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► To cite this version:

Lionel G Vacher, Laurette Piani, Thomas Rigaudier, Dorian Thomassin, Maxime Piralla, et al.. Hydrogen in chondrites: Influence of parent body alteration and 1 atmospheric contamination on primordial components. *Geochimica et Cosmochimica Acta*, 2020, 281, pp.53-66. 10.1016/j.gca.2020.05.007 . hal-03009498

HAL Id: hal-03009498

<https://hal.univ-lorraine.fr/hal-03009498>

Submitted on 17 Nov 2020

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1 **Hydrogen in chondrites: Influence of parent body alteration and**
2 **atmospheric contamination on primordial components**

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22 **Abstract**

23
24 Hydrogen occurs at the near percent level in the most hydrated chondrites (CI and
25 CM) attesting to the presence of water in the asteroid-forming regions. Their H abundances
26 and isotopic signatures are powerful proxies for deciphering the distribution of H in the
27 protoplanetary disk and the origin of Earth's water. Here, we report H contents and isotopic
28 compositions for a set of carbonaceous and ordinary chondrites, including previously
29 analyzed and new samples analyzed after the powdered samples were degassed under vacuum
30 at 120°C for 48 hours to remove adsorbed atmospheric water. By comparing our results to
31 literature data, we reveal that the H budgets of both H-poor and H-rich carbonaceous
32 chondrites are largely affected by atmospheric moisture, and that their precise quantification
33 requires a specific pre-degassing procedure to correct for terrestrial contamination. Our results
34 show that indigenous H contents of CI carbonaceous chondrites usually considered the more
35 hydrated meteorites are almost a factor of 2 lower than those previously reported, with
36 uncontaminated D/H ratios differing significantly from that of Earth's oceans. Without pre-

37 degassing, the H concentrations of H-poor samples (e.g., CVs chondrites) are also affected by
38 terrestrial contamination. After correction for contamination, it appears that the amount of
39 water in chondrites is not controlled by the matrix modal abundance, suggesting that the
40 different chondritic parent bodies accreted variable amounts of water-ice grains. Our results
41 also imply that (i) thermal metamorphism play an important role in determining the H content
42 of both CV and ordinary chondrites but without affecting drastically their H isotopic
43 composition since no clear D enrichment is observed with the increase of petrographic type
44 and (ii) the D enrichment of ordinary chondrites does not result from the loss of isotopically
45 light H₂ induced by metal oxidation but is rather linked to the persistence of a thermally
46 resistant D-rich component.

47

48 **1. Introduction**

49

50 Hydrogen is the most abundant element in the solar system, but is present in only
51 minor to trace amounts in asteroids and planetary bodies. Throughout the solar system and its
52 history, H has occurred in various species, most commonly (i) H₂ in the gas phase, (ii) H₂O
53 molecules mostly as vapor and ice, (iii) OH⁻ or H₂O in silicate minerals, melts, and glasses,
54 (iv) C-bonded H in organic compounds and (v) minor volatile species, such as NH₃ and H₂S.
55 Today, water is present as ice, vapor, and/or liquid on Earth and other planets, dwarf planets,
56 moons, and comets. The presence of OH⁻ or H₂O in the silicate minerals comprising primitive
57 meteorites attests to the past existence of water on their asteroidal parent bodies and
58 constitutes an invaluable record of the distribution and composition of water in the
59 protoplanetary disk.

60 The H isotopic composition of water (expressed as D/H or per-mil deviations relative
61 to Vienna standard mean ocean water, VSMOW, as $\delta D = [(D/H_{\text{sample}}/D/H_{\text{VSMOW}}) - 1] \times 1000$,

62 where $D/H_{\text{VSMOW}} = 155.76 \times 10^{-6}$) in planetary objects is generally thought to increase with
63 increasing heliocentric distance, from H_2O that was in isotopic exchange with D-poor H_2 near
64 the sun ($D/H \approx 21 \times 10^{-6}$; Geiss and Gloeckler, 2003), to intermediate D/H ratios in inner
65 solar system objects (e.g., Earth's ocean, $D/H \approx 156 \times 10^{-6}$), to D-enriched outer solar system
66 objects such as comets (D/H up to 530×10^{-6} in comet 67P/Churyumov-Gerasimenko;
67 Altwegg et al., 2014). Determining the precise abundances and isotopic compositions of H in
68 meteorites—as representative samples of asteroids—is thus of fundamental importance for
69 understanding the source(s) of water in the asteroid-forming regions (Alexander et al., 2012;
70 Piani et al., 2015, 2018; Piani and Marrocchi, 2018) and potential radial mixing during the
71 evolution of the protoplanetary disk (Vacher et al., 2016; Piani et al., 2018).

72 The origin of water on Earth and other telluric planets is also a long-standing question
73 (Alexander, 2017; McCubbin and Barnes, 2019). Although it is commonly assumed that the
74 building blocks of the terrestrial planets derived from a cosmochemical reservoir best
75 represented by enstatite chondrites, they accreted in a region where temperatures were too
76 high for water and other volatiles to condense (Albarede, 2009). Therefore, these bodies are
77 expected to be highly volatile-depleted, though they have tenuous atmospheres and water is
78 largely present on Earth and Mars (Taylor, 2013), and possibly in the Venusian and
79 Mercurian atmospheres (Smrekar and Sotin, 2012). For Earth, this paradox is generally
80 bypassed by invoking the late accretion of 1–4 wt% of hydrated carbonaceous chondrites (i.e.,
81 CI-CM chondrites), which arrived at Earth due to the formation and subsequent migration of
82 Jupiter (Morbidelli et al., 2000; Marty, 2012; Marty et al., 2016; Raymond and Izidoro, 2017).

83 Such a conclusion is highly dependent on our ability to precisely quantify the bulk H
84 contents and D/H ratios of extraterrestrial materials. This point is not trivial since, despite
85 being commonly measured, H is the most prone **element** to terrestrial contamination (except
86 for noble gases; Protin et al., 2016). For instance, it was recently shown that Martian

87 meteorites suffer rapid (<1 yr) terrestrial weathering and contamination that largely affect the
88 water contents and H isotopic compositions of minerals exposed at their surface (Stephant et
89 al., 2018). Moreover, water and volatile hydrocarbons (CH₂ and CH₃) quickly adsorb onto
90 rock surfaces, raising potential H and C contamination issues during sample preparation
91 (Salisbury et al., 1991). This implies that the H budget estimated for a given meteorite is a
92 complex mixture between indigenous hydrogen in minerals and organics and exogenous H
93 due to terrestrial weathering and adsorption. Corrections for exogenous H could be achieved
94 by performing (i) stepwise heating measurements to decipher between adsorbed, alteration,
95 and indigenous water (Robert and Epstein, 1982; Kerridge, 1985) or (ii) sample degassing
96 under vacuum at low temperature (e.g., Vacher et al., 2016). Bulk H budget reconstructions
97 from stepwise heating experiments are difficult due to the presence of different water-bearing
98 minerals characterized by variable contamination susceptibilities that release adsorbed water
99 at various temperatures. Furthermore, there is no consensus on a procedure to remove
100 adsorbed water from both terrestrial and extraterrestrial materials, and different studies have
101 employed various temperatures (70–350 °C) and durations (2–480 h) of heating under
102 vacuum (Savin and Epstein, 1970; Girard et al., 2000; Gong et al., 2007; Lupker et al., 2012).
103 Of note, a recent estimation of the bulk H contents and isotopic compositions of a large set of
104 chondrites was performed without a pre-degassing protocol (Alexander et al., 2012), and
105 therefore may have overestimated the H budget of primitive meteorites. In addition to
106 terrestrial contamination, meteorites experienced parent-body alteration processes
107 (hydrothermal alteration and thermal metamorphism; Brearley, 2006; Huss et al., 2006;
108 Marrocchi et al., 2018) whose influence on their pre-accretion H characteristics remains
109 unclear.

110 Here, to better understand the origin of hydrogen accreted by chondrites, we report the
111 H concentrations and isotopic compositions of a large set of carbonaceous and ordinary

112 chondrites. To test the influence of laboratory pre-degassing temperature, we selected
113 chondrites that have been previously characterized (C2-ung. = Tagish Lake; CI = Orgueil and
114 Alais; **CY = Yamato 980115; King et al., 2019a**; CM = Murchison, Mighei, and Murray; CV
115 = Allende, Bali, Grosnaja, Kaba, and Vigarano; Robert and Epstein, 1982; Kerridge, 1985;
116 Pearson et al., 2001; Alexander et al., 2012). As CM chondrites represent 25% of all
117 carbonaceous chondrite falls (Gounelle et al., 2005), several new CMs have recently been
118 described. We thus report the H concentrations and isotopic compositions of new CM falls
119 (Aguas Zarcas, Maribo and Munkundpura; Haack et al., 2012; Rudraswami et al., 2019) and
120 finds (Lonewolf Nunataks (LON) 94101 and Jbilet Winselwan; Lindgren et al., 2013; **King et**
121 **al., 2019b**). In addition, several ordinary chondrites and a CK-type carbonaceous chondrite
122 covering the entire range of thermal metamorphism (Tait et al., 2014) were analyzed to test
123 the influence of parent-body metamorphism on H characteristics (L3 = Grosvenor Mountains
124 (GRO) 95502, CK4 = Karoonda, L4 = Saratov, LL/L4 = Bjurbole, H4 = Sainte Marguerite,
125 and H6 = Kernouve). We use our results to discuss the nature of primordial components
126 accreted by chondrites and the influence of post-accretion effects, either in asteroidal bodies
127 or at Earth's surface.

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134 2. Materials and method

135

136 2.1 Bulk measurements of H contents and isotope compositions

137

138 Hydrogen concentrations, [H], and isotopic compositions, D/H, of the different
139 meteoritic samples were determined on-line using the Thermo Scientific EA IsoLink -
140 deltaV IRMS System at CRPG laboratory (Nancy, France) according to the procedure
141 developed by Lupker et al. (2012). First, meteoritic samples were crushed into powder in an
142 agate mortar, weighed in tin capsules (CM, CI, **CY** and Tagish Lake: 1–3 mg; CV, CK, and
143 OC: 4–10 mg), and the samples and capsules were loaded into a sample carousel and
144 degassed under vacuum at 120 °C for 48 h in a degassing canister (Lupker et al., 2012;
145 Gaudin et al., 2015; Fig. 1A); degassing for less than 48 h was shown to result in increased
146 variability of water contents and isotopic compositions due to remaining absorbed water
147 (Lupker et al., 2012). After dehydration, the degassing canister was opened in a dry, N₂-
148 flushed glove box and the sample carousel was transferred into a sealed auto-sampler
149 initially pre-flushed with He (Fig. 1B). The auto-sampler was then connected to the EA and
150 pumped out during 20 min before opening it to the reduction column as a precautionary step
151 to suppress any potential rehydration of the samples in the event of atmospheric contact
152 between degassing and analysis.

