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► **To cite this version:**

Laurette Piani, Yves Marrocchi, Thomas Rigaudier, Lionel G Vacher, Dorian Thomassin, et al.. Earth's water may have been inherited from material similar to enstatite chondrite meteorites. *Science*, 2020, 369 (6507), pp.1110-1113. <10.1126/science.aba1948>. <hal-02933795>

HAL Id: hal-02933795

<https://hal.science/hal-02933795v1>

Submitted on 8 Sep 2020

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1 **Earth's water may have been inherited from material similar to enstatite chondrite**
2 **meteorites**

3
4 Laurette Piani^{1*}, Yves Marrocchi¹, Thomas Rigaudier¹, Lionel G. Vacher^{1,2}, Dorian
5 Thomassin¹, Bernard Marty¹

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7
8 ¹Centre de Recherches Pétrographiques et Géochimiques (CRPG), Centre National de
9 Recherche Scientifique (CNRS)-Université de Lorraine, Vandoeuvre-les-Nancy, F-54500,
10 France

11
12 ²Department of Physics, Washington University in St. Louis, St. Louis, MO, USA

13
14 *Correspondence to: laurette.piani@univ-lorraine.fr

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17
18 **Abstract (132/125 words):**

19 The origin of Earth's water remains unknown. Enstatite chondrite (EC) meteorites have
20 similar isotopic composition to terrestrial rocks, so are often considered representative of the
21 material which formed Earth. ECs are presumed to be devoid of water because they formed in
22 the inner Solar System. Earth's water is thus generally attributed to a late addition of a small
23 fraction of hydrated materials, such as carbonaceous chondrite meteorites, which originated in
24 the outer Solar System where water was more abundant. We show that EC meteorites contain
25 sufficient hydrogen to have delivered to Earth at least three times the mass of water in its
26 oceans. Because EC hydrogen and nitrogen isotopic compositions match those of Earth's
27 mantle, EC-like asteroids might have been major contributors of these volatile elements to the
28 Earth's crust and mantle.

29
30 **Main text (2756 words):**

31 The origin of Earth's water is debated. The isotopic composition of Earth suggests that it
32 is composed of material from the inner Solar System, such as enstatite chondrite (EC)
33 meteorites (1-3). The inner Solar System was too warm to have retained water ice, so
34 terrestrial water is thought to have been supplied by hydrated materials that originally formed
35 in the outer Solar System before migrating inwards (4, 5).

36 Hydrogen isotopic compositions are conventionally expressed as $\delta D \equiv$
37 $[(D/H)_{\text{sample}}/(D/H)_{\text{SMOW}} - 1] \times 1000$, where D/H is the ratio of ²H (deuterium) to ¹H
38 (protium), and standard mean ocean water (SMOW) is the mean value of Earth's oceans. δD
39 varies greatly among Solar System objects. The bulk protosolar nebula had $\delta D = -865\text{‰}$,
40 estimated from the solar wind and remnant H₂ in the atmospheres of the giant planets (6).
41 Compared to this protosolar value, all other materials in the Solar System are enriched in
42 deuterium, with inner Solar System objects having intermediate values e.g., $\delta D \equiv 0\text{‰}$ in
43 terrestrial oceans and -165 to $+800\text{‰}$ in asteroids, as recorded in primitive meteorites (7).
44 Outer Solar System objects, such as comets, are even more enriched in deuterium, up to $\delta D =$
45 $2,400\text{‰}$ in comet 67P/Churyumov-Gerasimenko (8). Among meteorites, water-rich
46 carbonaceous chondrites of Ivuna-type (CI) and Mighei-type (CM) are potential sources of
47 terrestrial water because their δD values are distributed around a value coincident with Earth's
48 oceans e.g., $\delta D \equiv 0\text{‰}$ (9). Dynamical simulations suggest that the so-called C-type
49 (carbonaceous) asteroids –whose optical properties suggest a genetic link to water-rich
50 carbonaceous chondrites– could have been scattered into the inner solar system from beyond

51 the orbit of Jupiter [e.g., (10)], potentially delivering water to Earth during or after its main
52 phase of accretion throughout the first millions of years of the Solar System formation.

