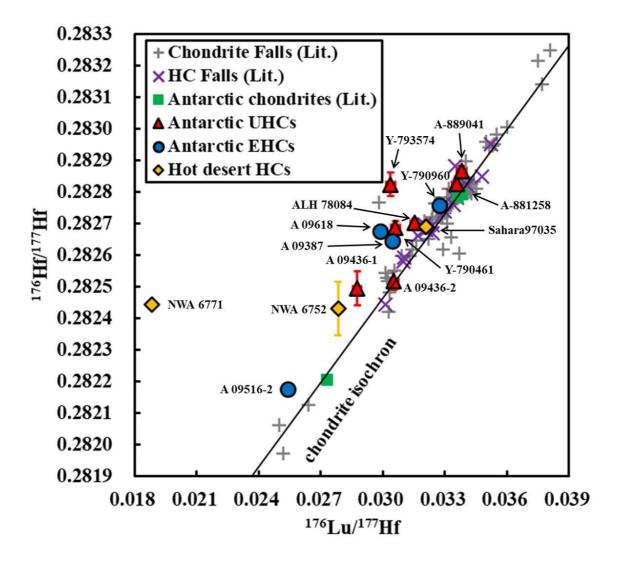
- 1102 Fig. 3. Mean CI-normalized REE, Th, and U abundances of Antarctic UHCs, Antarctic EHCs, and Antarctic
- 1103 HCs (n = 7, 3, and 10, respectively). The mean values of non-Antarctic HCs are also shown for comparison
- 1104 (Nakamura, 1974 for REEs; Tatsumoto et al., 1973 for Th and U). The uncertainties are 95% confidence
- 1105 interval of the mean values. CI values from Anders and Grevesse (1989).







- 1127 Fig. 5. Lu-Hf isotopic compositions of Antarctic UHCs, Antarctic EHCs, and HCs collected from hot deserts.
- 1128 The isotope compositions of chondrite falls, HC falls, and Antarctic chondrites from literature are also shown
- 1129 for comparison (Blichert-Toft and Albarède, 1997; Bizzarro *et al.*, 2003; Patchett *et al.*, 2004; Bouvier *et al.*,
- 1130 2008; Dauphas and Pourmand, 2011; see Supplementary data for the details). Note that Blichert-Toft and
- 1131 Albarède (1997) did not use high-pressure vessels during the sample digestion. The data of Farmington (L5),
- 1132 Adhi Kot (EH4), and Jajh Deh Kot Lalu (EL6) from Dauphas and Pourmand (2011) plot outside the range of
- 1133 the figure. The uncertainties for the 176 Hf/ 177 Hf values are 2SE of the internal reproducibility, and the 1134 uncertainties for the 176 Lu/ 177 Hf values are 2SE of the total uncertainty derived from the uncertainty of the
- spike calibration (see subsection 2.3.2). Error bars are often smaller than the symbol size. The analytical
- 1136 uncertainties for the literature data are not shown. The chondrite isochron is from Bouvier *et al.* (2008).

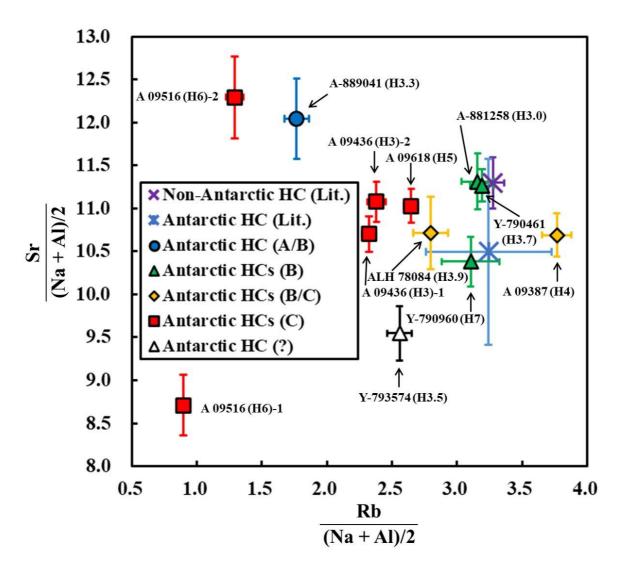


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- Fig. 6. Comparison of Rb and Sr abundances normalized by the mean Na and Al abundances, as a measure for the amount of feldspar in the sample. The plotted groups are divided based on their weathering index. The values for Non-Antarctic HC (Lit.) and Antarctic HC (Lit.) are calculated from Wasson and Kallemeyn (1988), Jarosewich (1990), and Nishikawa *et al.* (1990), shown in Table 5. The uncertainties are derived from the individual uncertainties of each elemental abundance, and those for literature values are 95% confidence
- 1146 interval of the mean values. Error bars are often smaller than the symbol size.