153 The samples were combusted at 1450 °C (**required temperature to ensure rapid sample**
154 **reduction and to prevent any CH₃ formation**) on a glassy carbon reaction tube filled with
155 glassy carbon chips and itself placed into a ceramic liner to reduce hydroxyls (OH) **released**
156 **by the samples into H₂ according to the reaction: 2OH + C (unlimited in reactor) → 2CO + H₂**
157 (Fig. 1C). The produced gases were separated on a chromatographic column maintained at 60
158 °C and the extracted H₂ was introduced into the mass spectrometer through an open-split and

159 its D/H isotopic composition **was** analyzed (Fig. 1D). The extracted H₂ was corrected for H₃⁺
160 contributions using in-house H₂ standard injections and [H] and D/H were determined by
161 comparison with seven internal standards routinely included during the analyses: (i)
162 phlogopite (Mica-Mg, 0.29 wt.% H, D/H = 143.8 × 10⁻⁶), (ii) muscovite (MuscD65, 0.474
163 wt.% H, D/H = 142.6 × 10⁻⁶), (iii) a fine-grained marine sediment from the Bay of Bengal
164 (SO188, 0.489 wt.% H, D/H = 142.7 × 10⁻⁶), and (iv) four basalts with different H
165 concentrations (CYP78, 0.14 wt.% H; JB3-GSJ, 0.20 wt.% H; SR2-02, 0.27 wt.% H; and
166 GMR-KS12, 0.42 wt.% H). Errors on hydrogen concentrations, calculated as twice the
167 standard deviation divided by the average, increase with decreasing standard water content
168 following the negative power law 2SD / [H] = 0.05 × [H]^{-0.43}, which was used to estimate the
169 2σ sample errors reported in Table 1. Hydrogen isotopic compositions are reported as D/H
170 ratios or as δD values (‰) relative to VSMOW. The reproducibility of this method is better
171 than 15% (2σ) for [H] and 0.5 × 10⁻⁶ for D/H (or 5‰ for δD).

172

173 2.2 Bulk measurements of C and N concentration and isotopic compositions

174

175 Carbon and nitrogen concentrations ([C] and [N], respectively) and carbon isotopic
176 compositions (δ¹³C, ¹³C/¹²C variations reported relative to Vienna Pee Dee Belemnite, VPDB,
177 where ¹³C/¹²C_{VPDB} = 0.01123720) of the different meteoritic samples were determined on-line
178 using the Thermo Scientific EA IsoLink - deltaV IRMS System at the CRPG laboratory.
179 Samples were crushed into fine powder and wrapped in tin capsules (2–29 mg). The capsules
180 were loaded in a specific auto-sampler connected to the EA and pumped out during 20 min
181 before opening it to the oxidation column. Samples were then **introduced into an oxidation**
182 **reactor made** of a quartz tube filled with chromium oxide, pure copper, and silvered cobalt
183 oxide **and heated at 1020 °C. A controlled pulse of oxygen causes the oxidation of tin**

184 capsules leading to an exothermic reaction that raises the sample temperature to about
185 1800°C. The produced gases (N₂, CO₂) were separated on a chromatographic column
186 maintained at 70 °C and the carbon isotopic composition of the extracted CO₂ was measured
187 with a Thermo Scientific Delta V Advantage continuous flow IRMS. Carbon isotopic
188 composition were determined by comparison with two internal and two international
189 standards routinely included during the analysis: (i) BFSd ($\delta^{13}\text{C} = -21.5\text{‰}$), (ii) CRPG_M2
190 ($\delta^{13}\text{C} = -24.98\text{‰}$), (iii) NBS22 ($\delta^{13}\text{C} = -30.03\text{‰}$) and (iv) USGS24 ($\delta^{13}\text{C} = -16.1\text{‰}$). Values
191 are quoted in the delta notation in ‰ relative to V-PDB and the reproducibility was better
192 than 0.2‰. Four internal standards were used to calculate [N] and [C]: (i) BFSd (0.53 wt.%
193 C), (ii) CRPG_M2 (0.408 wt.% C), (iii) Eurovector Synthetic Soil Mix #1 (0.216 wt.% N, 3.5
194 wt.% C) and (iv) Eurovector Synthetic Soil Mix #4 (0.048 wt.% N, 2.417 wt.% C). Errors
195 (2σ) on [N] are estimated to be 10% and 30% for samples containing more and less than 0.08
196 wt.% N, respectively. For carbon analyses, 2σ errors are expected to be 2% for [C] and 2‰
197 for $\delta^{13}\text{C}$.

198

199 3. Results

200

201 3.1 Hydrogen characteristics of chondrites

202

203 Results from hydrogen extractions and associated D/H isotopic analyses are presented
204 in Table 1 and Figure 2. For most samples (excluding C2-ung. Tagish Lake and CI Alais for
205 their D/H variability), replicate aliquots of the same meteorite exhibit good reproducibility,
206 with 2σ standard deviations less than 15% for [H] and 5‰ for δD .

207 Hydrogen abundances of CI, **CY** and CM chondrites vary widely, but exhibit
208 comparable ranges, from 0.47 to 1.01 wt.% H (Fig. 2A). The lowest hydrogen contents were
209 measured in CM Jbilet Winselwan, possibly due to dehydration during parent-body
210 metamorphism (King et al., 2019b). **CMs are depleted in deuterium compared to CIs (D/H_{CM}
211 $= 131\text{--}170 \times 10^{-6}$ vs. $D/H_{CI} \approx 177 \times 10^{-6}$) but show similar range of D/H values as the **CY**
212 **Antarctica Y-980115 ($D/H_{CY} = 139 \times 10^{-6}$, Fig. 2B). The low hydrogen content of Y-980115**
213 **(0.71 wt.% H) relative to CI-CM chondrites and its D-poor isotopic composition might be**
214 **might be a consequence of (i) thermal metamorphosed at temperature $> 500^\circ\text{C}$ (King et al.,**
215 **2019a) and/or (ii) terrestrial weathering (e.g., Alexander et al., 2018a). The ungrouped C2**
216 **carbonaceous chondrite Tagish Lake exhibits a hydrogen content similar to the lowest value**
217 **measured for CM chondrites (0.43 wt.% H), but this unique carbonaceous chondrite is the**
218 **most D enriched ($D/H = 225 \times 10^{-6}$, Fig. 2A and B), consistent with previous measurements**
219 **(Pearson et al., 2001; Alexander et al., 2012). CV chondrites (0.06–0.19 wt% H, Fig. 2A)**
220 **contain lower hydrogen concentrations than CM and CI chondrites, whereas their hydrogen**
221 **isotopic compositions are highly variable ($D/H = 139\text{--}161 \times 10^{-6}$, Table 1) over a range**
222 **similar to that of CM chondrites (Fig. 2B). Ordinary chondrites and CK-type carbonaceous**
223 **chondrites present the lowest hydrogen concentrations (≤ 0.03 wt.% H, excluding Grosvenor****

224 Mountains (GRO) 95502, an Antarctic find possibly affected by terrestrial weathering; Fig.
225 2A), and have relatively D-poor hydrogen isotopic compositions ($D/H = 134\text{--}144 \times 10^{-6}$, Fig.
226 2B).

227

228 3.2 Nitrogen and carbon characteristics of chondrites

229

230 Table 2 summarizes the carbon and nitrogen contents and carbon isotopic
231 compositions of the analyzed chondrites. CI, **CY** and CM chondrites contain the highest N
232 and C contents in this study, with 0.09–0.17 wt.% N and 1.54–3.04 wt.% C in CMs and 0.09–
233 0.21 wt.% N and 3.06–3.35 wt.% C in CIs (Fig. 3). They both show comparable ranges of
234 carbon isotopic compositions, with $\delta^{13}\text{C}$ ranging from -16 to -13‰ and -14 to -4‰ for CIs
235 and CMs, respectively. The nitrogen and carbon concentrations in Tagish Lake (0.11 wt.% N
236 and 2.14 wt.% C) are comparable to those in CM chondrites, but with a more ^{13}C -enriched
237 isotopic composition ($\delta^{13}\text{C} \approx +5\text{‰}$). Compared with these C- and N-rich chondrites, CV, CK,
238 and ordinary chondrites are depleted in N (≤ 0.03 wt.%) and C (≤ 1.42 wt.%), and have **lower**
239 carbon isotopic compositions than hydrated chondrites, with $\delta^{13}\text{C} \approx -27$ to -18‰ .

240

241

242

243 **4. Discussion**

244

245 4.1 Effect of terrestrial contamination in carbonaceous chondrites

246

247 4.1.1 Influence of pre-degassing temperature on the total H budget

248

249 Immediately after falling to Earth's surface, meteorites are exposed to terrestrial
250 moisture and precipitation that could significantly affect their H characteristics, even in dry
251 environments such as hot deserts (Stephant et al., 2018). Consequently, bulk meteorite H
252 budgets are complex mixtures of indigenous hydrogen, terrestrial weathering, and adsorbed
253 water hosted by different minerals/phases with variable thermal behaviors.
254 Thermogravimetric analysis and derivate curves obtained in the range 25–1200 °C (Garenne
255 et al., 2014; King et al., 2015; Gilmour et al., 2019) suggest a complex thermal evolution of
256 H-bearing phases with the following release temperatures: (i) 25–200 °C for absorbed H₂O,
257 (ii) 200–400 °C for water in hydroxides and H-bearing organic matter, (iii) 400–770 °C for
258 OH⁻ in phyllosilicates, and (iv) 770–900 °C for H₂O in sulfates. Although suspected to occur
259 to some degree at 200–400 °C, it has been proposed that adsorbed terrestrial water is mainly
260 released from hydrated carbonaceous chondrites at 25–250 °C (Boato, 1954; Kerridge, 1985;
261 Baker et al., 2002; Garenne et al., 2014; Gilmour et al., 2019).