53 An alternative possibility is that EC-like materials, which have similar isotopic abundances
54 to Earth for some key elements like oxygen, titanium, calcium, molybdenum (3), could
55 contain enough hydrogen with the appropriate isotopic composition to provide water to the
56 growing proto-Earth. Testing this scenario has been hampered by difficulties in measuring H
57 concentrations and isotopic ratios in ECs, due to their assumed low H abundances and the
58 potential for terrestrial contamination (7).

59 We measured H abundances and isotopic compositions for a suite of thirteen ECs that
60 spans the full range of thermal metamorphism degrees (i.e., represented by the petrologic
61 types 3 for the least metamorphosed to 6 for the chondrites the most altered by thermal
62 metamorphism on the parent asteroid) to evaluate potential effects of parent body processing
63 that could have altered the original water content and H isotopic composition (11). We also
64 analyzed one aubrite meteorite (Norton County), which represents an evolved planetary body
65 of enstatite meteorite composition for which metal-silicate segregation (differentiation)
66 occurred (11). Hydrogen measurements were performed at the Centre de Recherches
67 Pétrographiques et Géochimiques (CRPG, Nancy, France), either on bulk samples with an
68 elemental analyzer (EA) coupled to a stable isotope ratio mass spectrometer (IRMS), or at the
69 micron scale using a secondary ion mass spectrometer (SIMS) (11).

70
71 We find the enstatite chondrites have bulk hydrogen contents (reported as water
72 equivalents) ranging from 0.08 to 0.54 wt.% H₂O (Fig. 1A, Table S2). The EH3 meteorites
73 are generally more H rich (0.44 ± 0.04 wt.% H₂O, all uncertainties are one standard deviation
74 unless otherwise noted) than the more metamorphosed groups EH4 (0.2 ± 0.1 wt.% H₂O),
75 EH5 (0.3 ± 0.1 wt.% H₂O), and EL6 (0.3 ± 0.3 wt.% H₂O). The Norton County aubrite is 0.3
76 ± 0.2 wt.% H₂O. These EC water contents are well below those measured in the water-rich
77 carbonaceous chondrites Orgueil (CI), Alais (CI), and Murchison (CM), which are 7.2–9.1
78 wt.% H₂O (Fig. 1A, Table S2). However, all analyzed ECs contain hydrogen concentrations
79 above the EA-IRMS detection limit (~ 0.05 wt.% H₂O; (11)). The least-altered EH chondrites
80 (EH3 and EH4) show δD values systematically below the current value of Earth's oceans
81 (averaging $\delta D = -103 \pm 3\text{‰}$), whereas metamorphosed ECs (EH5 and EL6) and the aubrite
82 have even lower values, averaging $\delta D = -127 \pm 15\text{‰}$ (Fig. 1B, Table S2).

83 If the hydrogen contents had been totally due to atmospheric contamination during sample
84 preparation, constant D/H ratios would be observed despite variable H concentrations.
85 Instead, our data follow a negative trend in a δD vs. 1/H diagram (Fig. S5), indicating possible
86 loss of D-enriched hydrogen-bearing organic molecules (12) during thermal metamorphism
87 within the EC parent asteroid(s). In the least metamorphosed sample we analyzed, the pristine
88 EH3 meteorite Sahara 97096 [EH3.1–3.4 (13); fragment of the same meteorite as Sahara
89 97116 in Table S2], only a thin (≈ 500 μm) surface layer of terrestrial alteration is visible (11);
90 its inner portions remain unaffected, as attested by the presence of unaltered oldhamite (CaS)
91 grains, a highly hydrophilic mineral (14, 15). The fresh interior of Sahara 97096 has lower
92 (and constant) H contents (0.5 ± 0.1 wt.% H₂O) than the altered parts (3.0 ± 1.1 wt.% H₂O),
93 and their H isotopic compositions are also different (fresh interior $\delta D = -103.6 \pm 0.7\text{‰}$,
94 altered surface $\delta D = -136.3 \pm 3.9\text{‰}$; Fig. S4, Table S2). Similar features have been observed
95 in Martian meteorites subjected to desert alteration, with the H content and isotopic
96 characteristics being modified only at the near surface (i.e., < 2 mm) (16). In addition, the
97 hydrogen contents and D/H ratios measured for the two meteorite finds (Sahara 97096 and
98 Yamato 791790) are very close to the ones measured for the Indarch chondrite that has
99 collected after its observed fall and consistent with a metamorphic evolution of the H
100 signatures in ECs (Table S2; Fig. S5B). These observations suggest that the H contents of