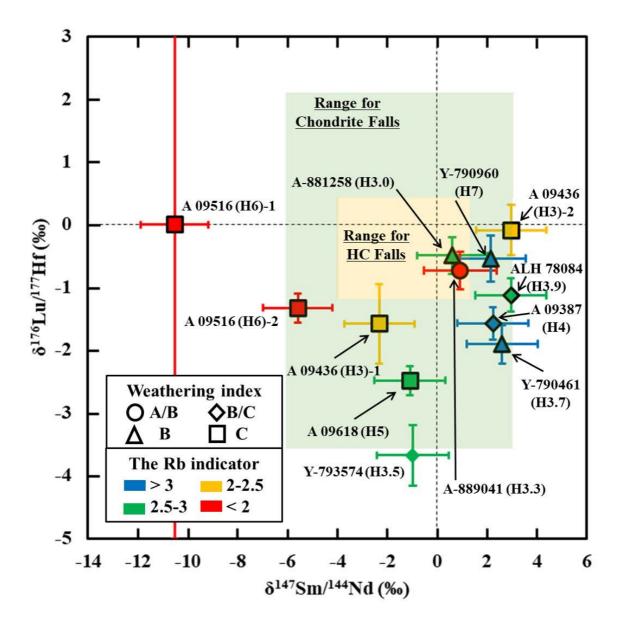
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1154 Hf systems. The symbols of each plot reflect the weathering index and the Rb indicator (Table 5) by shape 1155 and color, respectively. Note that no weathering index for Y-793574 (H3.5) and ¹⁷⁶Hf/¹⁷⁷Hf of A 09516 (H6)-1156 1 have been determined, hence the symbol of Y-793574 (H3.5) is not surrounded by black line and the 1157 $\delta^{176}Lu/^{177}$ Hf of A 09516 (H6)-1 was not determined. The ranges for chondrite falls and HC falls are calculated 1158 from the literature values shown in Figs. 4 and 5 (Supplemental data), and are shown by light green and light 1159 red, respectively. The respective isochrons used for the calculations are from Bouvier *et al.* (2008). The 1160 uncertainties except for the $\delta^{176}Lu/177$ Hf of A 09516 (H6)-1 are derived from the uncertainties of each

Fig. 7. Deviations of ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁷⁶Lu/¹⁷⁷Hf from the respective chondrite isochron on Sm-Nd and Lu-

1161 isotopic ratios. The uncertainties of the isochrons are not considered.



