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Contribution of Ryugu-like material to Earth's volatile inventory by Cu and Zn isotopic analysis

Marine Paquet^{1*}, Frederic Moynier^{1*}, Tetsuya Yokoyama², Wei Dai¹, Yan Hu¹, Yoshinari Abe³, Jérôme Aléon⁴, Conel M. O'D. Alexander⁵, Sachiko Amari⁶, Yuri Amelin⁷, Ken-ichi Bajo⁸, Martin Bizzarro^{1,9}, Audrey Bouvier¹⁰, Richard W. Carlson⁵, Marc Chaussidon¹, Byeon-Gak Choi¹¹, Nicolas Dauphas¹², Andrew M. Davis¹², Tommaso Di Rocco¹³, Wataru Fujiya¹⁴, Ryota Fukai¹⁵, Ikshu Gautam², Makiko K. Haba², Yuki Hibiya¹⁶, Hiroshi Hidaka¹⁷, Hisashi Homma¹⁸, Peter Hoppe¹⁹, Gary R. Huss²⁰, Kiyohiro Ichida²¹, Tsuyoshi Iizuka²², Trevor R. Ireland²³, Akira Ishikawa², Motoo Ito²⁴, Shoichi Itoh²⁵, Noriyuki Kawasaki⁸, Noriko T. Kita²⁶, Kouki Kitajima²⁶, Thorsten Kleine²⁷, Shintaro Komatani²¹, Alexander N. Krot²⁰, Ming-Chang Liu²⁸, Yuki Masuda², Kevin D. McKeegan²⁸, Mayu Morita²¹, Kazuko Motomura²⁹, Izumi Nakai²⁹, Kazuhide Nagashima²⁰, David Nesvorný³⁰, Ann N. Nguyen³¹, Larry Nittler⁵, Morihiko Onose²¹, Andreas Pack¹³, Changkun Park³², Laurette Piani³³, Liping Qin³⁴, Sara S. Russell³⁵, Naoya Sakamoto³⁶, Maria Schönbächler³⁷, Lauren Tafla²⁸, Haolan Tang²⁸, Kentaro Terada³⁸, Yasuko Terada³⁹, Tomohiro Usui¹⁵, Sohei Wada⁸, Meenakshi Wadhwa⁴⁰, Richard J. Walker⁴¹, Katsuyuki Yamashita⁴², Qing-Zhu Yin⁴³, Shigekazu Yoneda⁴⁴, Edward D. Young²⁸, Hiroharu Yui⁴⁵, Ai-Cheng Zhang⁴⁶, Tomoki Nakamura⁴⁷, Hiroshi Naraoka⁴⁸, Takaaki Noguchi²⁴, Ryuji Okazaki⁴⁸, Kanako Sakamoto¹⁵, Hikaru Yabuta⁴⁹, Masanao Abe¹⁵, Akiko Miyazaki¹⁵, Aiko Nakato¹⁵, Masahiro Nishimura¹⁵, Tatsuaki Okada¹⁵, Toru Yada¹⁵, Kasumi Yogata¹⁵, Satoru Nakazawa¹⁵, Takanao Saiki¹⁵, Satoshi Tanaka¹⁵, Fuyuto Terui⁵⁰, Yuichi Tsuda¹⁵, Sei-ichiro Watanabe¹⁷, Makoto Yoshikawa¹⁵, Shogo Tachibana⁵¹, Hisayoshi Yurimoto⁸

Affiliations:

¹Université Paris Cité, Institut de physique du globe de Paris, CNRS; 75005 Paris, France

²Department of Earth and Planetary Sciences, Tokyo Institute of Technology; Tokyo 152-8551, Japan.

³Graduate School of Engineering Materials Science and Engineering, Tokyo Denki University; Tokyo 120-8551, Japan.

⁴Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie, Sorbonne Université, Museum National d'Histoire Naturelle, CNRS UMR 7590, IRD; 75005 Paris, France.

⁵Earth and Planets Laboratory, Carnegie Institution for Science; Washington, DC, 20015, USA.