101 ECs were established during the early Solar System and have experienced minimal
102 modifications on Earth.

103 To confirm the indigenous nature of H in the interiors of ECs, we used SIMS to analyze
104 the H contents and isotopic compositions of the glass fraction (mesostasis) in four chondrules
105 -sub-millimetre silicate spherules that correspond to the main mineral assemblages in ECs- of
106 Sahara 97096, the least-metamorphosed EC in our sample (11, 12). For comparison, we
107 performed similar measurements in two CV chondrites (Kaba and Vigarano) having the same
108 degree of metamorphism (17). The glassy mesostasis of Sahara 97096 contains between 2,700
109 and 12,300 wt. ppm H₂O equivalent, with an average value of $7,560 \pm 1,546$ wt. ppm H₂O
110 (two standard deviations, 2σ) for 16 measurements (Fig. 1C, Table S4). Although their
111 metamorphic grades are similar to Sahara 97096, the CV chondrites Vigarano and Kaba show
112 lower H contents of 330 ± 140 and 210 ± 120 wt. ppm H₂O, respectively (Fig. 1C, Table S4).
113 The H isotopic composition of Sahara 97096 mesostasis is homogeneous with an average δD
114 value of $-147 \pm 16\%$ (2σ ; Fig. 1D, Table S4), whereas Vigarano and Kaba show $\delta D = -261 \pm$
115 25 and $+17 \pm 95\%$, respectively (2σ ; Fig. 1D, Table S4). Among these three meteorites, only
116 Kaba underwent aqueous alteration of its chondrules (18), which accounts for the variable
117 D/H ratio of its mesostasis. Vigarano contains traces of incipient aqueous alteration restricted
118 to its fine-grained matrix (19). As Sahara 97096 does not show any evidence of aqueous
119 alteration, the abundant H contents of its chondrule mesostases was thus acquired before the
120 EC parent asteroid evolution, possibly when chondrule formed.

121
122 Considering the H concentration of Sahara 97096 mesostasis, the modal abundance of
123 mesostasis in Sahara 97096 chondrules [16.2 vol.% (15)] and the modal abundance of
124 chondrules in ECs [~ 70 vol.% (20)], H in chondrule mesostases accounts for 702 ± 117 wt.
125 ppm H₂O of the bulk rock, or $\sim 13\%$ of the bulk-rock H content. Another potential source of H
126 in ECs is insoluble organic matter (IOM). However, given the low abundance of IOM in ECs
127 (0.6 wt.%) and the H concentration in EC organics [0.7 wt.% (12)], the contribution of IOM
128 to the bulk-rock H content is only 380 ppm H₂O, or 7.7% of the total H content of the
129 chondrite. Combined, H hosted in chondrule mesostases and IOM can only account for $\sim 20\%$
130 of the total H measured in the Sahara 97096 bulk rock.

131 H concentrations have been reported in low-calcium pyroxenes from non-carbonaceous
132 chondrite materials from (i) the S-type asteroid Itokawa (700–1,000 wt. ppm H₂O) brought
133 back by the Hayabusa sample-return spacecraft and (ii) the metamorphosed ordinary
134 chondrite Larkman Nunatak 12036 (600–1,300 wt. ppm H₂O) (21). We found an even higher
135 H concentration (5,300 wt. ppm H₂O; Table S2) for the pyroxene fraction of the Norton
136 County aubrite. The modal abundance of low-calcium pyroxene in ECs is ~ 50 vol.% (15), so
137 enstatite pyroxenes could account for $\sim 15\%$ or $\sim 58\%$ of the bulk H content of ECs
138 considering the highest H-content of OC pyroxenes (1,300 wt. ppm H₂O) or the aubrite
139 pyroxene fraction, respectively. The remaining $\sim 65\%$ to 20% of H measured in the bulk rock
140 may be contributed by other unknown carriers such as sulfur-rich carbon-bearing porous
141 amorphous silica (22), due to an underestimation of the H content in EC pyroxenes, and/or
142 could derive from some pervasive terrestrial contamination, although a limited amount of
143 contamination is expected otherwise it would have erased the thermal metamorphism effect
144 visible on the δD vs. $1/H$ diagram (Fig. S5). Our SIMS measurements demonstrate that at
145 least 20% of the H in ECs has an identified carrier phase (mesostasis or IOM) and that most
146 of the remaining 80% is probably of indigenous origin. This implies that terrestrial hydrogen
147 could be derived from ECs without invoking additional sources.