262 Alexander et al. (2012, 2013) reported H concentrations and isotopic compositions for
263 a large set of carbonaceous chondrites that were stored in desiccators at room temperature for
264 days to weeks prior to analyses. Their data define a positive correlation in a D/H vs. 1/H
265 diagram (Fig. 4) that reveals the increasing contribution of H-bearing, D-rich, insoluble
266 organic matter (IOM; Alexander et al., 2007; Alexander et al., 2012) to the total H budget, the
267 largest contributions coming from phyllosilicates in hydrated chondrites. Our data define a

268 similar positive trend, but with a statistically different slope (Fig. 4) that reveals the influence
269 of the pre-degassing temperature on observed meteorite H features. Thereby, our bulk H
270 measurements of samples also measured by Alexander et al. (2012) demonstrate that pre-
271 degassed CM and CI chondrites contain ~10–30% and ~40% less H, respectively, than non-
272 degassed samples (Fig. 4). Several reasons can be invoked to explain such important
273 differences: (i) the significant loss of indigenous hydrogen induced by the pre-degassing
274 protocol, leading to an underestimated H budget, (ii) sample heterogeneity, the effects of
275 which are apparent due to the small amounts of material analyzed, and/or (iii) the intended
276 removal of absorbed terrestrial water.

277 To test these different scenarios, we estimated the H isotopic composition of the
278 hydrogen lost during pre-degassing at 120 °C for 48 h using the following mass balance
279 equation:

280

$$281 \quad [H]_{\text{lost}} \times D/H_{\text{lost}} = [H]_{25\text{ }^{\circ}\text{C}} \times D/H_{25\text{ }^{\circ}\text{C}} - [H]_{120\text{ }^{\circ}\text{C}} \times D/H_{120\text{ }^{\circ}\text{C}} \quad (1)$$

282

283 where $[H]_{25\text{ }^{\circ}\text{C}}$, $[H]_{120\text{ }^{\circ}\text{C}}$, $D/H_{25\text{ }^{\circ}\text{C}}$, and $D/H_{120\text{ }^{\circ}\text{C}}$ correspond to the bulk H concentrations and
284 isotopic compositions of the samples determined at room temperature (i.e., 25 °C) and after
285 pre-degassing at 120 °C for 48 h, respectively. The hydrogen lost during pre-degassing
286 corresponds to $[H]_{\text{lost}} = [H]_{25\text{ }^{\circ}\text{C}} - [H]_{120\text{ }^{\circ}\text{C}}$. Consequently, if the H lost during pre-degassing
287 was adsorbed terrestrial water, its calculated H isotopic composition should be (i) similar
288 from one sample to the next and (ii) within the range of terrestrial values. On the other hand,
289 large H isotopic variations are expected if the H lost resulted from sample heterogeneity or
290 the removal of indigenous hydrogen. The H isotopic compositions of pre-degassed H range
291 from $D/H \sim 150\text{--}155 \times 10^{-6}$ for Murchison, Mighei, Murray, and Orgueil. These values are in
292 the range of H isotopic ratios reported for terrestrial hydrosphere reservoirs ($115\text{--}165 \times 10^{-6}$;

293 Lécuyer et al., 1998; Frankenberg et al., 2009; Clog et al., 2013), but differ strongly from the
294 (i) D-poor phyllosilicates that bear the signature of asteroidal water in CM and CI chondrites
295 (D/H $\sim 85\text{--}125 \times 10^{-6}$; Alexander et al., 2012; Piani et al., 2018, 2019) and (ii) D-rich organic
296 matter (D/H $\sim 265\text{--}310 \times 10^{-6}$; Alexander et al., 2007). Consequently, we interpret the
297 differences in H concentrations obtained with and without the pre-degassing routine as mostly
298 due to contamination of primitive meteorites by atmospheric moisture. **This scenario cannot,**
299 **however, explain the D/H ratio calculated of the H degassed from LON 94101, which display**
300 **a significantly higher D/H value compared to the other common samples ($\sim 400 \times 10^{-6}$).** By
301 analogy with other CM chondrites, the removal of indigenous H during pre-degassing would
302 result in a D-poor signature for $[\text{H}]_{\text{lost}}$, at odds with our calculations. Alternatively, the
303 preferential degassing of H from D-rich IOM or **H-bearing organic compounds (i.e., small**
304 **hydrocarbons or molecules found in the soluble part of the meteoritic organic matter)** might
305 explain such high D/H values **and contributed to the H budget removed during our pre-**
306 **degassing procedure. However, given that (i) the H isotopic compositions of $[\text{H}]_{\text{lost}}$ are D-poor**
307 **compared to organic molecules in carbonaceous chondrites (Pizzarello et al., 1994; 2006;**
308 **Yamashita & Naraoka, 2014), (ii) stepwise pyrolysis performed by Robert and Epstein (1982)**
309 **on Murchison and Murray indicates that only few amount of carbon (~ 0.2 mol% of the total**
310 **C) were lost compared to water below 200 °C (11.5 and 10.8 mol% of the total H,**
311 **respectively), and (iii) the decomposition of IOM likely occurs at 200–500 °C (Gilmour et al.,**
312 **2019; Remusat et al., 2019), we concluded that the amount of indigenous organics volatile**
313 **lost during pre-degassing is not significant and the D-rich compositions of the $[\text{H}]_{\text{lost}}$ by LON**
314 **94101 is rather a consequence of sample heterogeneity (Lindgren et al., 2013).**
315

4.1.2 Quantification of terrestrial contamination in carbonaceous chondrites

316

317

318 To estimate the influence of pre-degassing, we compare hydrogen concentrations and
319 isotopic compositions measured in different studies for CI Orgueil and CM Murchison as a
320 function of pre-degassing temperature. Murchison shows ~30% less H after pre-degassing at
321 200 °C compared to measurements performed without pre-degassing (Fig. 5A–C).
322 Isotopically, this sample does not show a large change, with D/H value of $\sim 150 \times 10^{-6}$ (Fig.
323 5C). In contrast, a sample of Orgueil pre-degassed at 120–350 °C shows (i) a H loss of up to
324 ~40–80% and (ii) a D/H increase up to $\sim 190 \times 10^{-6}$ relative to samples measured without pre-
325 degassing (D/H $\sim 150 \times 10^{-6}$; Figs. 5B–D). This disparity between CM and CI chondrites
326 probably derives from the fact that the bulk D/H values of CMs, which result from the
327 combined contributions of D-poor water and D-rich OM, are fortuitously close to the D/H
328 values imparted by atmospheric contamination (D/H_{CM} $\sim 142 \times 10^{-6}$ vs. D/H<sub>[H]_{lost} $\sim 150 \times 10^{-6}$),
329 and contamination thus produces only minor changes. In contrast, water and OM in CIs are
330 D-rich compared to CMs (Busemann et al., 2006; Alexander et al., 2007; Piani et al., 2018,
331 2019), with bulk H isotopic compositions (D/H_{CI} $\sim 177 \times 10^{-6}$) markedly higher than that of
332 the contaminating water. Consequently, the effects of atmospheric moisture are more
333 perceptible in CIs than in CMs.</sub>

334 To better quantify the influence of terrestrial contamination, we compared [H]_{lost} (=
335 [H]_{25 °C} – [H]_{120 °C}) for different chondrites as a function of the H concentration determined
336 after pre-degassing at 120°C. We thusly obtained separate linear trends for CI, CM and CV
337 carbonaceous chondrites, indicating that meteorites containing more H are more prone to
338 contamination (Fig. 6). Interestingly, CM- and CI-type chondrites seem to define a single
339 trend ($r^2 = 0.70$; MSWD = 1.5), suggesting that CIs would be more affected by H
340 contamination than CMs (Fig. 6). The H concentrations of Tagish Lake are highly

341 heterogeneous ($H = 1.37$ and 0.85 wt.%, from Pearson et al., 2001, and Alexander et al.,
342 2012, respectively) and have been shown to depend strongly on the studied lithology ($H =$
343 0.74 – 0.94 wt.%; Alexander et al., 2012); however, as we do not have the petrological details
344 of the Tagish lake pieces measured in our study, we omit Tagish Lake from further
345 discussion.

346 The difference in the amount of H loss between CI and CMs probably reflects
347 secondary mineralogical diversity between these two groups of chondrites (Fig. 6). CI
348 chondrites are mainly dominated by serpentine and saponite minerals, the latter being clay
349 minerals with the capacity to absorb significant amounts of water into their inter-layers
350 (Tomeoka and Buseck, 1988; King et al., 2015). Thus, it is likely that CI chondrites contains
351 indigenous inter-layer water that could be removed alongside with terrestrial water during
352 pre-degassing, leading to an underestimation of the H budget in CI chondrites. Nevertheless,
353 it appears that the high abundance of inter-layer water in Orgueil is probably related to its
354 long residence time on Earth (since 1864) and it is likely that this meteorite was devoid of
355 inter-layer water prior to its arrival on Earth (Baker et al., 2002). In addition, stepwise heating
356 performed on Orgueil demonstrated that water released at temperature lower than 200 °C is
357 characterized by terrestrial-like values (i.e., $\Delta^{17}\text{O} = 0$ ‰ and $D/H = 153 \times 10^{-6}$), supporting
358 that only minor indigenous water was present in the inter-layers of Orgueil (Robert and
359 Epstein, 1982; Baker et al., 2002). In contrast, CM chondrites are almost saponite-free, and
360 are instead dominated by Fe-Mg-rich serpentine that retains less absorbed water in its silicate
361 layers than saponite (Barber 1981; Howard et al., 2015). It is unclear why the CV trend is
362 separate from the CI-CM trend, but the amount of H lost during pre-degassing suggests that
363 this group of chondrites is also very sensitive to terrestrial alteration and can absorb similar H
364 proportions as CM chondrites (Fig. 6). Phyllosilicates are modally less abundant in CV (up to
365 4 vol.%; Howard et al., 2010) than in CM chondrites (~ 75 vol.%; Howard et al., 2015).

366 Assuming that phyllosilicates are the main storage sites for atmospheric moisture in hydrated
367 carbonaceous chondrites, water adsorption should be vastly lower in CV than in CM
368 chondrites (even accounting for the mineralogical differences of their phyllosilicates).
369 Alternatively, CI and CM phyllosilicates are almost water saturated—i.e., their water
370 adsorption capacity is limited—whereas CV phyllosilicates, despite being less abundant, have
371 more adsorption sites available for accommodating atmospheric moisture. Whatever the
372 reason, it is apparent that terrestrial contamination can greatly affect the H budgets of the
373 most H-poor (CV-like) and H-rich (CI-like) chondrites alike (Figs. 5 and 6). The bulk H
374 contents and D/H ratios of chondrites should thus be measured after pre-degassing under
375 vacuum at ≥ 120 °C. The consequences of correcting the H compositions of chondrites for
376 terrestrial contamination are discussed in §4.3.