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1164 SUPPLEMENTARY DATA

- 1165 Supplementary data-1. Sm-Nd and Lu-Hf isotopic compositions of H chondrite falls, chondrite falls, and Antarctic chondrites (WI: weathering index for Antarctic chondrites).
- 1166 The weathering indexes for Antarctic chondrites are from Antarctic Meteorite Newsletter, NASA. The chondrite fall mean includes H chondrites.
- 1167 *MAC 88136 and MAC 88180 are possibly paired (Antarctic Meteorite Newsletter 13, NASA).
- ¹¹⁶⁸ ^a Jacobsen and Wasserburg (1980. ^b Jacobsen and Wasserburg (1984). ^c Blichert-Toft and Albarède (1997). ^d Bizzarro *et al.* (2003). ^e Patchett *et al.* (2004). ^f Boyet and Carlson (2005). ^g Carlson *et al.*
- 1169 (2007). ^h Bouvier et al. (2008). ⁱ Dauphas and Pourmand (2011). ^j Gannoun et al. (2011). ^k Burkhardt et al. (2016). ¹ Fukai and Yokoyama (2017).
- 1170 Supplementary data-2. Average Sm-Nd and Lu-Hf isotope compositions in each class calculated using only falls from the supplementary data-1.
- 1171 The uncertainties associated with the isotopic ratios are simply calculated at 2SE, except for N = 2, where the values reflect the range of values. Tieschitz and Bjurböle are excluded from this calculation
- 1172 due to their intermediate petrologic features. The following samples are excluded from the respective means because their values seem not to be representative: Farmington from Dauphas and Purmand
- 1173 (2011), M'Bale from Bizzarro et al. (2003), and Peace River from Carlson et al. (2007) for the means of L chondrites; Paragould from Dauphas and Purmand (2011) and Saint-Séverin (Light) and
- 1174 (Dark) from Jacobsen and Wasserburg (1980) for the means of LL chondrites; Orgueil from Blichert-Toft and Albarède (1997) for the means of CI chondrites; the Sm-Nd values of Alende piece from
- 1175 Patchett et al. (2004) and Grosnaja from Carlson et al. (2007) for the means of CV chondrites; Abee and Abee (bis) from Burkhardt et al. (2016) and Adhi Kot, St. Mark's, and Saint-Sauveur from
- 1176 Dauphas and Purmand (2011) for the means of EH chondrites; Eagle, Jajh Deh Kot Lalu, and Khairpur from Dauphas and Purmand (2011) for the means of EL chondrites.
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Meteorite	Class	Nd (ppb)	Sm (ppb)	$^{147}{\rm Sm}/^{144}{\rm Nd}$	2SE	143Nd/144Nd	2SE	Lu (ppb)	Hf (ppb)	¹⁷⁶ Lu/ ¹⁷⁷ Hf	2SE	¹⁷⁶ Hf/ ¹⁷⁷ Hf	2SE	Ref.
<u>H chondrite Falls</u>														
Sharps	H3.4	572	186	0,1959	0,0002	0,512624	0,000006							f
Sharps (bis)	H3.4	592	190	0,1951	0,0002	0,512622	0,000002							f
Sharps	H3.4	570	185	0,1963	0,0010	0,512644	0,000009	33,6	143	0,0334	0,0002	0,282791	0,000007	h
Dhajala	H3.8	623	201	0,1954	0,0002	0,512621	0,000003							f
Dhajala	H3.8	576	186	0,1958	0,0010	0,512633	0,000008	33,2	143	0,0330	0,0002	0,282740	0,000006	h
Ankober	H4	541	175	0,1960	0,0004	0,512604	0,000006	30,7	137	0,0317	0,0002	0,282661	0,000008	e
Avanhandava	H4	562	182	0,1961	0,0004	0,512617	0,000005	32,8	144	0,0324	0,0002	0,282669	0,000018	e
Bielokrynitschie	H4							31,4	144	0,0310	0,0004	0,282581	0,000036	i
Ochansk	H4	491	158	0,1941	0,0004	0,512638	0,000006	31,6	149	0,0301	0,0002	0,282444	0,000009	e
Ochansk (bis)	H4	618	200	0,1960	0,0004	0,512630	0,000009	35,3	150	0,0334	0,0002	0,282761	0,000016	e
Ochansk	H4							34,0	151	0,0320	0,0004	0,282699	0,000036	i

Sm-Nd and Lu-Hf isotope compositions of H chondrite falls, chondrite falls, and Antarctic chondrites (WI: weathering index for Antarctic chondrites).