148 We incorporated the H concentrations measured in ECs (Tables S2 and S4) into theoretical
149 models of Earth's formation that invoke mixing of chondrite-like materials. These models
150 utilize various proportions of enstatite, ordinary, and carbonaceous chondrite-like materials,

151 e.g. (1-3, 5, 23), so we explored three end-member compositions: (i) 100% EC-like materials
152 (1), (ii) 68% EC-like and 32% carbonaceous chondrite-like materials (2, 23), although these
153 proportions could be inconsistent for the isotopic composition of the Earth's mantle (11), and
154 (iii) ~70% EC-like, ~25% ordinary chondrite-like, and ~5% carbonaceous chondrite-like
155 materials (3) (Fig. 2). In all cases, EC-like materials contribute substantially to Earth's water
156 budget, supplying 3.4 to 23.1 times the mass of Earth's oceans (1.4×10^{21} kg) once our H
157 content measurements are included (Table S2). These estimates are upper limits because loss
158 of volatiles might have occurred during Earth's accretion e.g. (24, 25). Considering only
159 identified H-bearing phases (mesostasis and IOM), EC-like materials alone contribute ~3 to
160 4.5 times the mass of Earth's oceans (Fig. 2). If Sahara 97096 is representative of the most
161 pristine ECs (Fig. 2), its bulk-rock hydrogen content (4,700 wt. ppm H₂O or ≥ 14 ocean
162 masses) is sufficient to explain the highest estimates of the water content of Earth's surface
163 and mantle, which are 1,000–3,900 ppm H₂O (5, 26). Mesostasis and IOM H contents alone
164 can account for 3 to 5 times the total mass of the oceans, corresponding to intermediate
165 estimates of Earth's mantle water content.

166 The hydrogen isotopic composition of ECs is depleted in deuterium relative to Earth's
167 oceans, but within the range of mantle values (Fig. 3). ECs are not the only group of
168 meteorites with D-poor values consistent with Earth's mantle, as CM-type carbonaceous
169 chondrites also span the range of mantle H isotopic compositions (Fig. 3). Nitrogen isotopes
170 provide additional constraints (27), as the mantle is depleted in both D and ¹⁵N relative to the
171 surface reservoirs (atmosphere and oceans). Combining H and N isotopic compositions
172 allows us to identify potential chondritic precursors (Fig. 4). Only ECs are compatible with
173 both the δD and $\delta^{15}N$ values measured in mantle-derived rocks (Fig. 4). D-poor solar
174 hydrogen ($\delta D = -865\%$; (6)) could also have contributed to the mantle composition (28),
175 although it would require extreme enrichments of solar H relative to solar Ne because mantle
176 neon is solar-like. The chondritic H/Ne ratio [4×10^8 (29)] is five orders of magnitude higher
177 than the solar ratio [7.9×10^3 (30)]. Therefore we favor the incorporation of EC-like material
178 in the silicate Earth (1).