377

378 4.2 The influence of thermal metamorphism

379

380 4.2.1 CV-type carbonaceous chondrites

381

382 CV chondrites are divided into three subgroups partly reflecting their various aqueous
383 and thermal alteration histories: reduced, CV_{Red}; oxidized Allende-like, CV_{OxA}; and oxidized
384 Bali-like, CV_{OxB}. Of the oxidized CVs, the CV_{OxB} subgroup is the most extensively altered
385 and contains abundant phyllosilicates and chondrules replaced by phyllosilicates and
386 magnetite. In contrast, the CV_{OxA} and CV_{Red} subgroups contain minor phyllosilicates and
387 abundant Ca-Fe-rich secondary phases such as andradite, hedenbergite, and kirschsteinite
388 (Krot et al., 2004; Brearley, 2006; Howard et al., 2010; Ganino and Libourel, 2017). CV3s
389 have experienced various degrees of metamorphism that are partly overprinted by subsequent
390 aqueous alteration and further obscured by brecciation (Bonal et al., 2006). Based on the

391 maturation grade of their organic matter, Bonal et al. (2006) assigned a range of petrographic
392 degrees for the CV chondrites Kaba (~3.1), Vigarano (3.1–3.4), Bali and Grosnaja (~3.6), and
393 Allende (>3.6), although they highlighted the occurrence of clasts of different petrologic types
394 within individual meteorites (e.g., Bali). The H-, C-, and N-concentrations and isotopic
395 compositions measured for these CV chondrites reflect the complex inputs of brecciation and
396 aqueous and thermal alteration, with no clear behavior of these elements and isotopes with
397 increasing petrologic type (Figs. 6 and 7a; Tables 1 and 2). Nevertheless, the most
398 metamorphosed CV_{OxA}, Allende, presents the lowest H concentrations, with its most H-
399 depleted lithologies (Yang and Epstein, 1983) having similar H contents as the
400 metamorphosed CK4 Karoonda (Fig. 7a). This result suggests that thermal metamorphism of
401 CV chondrites could induce a significant H loss for petrologic types >3.6 (Fig. 7a).

402

403 4.2.2 Ordinary chondrites

404

405 Ordinary chondrites (OCs) from the H, L, and LL groups exhibit pervasive imprints of
406 thermal metamorphism and, to a lesser degree, aqueous alteration. Semarkona (LL3.00) and
407 Bishunpur (LL3.15) are among the rare unequilibrated OCs to contain hydrated minerals
408 (smectites) and other secondary phases (calcites) thought to have formed on their parent body
409 from an oxidizing fluid (Alexander et al., 1989). Water measured in these chondrites is
410 enriched in D, as attested by (i) their bulk D/H values up to 600×10^{-6} (McNaughton et al.,
411 1982; Yang and Epstein, 1983) and (ii) *in-situ* measurements of D/H values up to 1800×10^{-6}
412 (Deloule and Robert, 1995; Piani et al., 2015). Such high D enrichments were interpreted
413 either as inherited from the accretion of D-rich ice that originated in cold regions of the solar
414 system (Deloule and Robert, 1995; Piani et al., 2015) or as the result of parent-body processes
415 (Alexander et al., 2012). Bleached chondrules found in LL3, L3, and more rarely in H3

416 chondrites attest that aqueous alteration was widespread in OCs, but was mostly erased by
417 subsequent thermal metamorphism (Grossman et al., 2000). This later metamorphic event
418 resulted in high degrees of equilibration of the three OC sub-groups spanning petrological
419 types 3–7 and equilibration temperatures up to 1000 °C (Tait et al., 2014).

420 The five OCs measured herein span the range of H concentrations reported in OCs,
421 with the most metamorphosed chondrites (H6 Kernouve) being the most depleted in hydrogen
422 (Fig. 7B). These five OCs exhibit D/H ratios similar to those previously measured in
423 equilibrated ordinary chondrites ($\sim 130\text{--}160 \times 10^{-6}$), and we observe no strong differences
424 between groups or petrological types (Fig. 7B). Furthermore, we do not observe D
425 enrichments with increased parent-body thermal metamorphism (Fig. 7B), in agreement with
426 previous observations (Robert et al., 1979; McNaughton et al., 1982). This is at odds with the
427 assumption that water in ordinary chondrites became increasingly enriched in deuterium due
428 to the oxidation of iron and subsequent loss of isotopically light H₂ (Alexander et al., 2010,
429 2012; Sutton et al., 2017). This assumption was initially proposed to explain the deuterium
430 enrichment of IOM in OCs with increasing degree of metamorphism, the organic matter
431 thereby being enriched by isotopic exchange with the remaining D-rich water (Alexander et
432 al., 2010). However, given that both the remaining water and OM should have been enriched
433 in D by this process, the bulk D/H ratios of OCs should increase with increasing degree of
434 thermal metamorphism. The absence of such D enrichments in the bulk chondrites thus
435 contradicts the implication of water in the D enrichment of the OM, instead supporting the
436 proposal that ordinary chondrites contain a thermally recalcitrant D-rich organic matter
437 component (Remusat et al., 2016). Overall, thermal metamorphism in OCs seems to have
438 mainly decreased the bulk-rock hydrogen contents without strongly influencing the hydrogen
439 isotopic ratios for petrological types ≥ 3.2 (Fig. 7B).

440 The percentage of fayalite (%Fa) in ordinary chondrites increases from H to L to LL
441 groups and is considered a robust proxy of the degree of hydration (i.e., the water content) of
442 their respective parent bodies (Rubin, 2005; Florin et al., 2020). However, the lack of
443 variation between the H contents of the different groups of OCs (Fig. 7B) suggests that they
444 could have accreted similar amounts of water. This could be interpreted as evidence for pre-
445 accretion hydration processes in different regions of the protoplanetary disk. Whereas there is
446 no clear limit between the H contents of unequilibrated and equilibrated OCs, the observed
447 decrease in H contents with increasing metamorphism is in line with the progressive increase
448 of %Fa and the loss of Fe-Ni metal during metamorphism (Florin et al., 2020).

449

450 4.3 Implication for the origin and abundance of H in chondrites

451

452 Our data suggest that terrestrial contamination can represent a significant part of the
453 measured chondritic H budget, which strongly depends on the analytical protocol and
454 particularly the pre-degassing procedure. The effects of terrestrial contamination are
455 especially important for **chondrites** with the **highest** indigenous hydrogen contents **within a**
456 **given chondrite group** (Figs. 4 and 6). However, within errors, we observe similar positive
457 correlations and zero intercepts ($D/H \sim 100 \times 10^{-6}$ or $\delta D \sim -400\text{‰}$) between D/H and C/H for
458 CMs (Fig. 8) as previously reported (Alexander et al., 2012). These values represent the
459 isotopic composition of water accreted by the CM parent body (Alexander et al., 2012). This
460 similarity indicates that previous estimations of the D/H ratios of water in the region of CM
461 parent-body accretion (Alexander et al., 2012) were not significantly affected by terrestrial
462 contamination, as also attested by the consistency with *in-situ* measurements via secondary
463 ion mass spectrometry (SIMS; Piani et al., 2018). The D/H ratio of water accreted by CV
464 chondrites was only estimated recently from SIMS measurements (Piani and Marrocchi,
465 2018), and our bulk data confirm their observation that CV water is enriched in deuterium
466 compared to CM water (Fig. 8). If CMs and CVs accreted beyond the orbit of Jupiter as
467 commonly proposed (Kruijjer et al., 2017; Nanne et al., 2019), this suggests the existence of
468 different water-ice reservoirs, with CV water being enriched in heavy H and O isotopes (Piani
469 et al., 2018; Piani and Marrocchi, 2018; Marrocchi et al., 2018).

470 Among chondrites, CIs are of primary importance as their bulk chemical compositions
471 are similar to that of the solar photosphere, which is representative of the average solar system
472 composition (Lodders, 2003). In addition, CIs are generally considered to be the most
473 hydrated chondrites based on previous H measurements (Pearson et al., 2001; Alexander et
474 al., 2012) and their high abundances of secondary minerals ($\sim 97\%$; King et al., 2015).

475 Although it remains a possibility that a small amount of the released water below 120°C could
476 have been initially indigenous, our data reveal that CIs analyzed without pre-degassing are
477 essentially contaminated by terrestrial moisture, and we estimate their indigenous water
478 contents to be almost a factor of 2 lower than previously reported (0.93 wt.% H compared to
479 1.56 wt.% H; Pearson et al., 2001; Alexander et al., 2012), in agreement with results from
480 stepwise heating analyses (Boato, 1954; Robert and Epstein, 1982; Yang and Epstein, 1983;
481 Kerridge, 1985; Eiler and Kitchen, 2004). Consequently, CI chondrites have indigenous water
482 contents similar to those estimated for CM chondrites, suggesting that CIs and CMs accreted
483 similar proportions of water, although they contain vastly different proportions of matrix (65
484 vol.% and 100 vol.% in CMs and CIs, respectively; Alexander et al., 2018b and references
485 therein). CV chondrites comprising 40 vol.% matrix have H abundances roughly one tenth
486 those of CM and CI chondrites. Together, our results for CV-, CM-, and CI-type
487 carbonaceous chondrites argue against the suggested dependency of the bulk H concentration
488 on matrix content (Alexander et al., 2018b). Nevertheless, our data for C and N
489 concentrations (Fig. 3) are consistent with a matrix content dependency for these elements
490 (Alexander et al., 2018b). Furthermore, correcting for terrestrial water contamination in CIs
491 largely affects their D/H isotopic compositions, which is not the case for CMs (Fig. 5). This
492 implies that CIs do not have H isotopic ratios as close to that of Earth's water as previously
493 estimated based on measurements performed without pre-degassing. This conclusion was
494 reported nearly 40 years ago (Robert and Epstein, 1982; Kerridge, 1985), but was
495 overshadowed by recent studies that did not quantify the effects of terrestrial contamination
496 (Alexander et al., 2012). Consequently, the large contamination of CI water and its non-
497 terrestrial H isotopic composition should be accounted for in models of Earth's formation
498 based on the mixing of chondrite-like materials, as considering CIs as the putative source of
499 Earth's oceans would also affect the ^{54}Cr mass balance calculation (Warren, 2011; Dauphas,

500 2017). In such a framework, CM chondrites thus appear to be the most isotopically similar
501 extraterrestrial materials to the Earth's surficial hydrogen reservoir.

502

503 **5. Conclusions**

504

505 We have determined the bulk hydrogen concentrations and isotopic compositions of a
506 large set of chondritic samples (CI, CM, CV, CK, and ordinary chondrites) using an elemental
507 analyzer coupled to an isotope ratio mass spectrometer. Samples were pre-degassed under
508 vacuum at 120 °C for 48 h before measurements. Our main results are:

509 1) CI and CM chondrites and Tagish Lake define a correlation in a D/H vs. 1/H
510 diagram induced by the increasing contribution of insoluble organic matter to the total H
511 budget. This trend is statistically distinct from that defined by similar chondrites analyzed
512 without pre-degassing at ≥ 120 °C.