Kesen	H4							32,4	133	0,0348	0,0004	0,282849	0,000036	i
Sainte Marguerite	H4	702	227	0,1955	0,0002	0,512635	0,000006							k
Sainte Marguerite (bis)	H4	773	250	0,1954	0,0002	0,512636	0,000005							k
Allegan	Н5	570	185	0,1958	0,0004	0,512646	0,000010	32,1	129	0,0352	0,0002	0,282951	0,000014	e
Allegan	Н5	584	188	0,1952	0,0002	0,512615	0,000003							k
Forest City	Н5	624	203	0,1964	0,0004	0,512634	0,000006	35,8	151	0,0336	0,0002	0,282782	0,000012	e
Forest City	Н5	675	217	0,1944	0,0002	0,512599	0,000003							k
Forest City	Н5			0,1960	0,0030	0,512633	0,000003							1
Gao	Н5							33,2	141	0,0335	0,0003	0,282884	0,000020	с
Herredia	Н5							32,4	143	0,0321	0,0003	0,282681	0,000037	с
Herredia	Н5							34,3	143	0,0341	0,0001	0,282821	0,000022	d
Jilin	Н5	550	173	0,1904		0,512581	0,000003							j
Pultusk	Н5	592	193	0,1973	0,0004	0,512663	0,000006	35,1	148	0,0337	0,0002	0,282810	0,000014	e
Pultusk	Н5	660	211	0,1934	0,0002	0,512608	0,000005							k
Richardton	Н5	562	184	0,1979	0,0004	0,512648	0,000006	33,3	153	0,0310	0,0002	0,282593	0,000009	e
Richardton	Н5	595	191	0,1942	0,0002	0,512622	0,000003							f
Guareña	H6	902	290	0,1947	0,0002	0,512560	0,000024							а
Kernouve	H6							29,3	129	0,0324	0,0004	0,282695	0,000036	i
Queens Mercy	H6	587	189	0,1946	0,0002	0,512597	0,000003							k
Ucera	Н	569	185	0,1963		0,512645	0,000001							g
HC fall mean		613	198	0,1953	0,0006	0,512623	0,000009	33,0	143	0,0328	0,0007	0,282730	0,000060	
N for the mean values		23			2	24		17				17		
Chondrite Falls														
Tieschitz	H/L3.4	633	203	0,1937	0,0010	0,512628	0,000007	36,0	157	0,0325	0,0002	0,282766	0,000006	h
Hallingeberg	L3.4	616	200	0,1960	0,0010	0,512629	0,000007	35,1	148	0,0336	0,0002	0,282781	0,000007	h
Hedjaz	L3.7							32,6	141	0,0329	0,0001	0,282726	0,000021	d
Mezo-Madaras	L3.7	621	201	0,1960	0,0010	0,512626	0,000009	35,6	152	0,0333	0,0002	0,282764	0,000004	h
Bovedy	L3.8	687	223	0,1961	0,0010	0,512629	0,000007	40,3	172	0,0333	0,0002	0,282657	0,000009	h

Bald Mountain	L4							37,1	147	0,0360	0,0004	0,283005	0,000036	i
Saratov	L4			0,1960	0,0002	0,512616	0,000005							1
Tennasilm	L4							36,6	154	0,0337	0,0003	0,282606	0,000025	с
Tennasilm	L4							33,8	146	0,0329	0,0001	0,282720	0,000025	d
Farmington	L5							44,2	148	0,0425	0,0004	0,283741	0,000036	i
Farmington	L5	655	210	0,1944	0,0002	0,512591	0,000006							k
Homestead	L5	678	218	0,1949	0,0004	0,512640	0,000008	35,2	150	0,0332	0,0002	0,282810	0,000009	e
Homestead	L5	583	188	0,1951		0,512625	0,000001							g
Alfianello	L6							33,5	155	0,0317		0,282599	0,000014	c
Alfianello	L6							42,3	169	0,0355	0,0001	0,282982	0,000013	d
Bruderheim	L6	530	173	0,1970	0,0004	0,512648	0,000006	31,0	145	0,0302	0,0002	0,282518	0,000022	e
Bruderheim	L6	540	175	0,1955	0,0004	0,512620	0,000003							f
Bruderheim	L6	909	291	0,1935	0,0002	0,512563	0,000003							k
Fisher	L6							37,1	150	0,0350	0,0004	0,282959	0,000008	c
Girgenti	L6	674	218	0,1954	0,0004	0,512643	0,000006	38,2	173	0,0313	0,0002	0,282597	0,000009	e
Harleton	L6							32,0	150	0,0303	0,0004	0,282421	0,000036	i
Holbrook	L6	557	182	0,1975	0,0004	0,512656	0,000008	33,9	159	0,0303	0,0002	0,282483	0,000012	e
Holbrook (bis)	L6	571	187	0,1974	0,0004	0,512677	0,000009	34,0	160	0,0301	0,0002	0,282544	0,000014	e
Leedey	L6			0,1966	0,0004	0,512629	0,000023							b
M'Bale	L6							35,4	152	0,0331	0,0003	0,282700	0,000016	с
M'Bale	L6							30,2	162	0,0264	0,0001	0,282127	0,000011	d
M'Bale (bis)	L6							36,8	171	0,0306	0,0001	0,282552	0,000025	d
Modoc (1905)	L6			0,1950	0,0020	0,512510	0,000005							1
Peace River	L6	606	82	0,1903		0,512641	0,000001							g
Bjurböle	L/LL4	650	208	0,1933	0,0004	0,512602	0,000007	38,9	170	0,0324	0,0002	0,282714	0,000008	e
Semarkona	LL3.0	662	215	0,1960	0,0010	0,512622	0,000012	37,8	162	0,0331	0,0002	0,282749	0,000005	h
Krymka	LL3.1	601	193	0,1945	0,0010	0,512575	0,000012	34,2	144	0,0337	0,0002	0,282786	0,000008	h
Chainpur	LL3.5	633	205	0,1956	0,0010	0,512623	0,000008	36,1	154	0,0333	0,0002	0,282786	0,000005	h
Parnallee	LL3.6	648	210	0,1961	0,0010	0,512622	0,000010	37,1	156	0,0339	0,0002	0,282809	0,000005	h