179 Earth's surficial (oceans and atmosphere) H and N isotopic compositions ($\delta D \equiv 0\%$ and
180 $\delta^{15}N \equiv 0\%$) cannot be easily derived from EC materials alone because they are more rich in D
181 by ~100–200‰ and ¹⁵N by 5–40‰. Atmospheric escape to space, often invoked for
182 atmospheric species including water vapor, could potentially achieve such enrichments, but
183 light noble gases do not support this hypothesis. Ne-Ar isotopic variations in the mantle-
184 atmosphere system do not follow elemental/isotopic variations that would sign atmospheric
185 escape; they are best reproduced by mixing between a solar-like component trapped in the
186 solid Earth and a carbonaceous chondrite-like component with a low ²⁰Ne/²²Ne ratio (5). A
187 contribution of carbonaceous chondrite-like materials, especially CIs, to the surface inventory
188 would also be consistent with the observed H and N isotopic variations (Fig. 4) and can be
189 computed independently from H and N isotopic ratios (11). We find a ~4% contribution from
190 CI-like material to the Earth surficial reservoir based on hydrogen abundances and isotopic
191 compositions, and a ~15% contribution based on equivalent data for nitrogen (11). Although
192 nitrogen isotopes could also have contributions from cometary materials (11), these estimates
193 are similar to those obtained from noble gases (5) and molybdenum isotopes (23). We cannot
194 determine exactly when the CI-like material was delivered to the surficial reservoir, but we
195 note that it must have been sufficiently late during Earth's formation to prevent a global re-
196 homogenization of the mantle with the surface.

197 198 **Acknowledgments:**

199 We thank the Field Museum (Chicago, USA), the French National Museum of Natural
200 History (Paris, France), the Japanese National Institute for Polar Research (Tokyo, Japan), the

201 University of New Mexico (Albuquerque, USA), Natural History Museum (Vienna, Austria)
202 and the CEREGE meteoritic collection (Aix en Provence, France) for providing the
203 meteorites samples. We thank Nordine Bouden and the members of the Ion Probe Team
204 Nancy (IPTN) for help with the SIMS. We thank E. Deloule, A. Gurenko, L. France, M.
205 Broadley, D. Bekaert, and C. Cartier for fruitful discussions. This is CRPG contribution
206 #2997. **Funding:** Supported by the French Research National Agency (grant No. ANR-19-
207 CE31-0027-01 to L.P.) and by the European Research Council (grant No. 695618 to B.M.).
208 **Author contributions:** L.P. designed the study; L.P., T.G., L.G.V., and D.T. performed the
209 bulk and *in-situ* analyses. Y.M. and B.M. contributed to the experimental design. L.P., Y.M.,
210 and B.M. discussed the data and wrote the manuscript. All authors provided input to the data
211 analysis and manuscript preparation. **Competing interests:** We declare no competing
212 interests. **Data and materials availability:** All meteorites we studied are deposited in public
213 museums, as listed in the supplementary material. Our measured H abundances and D/H
214 ratios, for the meteorite samples and standards, are listed in Tables S1-S4 and Data S1.