513 2) No clear D enrichment is observed with the increase of parent-body thermal
514 metamorphism for ordinary and CV-type carbonaceous chondrites.

515 From these results, we drew the following conclusions:

516 1) Chondrites are largely affected by terrestrial contamination, with the most hydrated
517 CI chondrites being the most prone to contamination because phyllosilicates are more
518 abundant in CIs compared to CMs and CVs. After correction of contamination, the primordial
519 H contents of CI, CM, and CV chondrites are significantly lower than generally reported and
520 is not related to their matrix modal abundance, implying that their respective parent bodies
521 accreted variable amounts of water-ice grains.

522 2) The D/H ratio of Orgueil is affected by terrestrial contamination, leading to an
523 important underestimation of its H isotopic composition. This result is important as it could

524 affects the amount of water delivery by CI chondrites in the models of Earth's formation
525 based on the mixing of chondrite-like materials.

526 3) Although based on a limited number of samples, thermal metamorphism suggests
527 that significant H were lost in CVs of petrological type >3.6. It also decreased the H contents
528 of ordinary chondrites, but minimally affected the D/H ratios of OCs of petrological type
529 ≥ 3.2 .

530 4) The absence of an increasing D/H ratio with increasing the metamorphism degree
531 for bulk ordinary chondrites does not support models in which water became enriched in
532 deuterium via the oxidation of iron and the loss of isotopically light H₂. On the contrary, this
533 observation supports the presence of thermally resistant D-rich organic matter in ordinary
534 chondrites.

535

536 **Acknowledgments**

537

538 We thank Mike Zolensky and Bernard Marty for providing samples. We also thank the
539 National Institute of Polar Research (Japan), the Natural History Museum of Denmark
540 (Copenhagen), the Muséum national d'Histoire naturelle (Paris, France), the Natural History
541 Museum of Vienna (Austria), and the Antarctic Search for Meteorites (ANSMET) program
542 for loaning samples. US Antarctic meteorite samples were recovered by ANSMET, funded by
543 NSF and NASA, and characterized and curated by the Department of Mineral Sciences of the
544 Smithsonian Institution and Astromaterials Curation Office at NASA Johnson Space Center.
545 This work was funded by l'Agence Nationale de la Recherche through grant ANR-14-CE33-
546 0002-01 SAPINS (PI Yves Marrocchi) and ANR-19-CE31-0027-01 HYDRaTE (PI Laurette
547 Piani) and by the European Research Council (PHOTONIS project, grant agreement No.
548 695618 to Bernard Marty). This is a CRPG contribution #2728.

549 **Research data**

550 Original data of this study are available in the supplementary file.

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798 **Figure captions**

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800 Fig. 1. Schematic representation of the analytical protocol used in this study. The samples
801 were first degassed under vacuum at 120 °C for 48 h in a degassing canister (A) before being
802 transferred into a sealed auto-sampler pre-flushed with He (B). Samples were combusted at
803 1450 °C (C) and the extracted H₂ was introduced into the mass spectrometer through an open
804 split and its D/H isotopic composition analyzed (D).

805

806 Fig. 2. (A) Average bulk hydrogen and water abundances (wt.%) for the different groups of
807 chondrites analyzed in this study. Half-filled markers and unfilled triangle represent samples
808 presumably affected by terrestrial weathering and/or metamorphism dehydration (Jbilet
809 Winselwan, Y-980115 and Gro 95502). (B) Distribution of the H isotopic compositions (bulk
810 D/H ratios and equivalent δD values) of chondrites ($n = 178$). The upper part of the graph
811 presents the H isotopic compositions measured in this study for each group of chondrites, and
812 the lower part presents values reported in literature: Boato (1954), Robert (1978), Robert et al.
813 (1979), Kolodny et al. (1980), Kerridge (1985), McNaughton et al. (1982), Robert and
814 Epstein (1982), Yang and Epstein (1983), Robert (2003), Pearson et al. (2001), and Alexander
815 et al. (2012).

816

817 Fig. 3. Relationship between bulk carbon and nitrogen abundances (wt.%) for all chondrites
818 analyzed. A positive trend (solid black line) is observed between the two parameters, with an
819 R^2 value of 0.77 (MSWD = 1.7). Dashed curves represent the 95% confidence interval of the
820 regression (errors are 2σ).

821

822 Fig. 4. Bulk hydrogen abundances (expressed as 1/H) and D/H isotopic ratios of CM
823 chondrites and the ungrouped Tagish Lake chondrite ('TL') from this study and Alexander et
824 al. (2012) and CM Paris (Vacher et al., 2016). Two positive trends (solid lines) with distinct
825 slopes but similar intercepts are observed for our data (red; $D/H = 70 (\pm 17) \times 1/H + 60 (\pm$
826 $24)$; 2σ , $R^2 = 0.91$, MSWD = 21) and the data from Alexander et al. (2012), excluding heated
827 CMs (gray; $D/H = 133 (\pm 28) \times 1/H + 28 (\pm 26)$; 2σ , $R^2 = 0.55$, MSWD = 466). Dashed
828 curves represent the 95% confidence intervals of the regressions (errors are 2σ).

829

830 Fig. 5. The effect of pre-degassing temperature on the measured (A, B) hydrogen abundances
831 and (C, D) hydrogen isotopic compositions of **CM Murchison** (red) and **CI Orgueil** (orange).
832 Measured hydrogen concentrations decrease and D/H ratios increase with increased pre-
833 degassing temperature for Orgueil. **Ranges of D/H values of D-rich organic matter (dark gray**
834 **area; Alexander et al., 2007), D-poor phyllosilicates (light gray area; Alexander et al., 2012;**
835 **Piani et al., 2018, 2019) and terrestrial hydrosphere (blue area; Lécuyer et al., 1998;**
836 **Frankenberg et al., 2009; Clog et al., 2013) are also represented for comparison.** Data from:
837 Boato (1954), Kolodny et al. (1980), Robert and Epstein ('R&E', 1982), Yang and Epstein
838 ('Y&E', 1983), Pearson et al. (2001), and Alexander et al. (2012).

839

840 Fig. 6. The amount of H lost during the pre-degassing procedure as a function of the
841 indigenous H content measured in CV-, CM-, and CI-type carbonaceous chondrites after pre-
842 degassing at 120 °C. The amount of pre-degassed H is calculated as the difference between
843 the hydrogen content measured in our samples after pre-degassing at 120 °C and that
844 measured in the same samples at room temperature (25 °C) using the same analytical
845 technique, but without pre-degassing (Pearson et al., 2001; Alexander et al., 2012) (**errors are**
846 **2σ**).

847

848 Fig. 7. Bulk D/H ratios as a function of bulk hydrogen abundances (expressed as 1/H) for all
849 reported analyses of (A) CV chondrites (this study; Boato, 1954; Robert and Epstein, 1982;
850 Kerridge, 1985; Pearson et al., 2001; Alexander et al., 2012) and (B) ordinary chondrites (this
851 study; Robert et al., 1979; McNaughton et al., 1982; Yang and Epstein, 1983; Alexander et
852 al., 2012) (errors are 2σ).

853

854 Fig. 8. Bulk hydrogen isotopic compositions of CV (black) and CM (red) chondrites as a
855 function of their bulk C/H ratios. Delta notation is used for H isotopic compositions to
856 facilitate comparison with Alexander et al. (2012). Solid lines and dashed curves represent
857 data regressions and their 95% confidence intervals, respectively (errors are 2σ).

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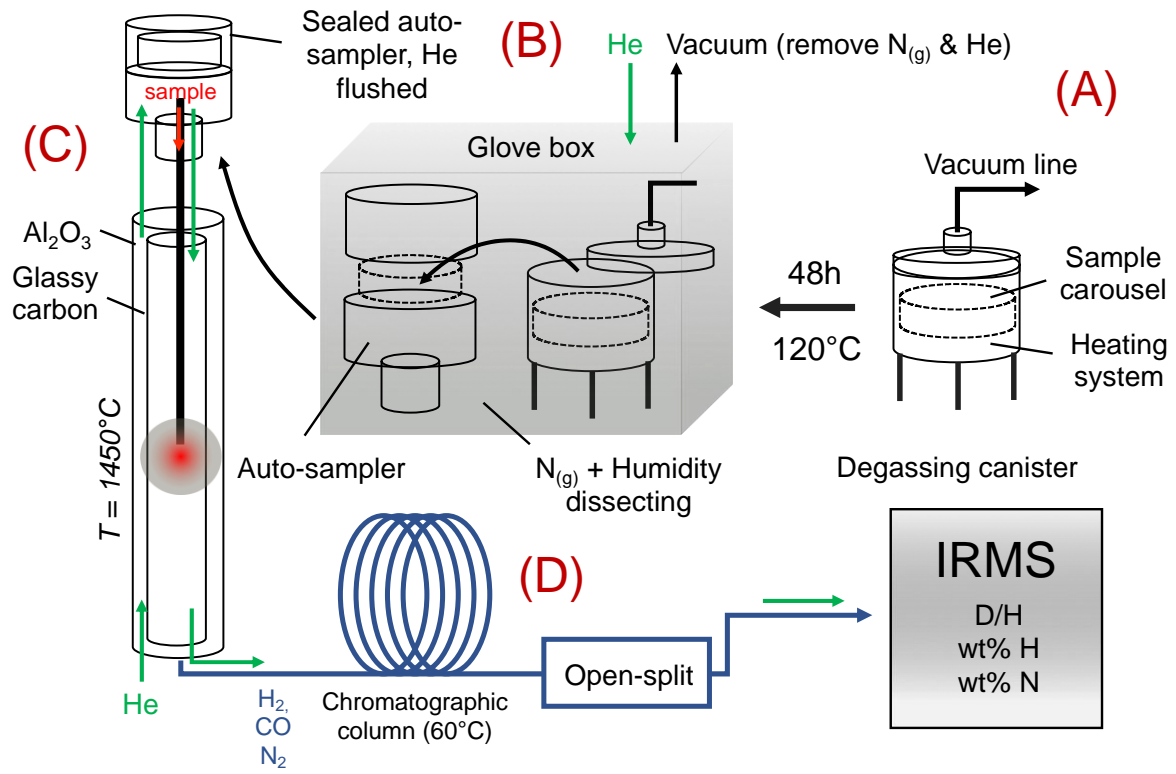
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Fig. 1