Hamlet	LL4							39,0	177	0,0314	0,0004	0,282619	0,000036	i
Soko-Banja	LL4							33,8	149	0,0324	0,0004	0,282722	0,000036	i
Chelyabinsk	LL5	661	215	0,1963	0,0002	0,512647	0,000003							k
Tuxtuac	LL5							38,9	168	0,0329	0,0004	0,282619	0,000036	i
Tuxtuac	LL5			0,1920	0,0020	0,512643	0,000005							1
Paragould	LL5							45,2	172	0,0375	0,0004	0,283217	0,000036	i
Dhurmsala	LL6	593	193	0,1965	0,0002	0,512637	0,000003							k
Saint-Séverin (Light)	LL6	1386	440	0,1920	0,0002	0,512464	0,000025							a
Saint-Séverin (Dark)	LL6	560	185	0,2000	0,0002	0,512735	0,000027							а
Saint-Séverin (WR)	LL6			0,1964	0,0002	0,512618	0,000020							b
Saint-Séverin	LL6							42,7	185	0,0330	0,0004	0,282735	0,000036	i
Saint-Séverin	LL6			0,1910	0,0020	0,512571	0,000007							1
Ivuna	CI1	410	133	0,1967	0,0010	0,512679	0,000013							h
Ivuna	CI1							28,0	116	0,0343	0,0004	0,282826	0,000036	i
Orgueil	CI1							27,4	130	0,0298	0,0003	0,282767	0,000033	c
Orgueil	CI1	462	150	0,1968		0,512639	0,000001							g
Orgueil	CI1	459	149	0,1964	0,0010	0,512609	0,000014	25,1	106	0,0335	0,0002	0,282784	0,000005	h
Cold Bokkeveld	CM2	602	195	0,1962		0,512639	0,000001							g
Mighei	CM2	612	199	0,1966	0,0004	0,512644	0,000006	35,0	147	0,0339	0,0002	0,282798	0,000007	e
Mighei	CM2	625	202	0,1953		0,512618	0,000001							g
Mighei	CM2							33,0	139	0,0338	0,0004	0,282795	0,000036	i
Murchison	CM2	635	207	0,1969	0,0002	0,512607	0,000025							а
Murchison (bis)	CM2	670	214	0,1932	0,0002	0,512529	0,000034							а
Murchison (ter)	CM2	643	209	0,1966	0,0002	0,512628	0,000025							а
Murchison	CM2			0,1966	0,0002	0,512627	0,000016							b
Murchison	CM2							30,8	129	0,0340	0,0003	0,282899	0,000035	c
Murchison	CM2							34,0	150	0,0323	0,0001	0,282679	0,000019	d
Murchison	CM2	632	207	0,1977	0,0004	0,512626	0,000008	35,6	150	0,0338	0,0002	0,282804	0,000010	e
Murchison	CM2			0,1970	0,0020	0,512613	0,000005							1