215

216 **Supplementary materials:**

217 Materials and Methods

218 Tables S1–S6

219 Figs. S1–S6

220 Data S1

221 References (36–62)

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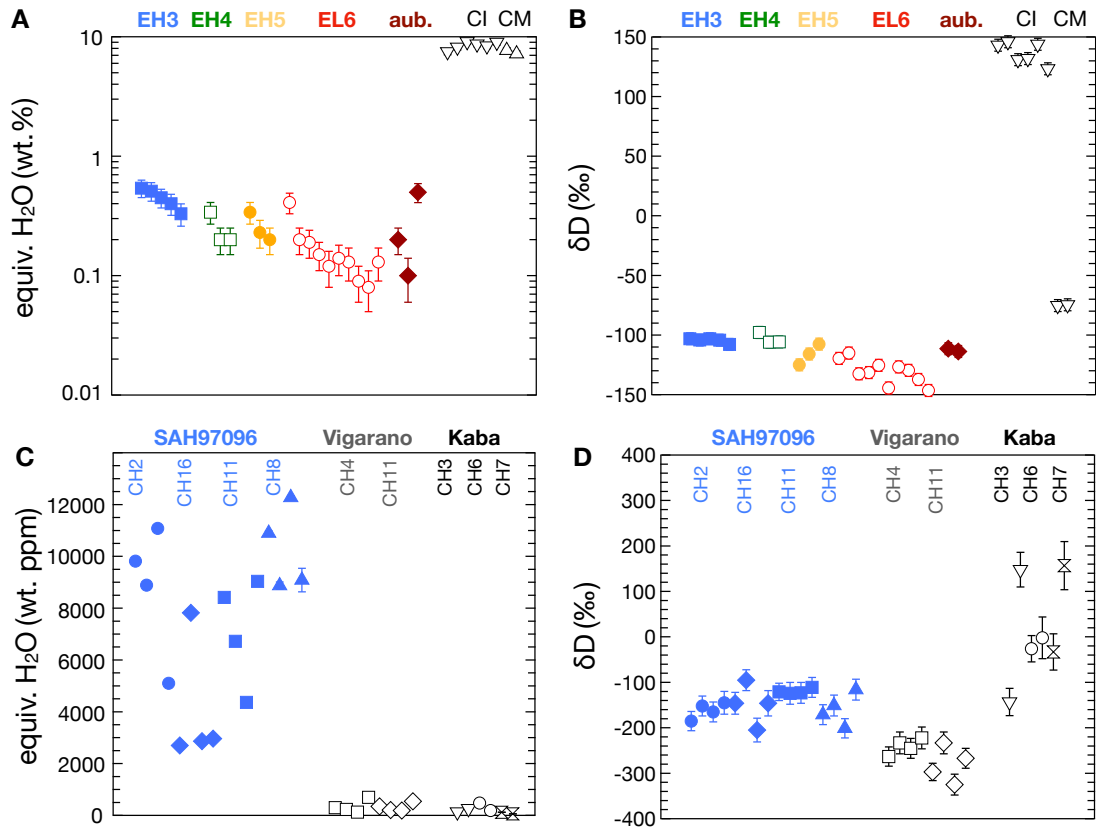
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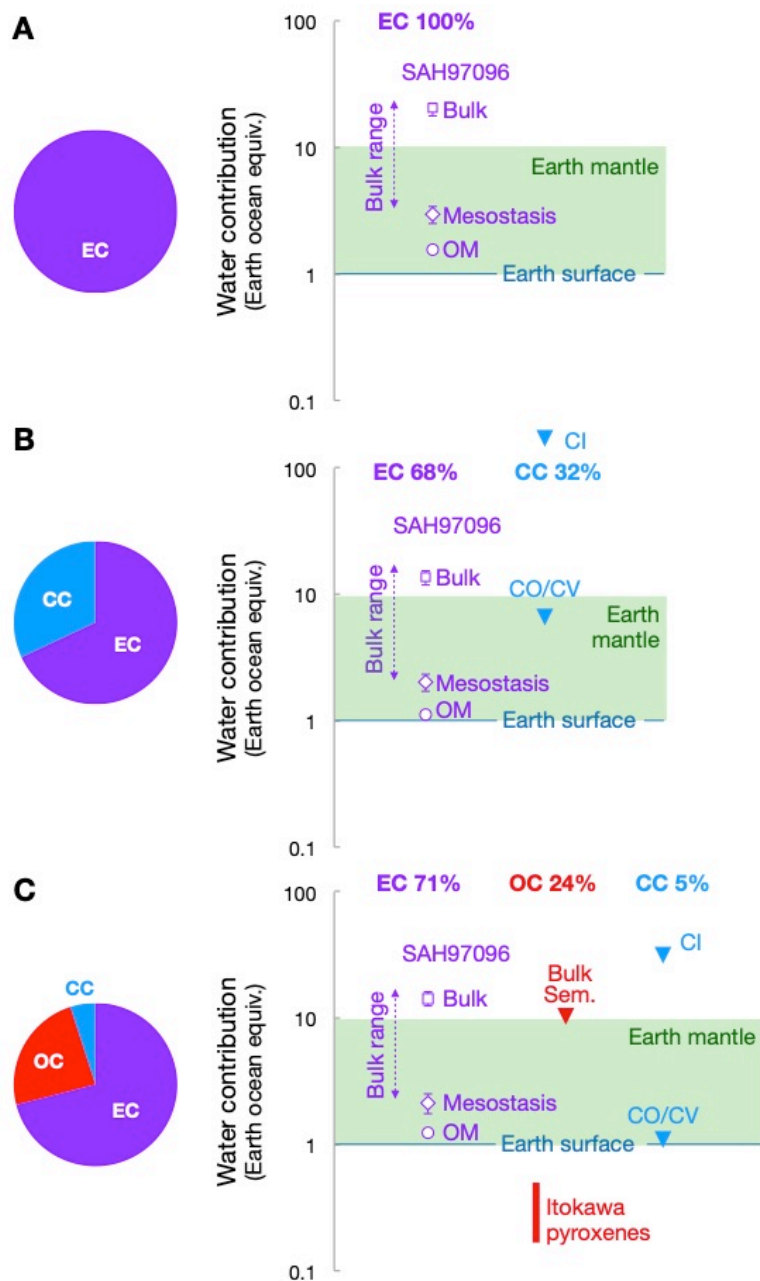
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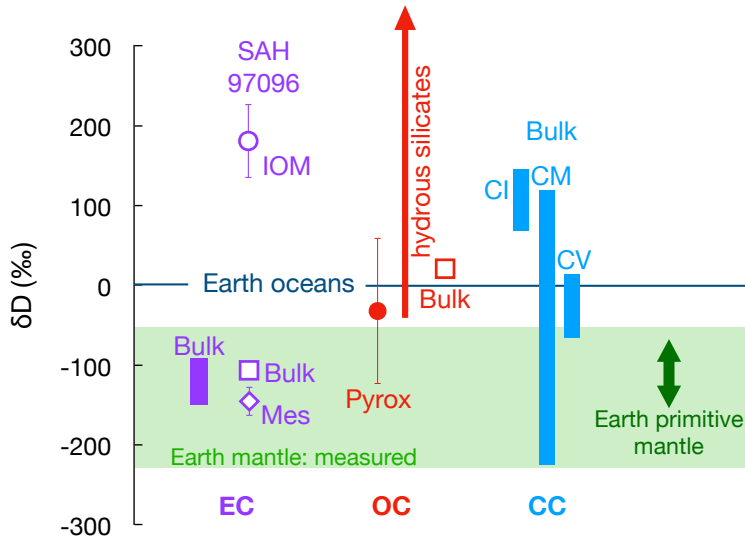
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 378 **Fig. 1. Hydrogen contents and isotopic compositions of enstatite chondrites.** (A) Bulk-
 379 rock hydrogen content, reported as water equivalent by weight on a logarithmic scale. (B)
 380 Bulk rock deuterium abundance, expressed as an offset from mean ocean water. (C)
 381 Mesostasis hydrogen content. (D) Mesostasis deuterium abundance. The CM- and CI-type
 382 carbonaceous chondrites Orgueil, Alais, and Murchison, and the aubrite Norton County, are
 383 included in panels A and B for comparison. Panels C and D show data for the primitive EC
 384 Sahara 97096 (SAH97096; EH3.1–3.4), and the two CV-type carbonaceous chondrites
 385 Vigarano and Kaba for comparison (11). Different symbols in panels C and D indicate
 386 different chondrules (labeled CH). Error bars are 2σ and are smaller than the symbol in
 387 panel B. The data are listed in Tables S2 and S4.