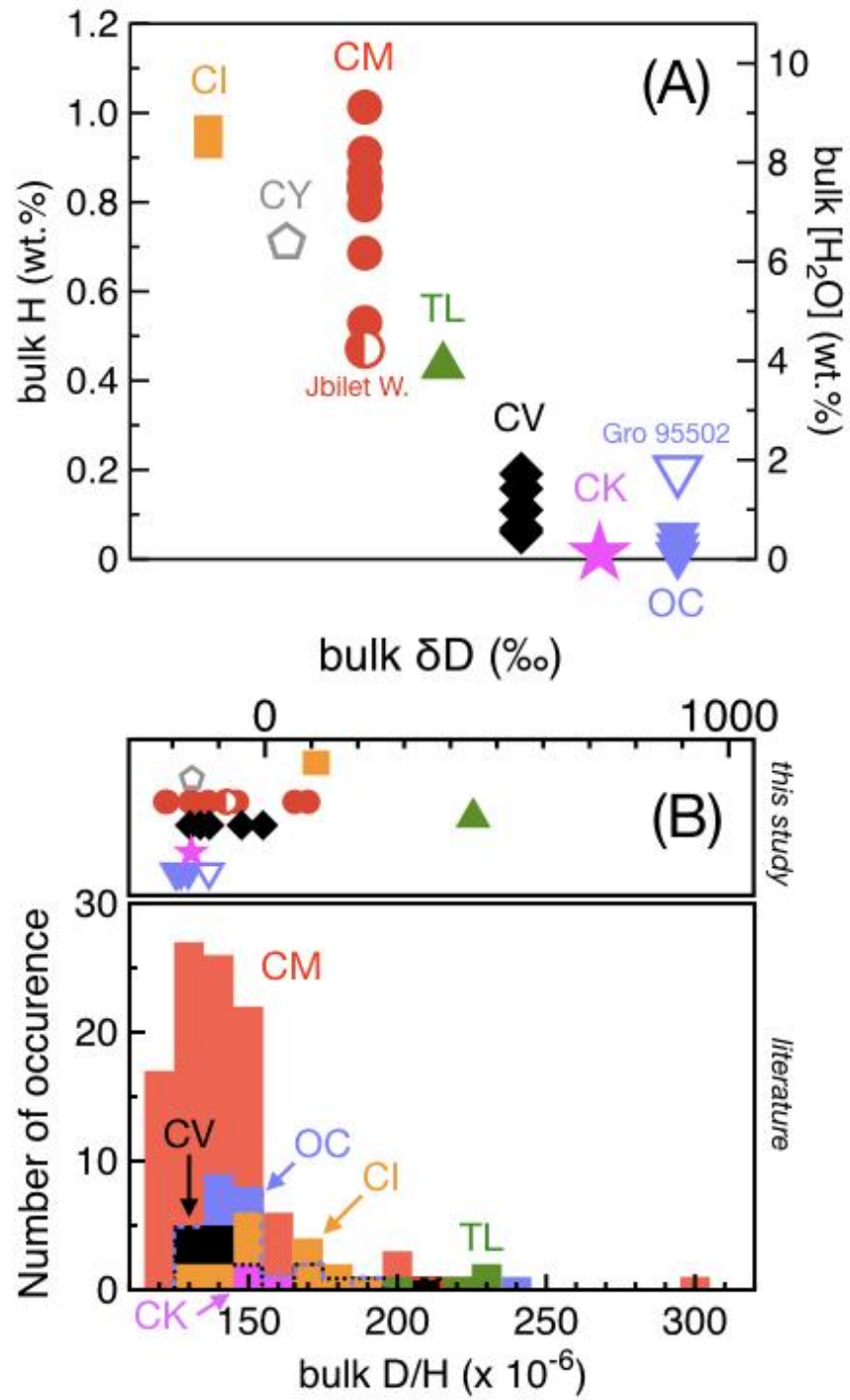


Fig. 2

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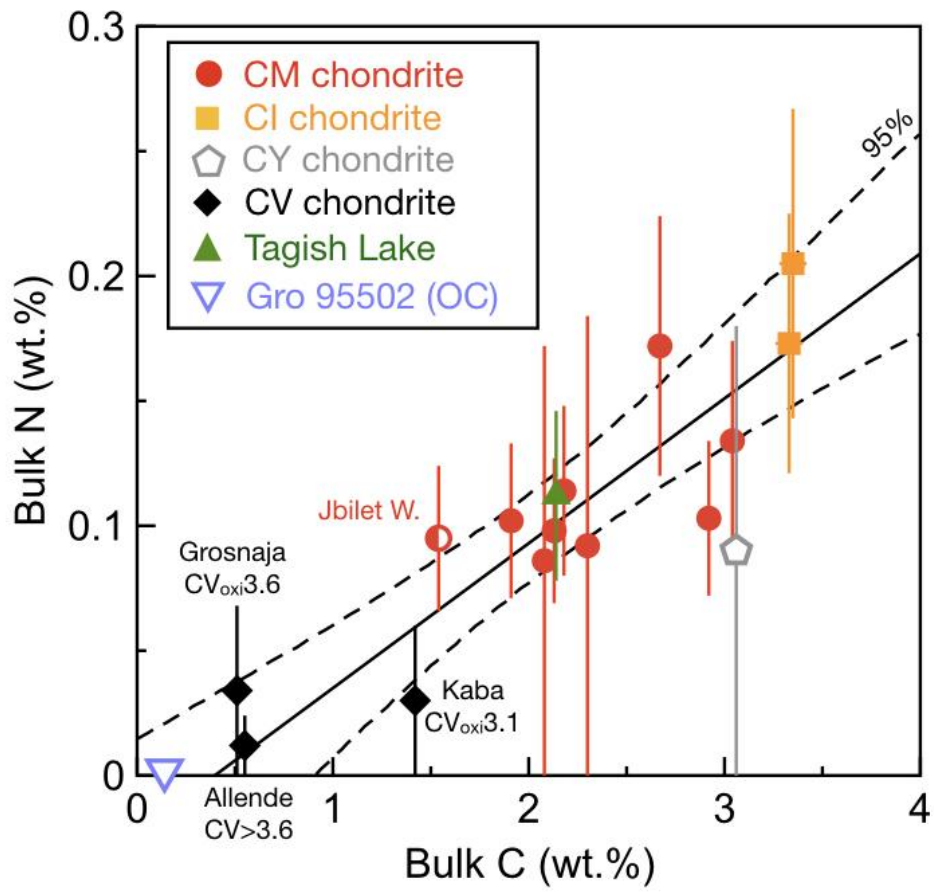
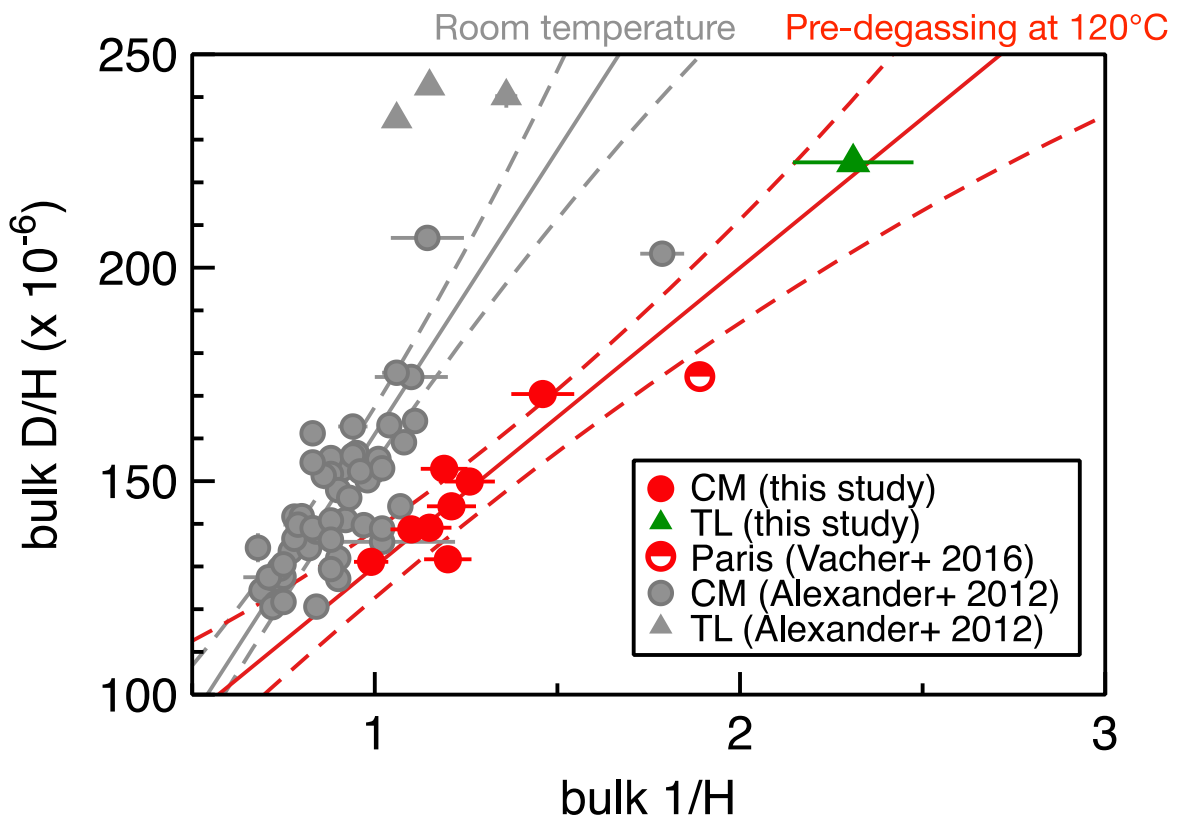