Murray	CM2	641	207	0,1949	0,0004	0,512632	0,000007	36,1	155	0,0330	0,0002	0,282742	0,000011	e
Murray	CM2	638	207	0,1962		0,512637	0,000001							g
Pollen	CM2			0,1967	0,0002	0,512641	0,000024							b
Al Rais	CR2	548	179	0,1970	0,0010	0,512647	0,000009							h
Felix	CO3.1	735	238	0,1959	0,0010	0,512620	0,000007	46,3	196	0,0335	0,0002	0,282777	0,000004	h
Kainsaz	CO3.2	727	237	0,1969	0,0004	0,512632	0,000006	59,9	242	0,0351	0,0002	0,282942	0,000006	e
Kainsaz	CO3.2							43,8	186	0,0335	0,0004	0,282817	0,000036	i
Lancé	CO3.5	733	239	0,1969	0,0010	0,512647	0,000014	40,5	169	0,0339	0,0002	0,282799	0,000004	h
Lancé	CO3.5							42,5	177	0,0333	0,0004	0,282762	0,000036	i
Bali	CV3.6	771	256	0,2006	0,0010	0,512763	0,000014	40,6	171	0,0338	0,0002	0,282838	0,000009	h
Allende	CV3.6	1020	327	0,1940	0,0010	0,512552	0,000006	45,6	191	0,0339	0,0002	0,282816	0,000018	h
Allende (bis)	CV3.6	1021	327	0,1939	0,0010	0,512559	0,000010	45,7	190	0,0340	0,0002	0,282824	0,000005	h
Allende	CV3							49,8	211	0,0335	0,0003	0,282776	0,000015	c
Allende (bis)	CV3							46,1	192	0,0341	0,0003	0,282825	0,000011	c
Allende	CV3							43,3	186	0,0331	0,0001	0,282783	0,000012	d
Alende piece	CV3	1319	406	0,1861	0,0004	0,512353	0,000007	42,4	177	0,0341	0,0002	0,282847	0,000007	e
Allende pdr	CV3	1010	325	0,1947	0,0004	0,512606	0,000009	49,6	206	0,0341	0,0002	0,282828	0,000017	e
Allende pdr (bis)	CV3	991	319	0,1947	0,0004	0,512589	0,000009	48,1	198	0,0344	0,0002	0,282834	0,000016	e
Allende	CV3	986	315	0,1930	0,0002	0,512573	0,000003							f
Allende (bis)	CV3	1010	324	0,1938	0,0002	0,512563	0,000002							f
Allende (ter)	CV3	980	313	0,1929	0,0002	0,512558	0,000003							f
Allende	CV3	854	275	0,1947		0,512566	0,000001							g
Allende	CV3							46,0	192	0,0341	0,0004	0,282835	0,000036	i
Allende (bis)	CV3							45,9	192	0,0341	0,0004	0,282795	0,000036	i
Allende (ter)	CV3							45,2	189	0,0340	0,0004	0,282839	0,000036	i
Allende (quarter)	CV3							48,5	206	0,0337	0,0004	0,282814	0,000036	i
Allende (quīnquiēs)	CV3							46,0	193	0,0340	0,0004	0,282824	0,000036	i
Allende (sexiēs)	CV3							46,4	193	0,0343	0,0004	0,282795	0,000036	i
Allende (septiēns)	CV3							46,1	192	0,0342	0,0004	0,282801	0,000036	i

Allende	CV3	735	241	0,1979		0,512670	0,000003							j
Allende	CV3	863	280	0,1959	0,0002	0,512651	0,000003							k
Allende (bis)	CV3	879	285	0,1961	0,0002	0,512664	0,000006							k
Allende (ter)	CV3	1009	325	0,1948	0,0002	0,512620	0,000005							k
Allende	CV3			0,1950	0,0020	0,512608	0,000019							1
Grosnaja	CV3	1076	360	0,2024		0,512841	0,000001							g
Grosnaja	CV3							39,1	162	0,0345	0,0004	0,282811	0,000036	i
Mokoia	CV3	766	249	0,1963		0,512639	0,000001							g
Vigarano	CV3	876	282	0,1951	0,0010	0,512609	0,000010	41,8	174	0,0340	0,0002	0,282811	0,000006	h
Vigarano	CV3							49,2	207	0,0338	0,0004	0,282789	0,000036	i
Ningqiang	CK3	1098	357	0,1965	0,0010	0,512639	0,000008	68,8	276	0,0353	0,0002	0,282947	0,000004	h
Karoonda	CK4	932	301	0,1951	0,0004	0,512602	0,000007	47,4	190	0,0354	0,0002	0,282951	0,000010	e
Qingzhen	EH3							24,5	107	0,0326	0,0004	0,282704	0,000036	i
Abee	EH4			0,2000	0,0002	0,512726	0,000022							b
Abee	EH4	464	146	0,1903	0,0002	0,512586	0,000008							f
Abee	EH4	229	71	0,1874	0,0002	0,512395	0,000006							k
Abee (bis)	EH4	278	88	0,1903	0,0002	0,512490	0,000005							k
Adhi Kot	EH4							23,9	73,3	0,0465	0,0004	0,284055	0,000036	i
Indarch	EH4	412	133	0,1957	0,0004	0,512603	0,000006	24,8	105	0,0335	0,0002	0,282797	0,000009	e
Indarch	EH4	365	117	0,1934		0,512623	0,000001							g
Indarch	EH4							23,5	104	0,0320	0,0004	0,282685	0,000036	i
Indarch	EH4	440	142	0,1953	0,0002	0,512622	0,000006							k
Indarch (bis)	EH4	439	141	0,1948	0,0002	0,512611	0,000005							k
St. Mark's	EH5							23,2	87,7	0,0377	0,0004	0,283143	0,000036	i
St. Mark's	EH5	452	146	0,1953		0,512604	0,000002							j
Saint-Sauveur	EH5							20,3	76,1	0,0381	0,0004	0,283250	0,000036	i
Saint-Sauveur	EH5	458	148	0,1956	0,0002	0,512624	0,000003							k
Daniel's Kuil	EL6							29,3	138	0,0302	0,0004	0,282528	0,000036	i
Eagle	EL6							31,2	176	0,0252	0,0004	0,281972	0,000036	i