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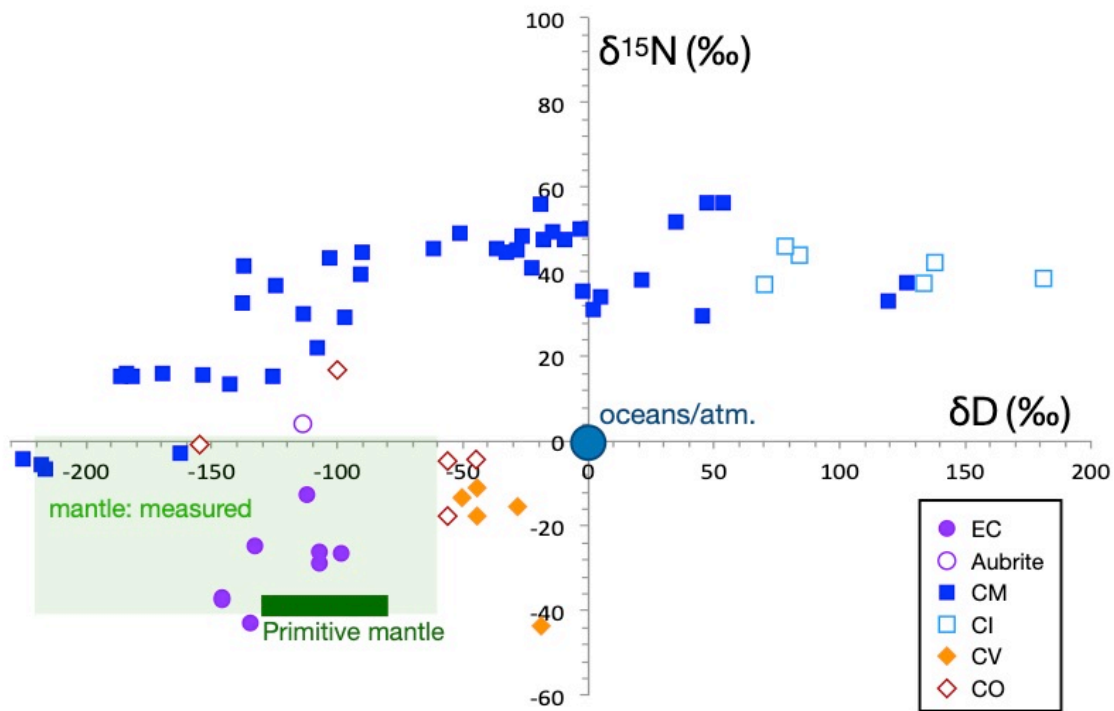
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Figure 2. Contributions of accreting materials to Earth's hydrogen budget in three different models. Values are expressed in equivalent ocean masses, where one ocean = 1.4×10^{21} kg H₂O. (A) A model Earth comprising only enstatite chondrites (1). (B) A model Earth comprising 68% ECs and 32% carbonaceous chondrites (CCs) (2). (C) A model Earth comprising 71% ECs, 24% ordinary chondrites (OCs), and 5% CCs [(3), their most extreme case]. The consistency of such models for the isotopic composition of the mantle is discussed in (11). SAH97096 = Sahara 97096 (EH3); Bulk = bulk rock; OM = organic matter; Sem. = Semarkona (LL3.0). Carbonaceous chondrites are divided into water-rich (CI-type) and water-poor groups (CV- and CO-type). Water contribution by ECs are in purple, by OCs are in red, and by CCs in blue. The water content of Earth's mantle (green band) is between 1 and ≥ 10 ocean masses (26). We consider 10 ocean masses in the mantle as a maximum value (green band). Regardless of the model considered, the water provided by ECs alone lies above the maximum value of 10 oceans and thus substantially contributes to the Earth's water budget. Data are from (5, 7, 11, 21) and this study (Tables S2 and S4). Error bars are 2σ .