Fig. 3

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Fig. 4

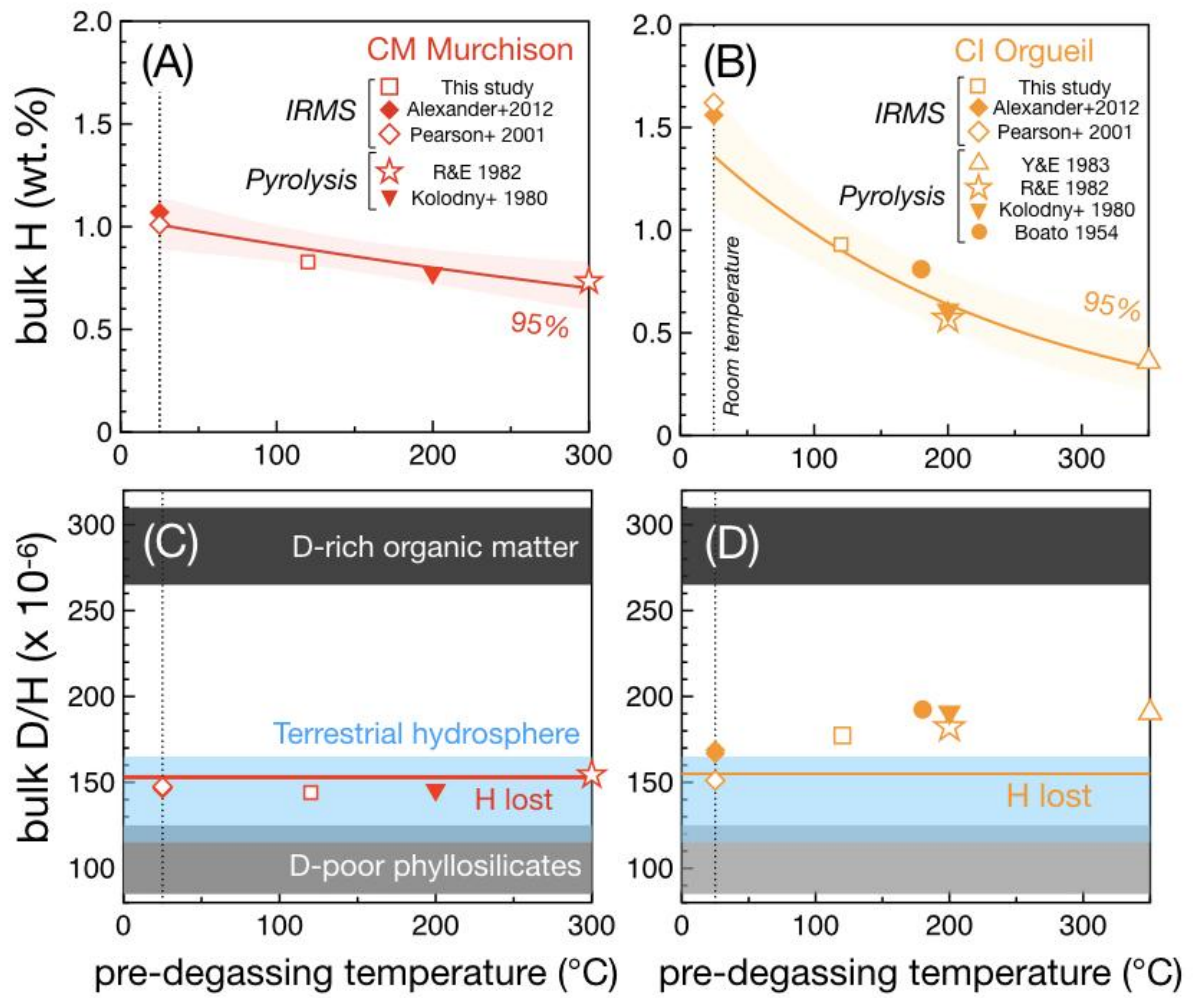


Fig. 5

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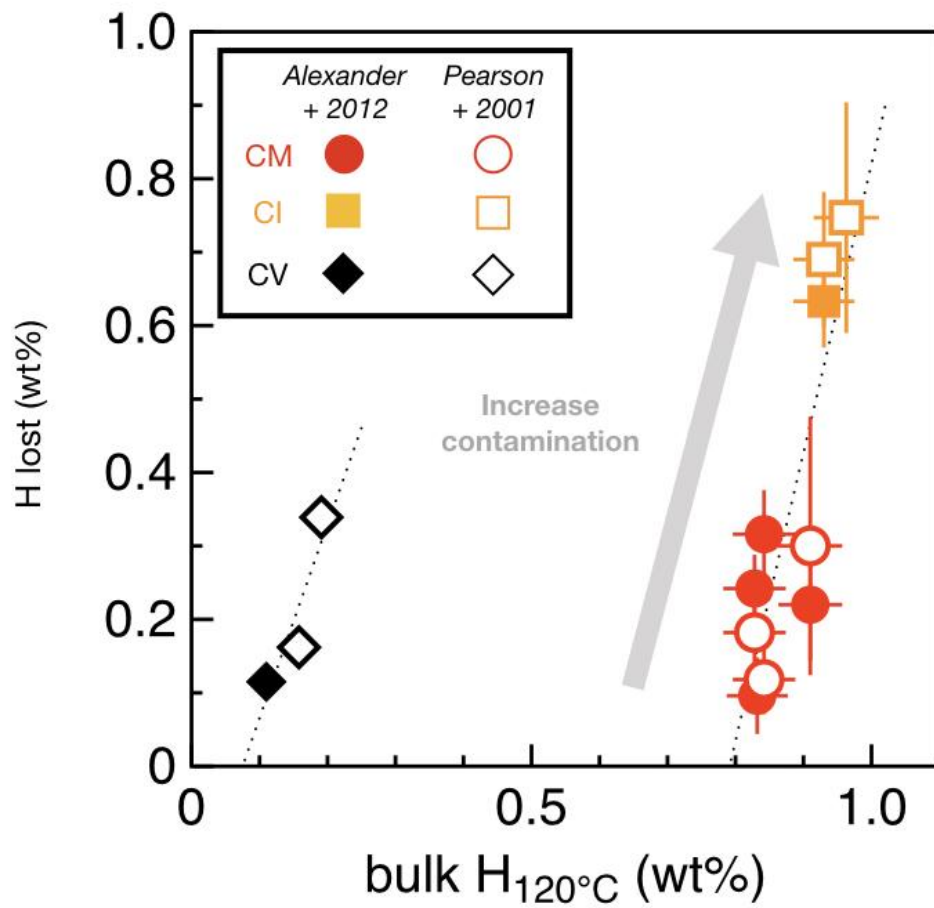


Fig. 6

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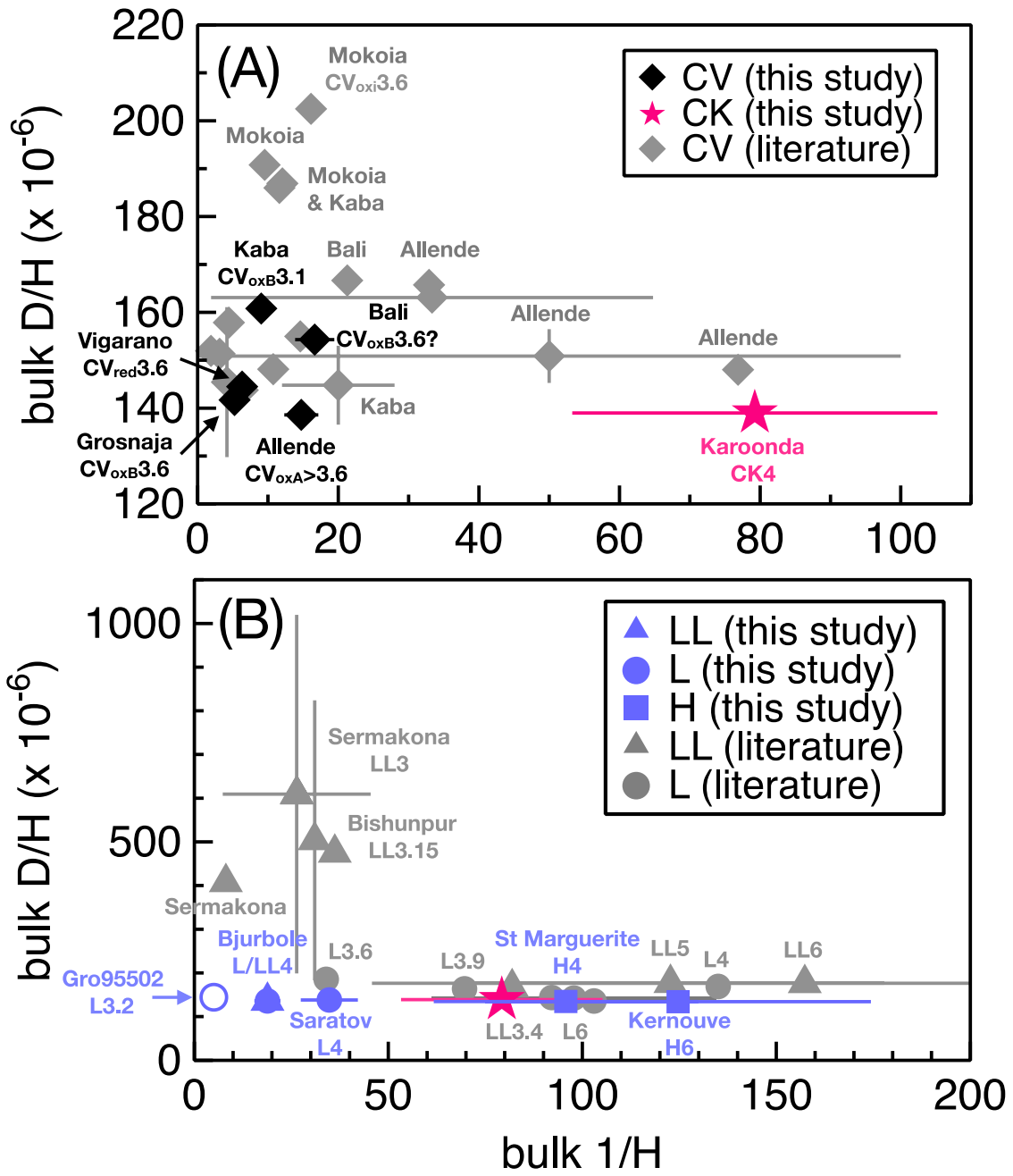