iv for the mean values						U				-		3		<u> </u>
N for the mean values	11			0,1970	-	0,512045 6	0,000049			0,0316	0,0043	0,282596 <i>3</i>	0,000388	
Antarctic chondrite mean		512	10/	0,1976 0,1976	0,0019	0,512642 0,512643				0.0316	0,0043	0,282596	0 000380	J
MAC 02837 (WI: C) MAC 02839 (WI: C)	EL3 EL3	497 572	163 187	0,1987 0,1976		0,512653 0,512642	0,000000 0,000003							J ;
MAC 88180 (WI: C)*	EL3	819	267	0,1974		0,512732	0,000003							J
MAC 88136 (WI: A)*	EL3	397	127	0,1936	0,0010	0,512553	0,000008	29,0	122	0,0338	0,0002	0,282794	0,000007	h
KLE 98300 (WI: A)	EH3	490	160	0,1976	0,0010	0,512668	0,000007	24,0	124	0,0273	0,0001	0,282208	0,000004	h
ALHA 77295 (WI: B)	EH3	344	114	0,2009		0,512607	0,000003							j
GRA 95229 (WI: A)	CR2							44,6	188	0,0336	0,0002	0,282787	0,000007	h
Antarctic chondrites														
Iv for the mean values					1	07						70		
<i>N</i> for the mean values				0,1755		07	0,00012			0,0551	0,0000	96	0,000050	
Chondrite fall mean	ELU			0,1955	0,0005	0,512622	0,000012	23,0	115	0,0310 0,0331	0,0004 0,0006	0,282048 0,282757	0,000036 0,000056	1
Pillistfer Pillistfer	EL6 EL6							26,8 25,0	119	0,0320	0,0003 0,0004	0,282708 0,282648	0,000014 0,000036	c i
Khairpur Billistfor	EL6 EL6	411	134	0,1965		0,512633	0,000002	26.0	119	0,0320	0.0002	0 202700	0.000014	J
Khairpur	EL6	411	124	0.1075		0.512(22	0.00000	24,1	137	0,0250	0,0004	0,282061	0,000036	1
Jajh Deh Kot Lalu	EL6							22,9	159	0,0205	0,0004	0,281675	0,000036	1
Hvittis (ter)	EL6	557	183	0,1993	0,0002	0,512766	0,000005							k
Hvittis (bis)	EL6	818	269	0,1986	0,0002	0,512753	0,000006							k
Hvittis	EL6	480	159	0,1999	0,0002	0,512758	0,000003							k
Hvittis	EL6	361	120	0,2007		0,512750	0,000003							j
Hvittis	EL6							30,4	134	0,0322	0,0004	0,282651	0,000036	i

The weathering indexes for Antarctic chondrites are from Antarctic Meteorite Newsletter, NASA. The chondrite fall mean includes H chondrite falls.

*MAC 88136 and MAC 88180 are possibly paired (Antarctic Meteorite Newsletter 13, NASA).

^a Jacobsen and Wasserburg (1980). ^b Jacobsen and Wasserburg (1984). ^c Blichert-Toft and Albarède (1997). ^d Bizzarro *et al.* (2003). ^e Patchett *et al.* (2004). ^f Boyet and Carlson (2005). ^g Carlson *et al.* (2007). ^h Bouvier *et al.* (2008). ⁱ Dauphas and Pourmand (2011). ^j Gannoun *et al.* (2011). ^k Burkhardt *et al.* (2016). ¹ Fukai and Yokoyama (2017).

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