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Figure 3. Hydrogen isotopic compositions of meteorites compared to Earth’s mantle and oceans. Data are shown for enstatite chondrites (ECs, bulk rock and mesostasis; in purple), ordinary chondrite (OC) pyroxenes and hydrous minerals (in red), and carbonaceous chondrites (CCs, bulk rock, CI-, CM-, and CV-type; in blue). SAH97096 = Sahara 97096 (EH3); Bulk = bulk rock; OM = organic matter; Mes = mesostasis; Pyrox = pyroxene. Meteorite data are from (7, 12, 21, 31) and this study (Tables S2 and S4). Values for Earth’s mantle ($\delta D = -220$ to -20‰ , green shaded area) are from (26), with the most D-depleted values reported in (28). The primitive mantle ranges from $\delta D = -130$ to -75‰ (32, 33).



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Figure 4. Hydrogen and nitrogen isotopic compositions of meteorites compared to Earth's mantle and surface. Meteorite bulk-rocks values are shown for enstatite chondrites (ECs) and CM-, CI-, CV-, and CO-type carbonaceous chondrites; data are from Table S2 and (7, 34, 35). Mantle δD values are from (26, 28). The primitive mantle δD is from (32) and (33) and $\delta^{15}N$ by the most negative values (-40‰) in (35).