Fig. 7

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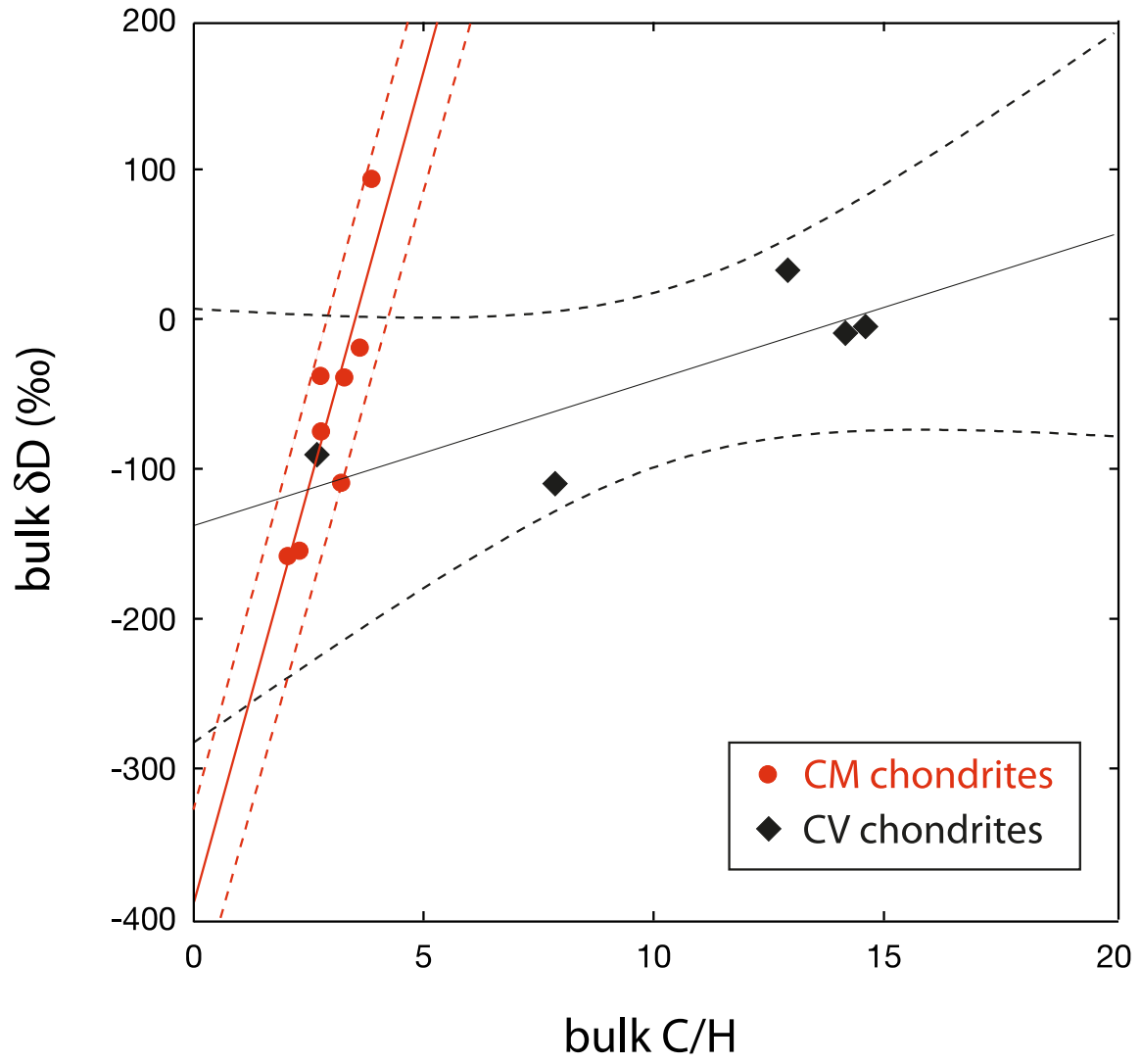


Fig. 8

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Table 1: Hydrogen abundances and isotopic compositions of chondrites.

| Meteorite | Type | wt% H | err. (2 σ) | 2 sd | δ D (‰) | err. (2 σ) | 2 sd | D/H ($\times 10^{-6}$) | err. (2 σ) | 2 sd |
|------------------------------------|--------|-------|-----------------------|-------|-------------------|-----------------------|------|-----------------------------|-----------------------|------|
| Aguas Zarcas ($n = 2$) | CM2 | 0.868 | 0.045 | 0.087 | -107.1 | 2 | 1.6 | 139.1 | 0.3 | 0.3 |
| Jbilet Winselwan ($n = 2$) | CM2 | 0.471 | 0.032 | 0.018 | -38.9 | 2 | 9.5 | 149.7 | 0.3 | 1.5 |
| LON 94101 ($n = 2$) ^c | CM2 | 0.832 | 0.045 | 0.012 | -154.6 | 2 | 3.1 | 131.7 | 0.3 | 0.5 |
| Maribo ($n = 2$) | CM2 | 0.794 | 0.044 | 0.028 | -37.9 | 2 | 1.1 | 149.9 | 0.3 | 0.2 |
| Mighei ($n = 2$) | CM2 | 0.910 | 0.047 | 0.038 | -109.4 | 2 | 0.8 | 138.7 | 0.3 | 0.1 |
| Mukundpura ($n = 4$) | CM2 | 1.013 | 0.048 | 0.139 | -158.2 | 2 | 6.7 | 131.1 | 0.3 | 1 |
| Murchison ($n = 2$) | CM2 | 0.828 | 0.046 | 0.081 | -74.9 | 2 | 0.9 | 144.1 | 0.3 | 0.1 |
| Murray ($n = 2$) | CM2 | 0.842 | 0.046 | 0.038 | -18.5 | 2 | 2.6 | 152.9 | 0.3 | 0.4 |
| Paris (altered area) ^a | CM2 | 0.686 | 0.040 | | 93.7 | 2 | | 170.4 | 0.3 | |
| Paris ^b | CM2 | 0.530 | 0.010 | | 120.3 | 1 | | 174.5 | 0.3 | |
| Tagish Lake ($n = 3$) | Ung. C | 0.433 | 0.031 | 0.007 | 442.3 | 2 | 24 | 224.7 | 0.3 | 3.7 |
| Alais ($n = 2$) | CI | 0.963 | 0.048 | 0.092 | 133.7 | 2 | 28.9 | 176.6 | 0.3 | 4.5 |
| Orgueil ($n = 4$) | CI | 0.930 | 0.045 | 0.148 | 138.0 | 2 | 15.3 | 177.3 | 0.3 | 2.4 |
| Y-980115 ($n = 3$) ^d | CY | 0.710 | 0.041 | 0.212 | -106.0 | 2 | 3 | 139.2 | 0.3 | 0.5 |
| Allende ($n = 2$) | CV | 0.068 | 0.011 | 0.011 | -109.9 | 2 | 2.4 | 138.6 | 0.8 | 0.4 |
| Bali | CV | 0.060 | 0.010 | | -9.3 | 5 | | 154.3 | 0.8 | |
| Grosnaja | CV | 0.191 | 0.019 | | -90.5 | 5 | | 141.7 | 0.8 | |
| Kaba | CV | 0.110 | 0.014 | | 32.6 | 5 | | 160.8 | 0.8 | |
| Vigarano | CV | 0.158 | 0.018 | | -72.6 | 5 | | 144.5 | 0.8 | |
| Karoonda | CK | 0.013 | 0.004 | | -107.4 | 5 | | 139.0 | 0.8 | |
| Bjurbole | LL/L4 | 0.053 | 0.009 | | -128.1 | 5 | | 135.8 | 0.8 | |
| GRO 95502 ^e | L3 | 0.199 | 0.020 | | -73.2 | 5 | | 144.4 | 0.8 | |
| Saratov ($n = 2$) | L4 | 0.029 | 0.006 | 0.011 | -113.7 | 5 | 2.3 | 138.1 | 0.8 | 0.4 |
| Sainte Marguerite | H4 | 0.010 | 0.004 | | -135.8 | 5 | | 134.6 | 0.8 | |
| Kernouve | H6 | 0.008 | 0.003 | | -138.3 | 5 | | 134.2 | 0.8 | |

922 ^aPiece of Paris provided by Bekaert et al. (2018). ^bData from Vacher et al. (2016) using the same

923 methodology as this study. ^cLonewolf Nunataks 94101. ^dYamato 980115. ^eGrosvenor Mountains.

924 Err. (2 σ): 2 σ sample error (see methods). 2 sd: 2 σ standard deviation on n replicates.

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926 Table 2: Carbon and nitrogen abundances and carbon isotopic compositions of chondrites.
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| Meteorite | Type | wt% N | err. (2σ) | 2 sd | wt% C | err. (2σ) | 2 sd | δ ¹³ C (‰) | err. (2σ) | 2 sd |
|----------------------------|--------|-------|--------------|-------|-------|--------------|------|--------------------------|--------------|------|
| Aguas Zarcas | CM2 | 0.098 | 0.029 | | 2.13 | 0.04 | | -9.8 | 0.2 | |
| Jbilet Winselwan | CM2 | 0.095 | 0.029 | | 1.54 | 0.03 | | -10.9 | 0.2 | |
| LON 94101 ^a | CM2 | 0.102 | 0.031 | | 1.91 | 0.04 | | -13.7 | 0.2 | |
| Maribo | CM2 | 0.114 | 0.034 | | 2.18 | 0.04 | | -5.5 | 0.2 | |
| Mighei | CM2 | 0.103 | 0.031 | | 2.92 | 0.06 | | -12.5 | 0.2 | |
| Mukundpura (<i>n</i> = 3) | CM2 | 0.086 | 0.086 | 0.008 | 2.08 | 0.04 | 0.04 | -3.9 | 0.2 | 2.3 |
| Murchison | CM2 | 0.092 | 0.092 | | 2.30 | 0.05 | | -10.4 | 0.2 | |
| Murray | CM2 | 0.134 | 0.040 | | 3.04 | 0.06 | | -11.7 | 0.2 | |
| Paris | CM2 | 0.172 | 0.052 | | 2.67 | 0.05 | | -7.5 | 0.2 | |
| Tagish Lake | Ung. C | 0.112 | 0.034 | | 2.14 | 0.04 | | 4.7 | 0.2 | |
| Alais | CI | 0.173 | 0.052 | | 3.33 | 0.07 | | -15.9 | 0.2 | |
| Orgueil (<i>n</i> = 2) | CI | 0.205 | 0.062 | 0.014 | 3.35 | 0.07 | 0.78 | -12.9 | 0.2 | 5.7 |
| Y-980115 ^b | CY | 0.090 | 0.090 | | 3.06 | 0.06 | | -12.9 | 0.2 | |
| Allende | CV | 0.012 | 0.012 | | 0.55 | 0.01 | | -23.7 | 0.2 | |
| Bali | CV | bdl | | | 0.85 | 0.02 | | -19.9 | 0.2 | |
| Grosnaja | CV | 0.034 | 0.034 | | 0.51 | 0.01 | | -21.6 | 0.2 | |
| Kaba | CV | 0.030 | 0.030 | | 1.42 | 0.03 | | -17.8 | 0.2 | |
| Vigarano | CV | bdl | | | bdl | | | bdl | 0.2 | |
| Karoonda | CK | bdl | | | 0.09 | 0.00 | | -20.7 | 0.2 | |
| Bjurbole | LL/L4 | bdl | | | 0.14 | 0.00 | | -26.5 | 0.2 | |
| GRO 95502 ^c | L3 | bdl | | | bdl | | | bdl | 0.2 | |
| Saratov | L4 | bdl | | | 0.12 | 0.00 | | -26.2 | 0.2 | |
| Sainte Marguerite | H4 | bdl | | | 0.08 | 0.00 | | -24.7 | 0.2 | |
| Kernouve | H6 | bdl | | | 0.10 | 0.00 | | -26.6 | 0.2 | |

928 ^aLonewolf Nunataks 94101, ^bYamato 980115 ^cGrosvenor Mountains.

929 Err. (2σ): 2σ sample error (see methods), 2 sd: 2σ standard deviation on *n* replicates.

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