⁶McDonnell Center for the Space Sciences and Physics Department, Washington University; St. Louis, MO 63130, USA.

⁷Guangzhou Institute of Geochemistry, Chinese Academy of Sciences; Guangzhou, GD 510640, China.

⁸Natural History Sciences, IIL, Hokkaido University; Sapporo 001-0021, Japan.

⁹Centre for Star and Planet Formation, GLOBE Institute, University of Copenhagen; Copenhagen, K 1350, Denmark.

- 40 ¹⁰Bayerisches Geoinstitut, Universität Bayreuth; Bayreuth 95447, Germany.
- 41 ¹¹Department of Earth Science Education, Seoul National University; Seoul 08826, Republic
42 of Korea.
- 43 ¹²Department of the Geophysical Sciences and Enrico Fermi Institute, The University of
44 Chicago, 5734 South Ellis Avenue, Chicago 60637, USA.
- 45 ¹³Faculty of Geosciences and Geography, University of Göttingen; Göttingen, D-37077,
46 Germany.
- 47 ¹⁴Faculty of Science, Ibaraki University; Mito 310-8512, Japan.
- 48 ¹⁵ISAS/JSEC, JAXA; Sagamihara 252-5210, Japan.
- 49 ¹⁶General Systems Studies, The University of Tokyo; Tokyo 153-0041, Japan.
- 50 ¹⁷Earth and Planetary Sciences, Nagoya University; Nagoya 464-8601, Japan.
- 51 ¹⁸Osaka Application Laboratory, SBUWDX, Rigaku Corporation; Osaka 569-1146, Japan.
- 52 ¹⁹Max Planck Institute for Chemistry; Mainz 55128, Germany.
- 53 ²⁰Hawai‘i Institute of Geophysics and Planetology, University of Hawai‘i at Mānoa;
54 Honolulu, HI 96822, USA.
- 55 ²¹Analytical Technology, Horiba Techno Service Co., Ltd.; Kyoto 601-8125, Japan.
- 56 ²²Earth and Planetary Science, The University of Tokyo; Tokyo 113-0033, Japan.
- 57 ²³School of Earth and Environmental Sciences, The University of Queensland; St Lucia QLD
58 4072, Australia.
- 59 ²⁴Kochi Institute for Core Sample Research, JAMSTEC; Kochi 783-8502, Japan.
- 60 ²⁵Earth and Planetary Sciences, Kyoto University; Kyoto 606-8502, Japan.
- 61 ²⁶Geoscience, University of Wisconsin-Madison; Madison, WI 53706, USA.
- 62 ²⁷Max Planck Institute for Solar System Research; 37077 Göttingen, Germany.
- 63 ²⁸Earth, Planetary, and Space Sciences, UCLA; Los Angeles, CA 90095, USA.
- 64 ²⁹Thermal Analysis, Rigaku Corporation; Tokyo 196-8666, Japan.
- 65 ³⁰Department of Space Studies, Southwest Research Institute, Boulder, CO 80302, USA.
- 66 ³¹Astromaterials Research and Exploration Science, NASA Johnson Space Center; Houston,
67 TX 77058, USA.
- 68 ³²Earth-System Sciences, Korea Polar Research Institute; Incheon 21990, Korea.
- 69 ³³Centre de Recherches Péetrographiques et Géo-chimiques, CNRS - Université de Lorraine;
70 54500 Nancy, France.
- 71 ³⁴CAS Key Laboratory of Crust-Mantle Materials and Environments, University of Science
72 and Technology of China, School of Earth and Space Sciences; Anhui 230026, China.
- 73 ³⁵Department of Earth Sciences, Natural History Museum; London, SW7 5BD, UK.
- 74 ³⁶IIL, Hokkaido University; Sapporo 001-0021, Japan.

- 75 ³⁷Institute for Geochemistry and Petrology, Department of Earth Sciences, ETH Zurich,
76 Zurich, Switzerland.
- 77 ³⁸Earth and Space Science, Osaka University; Osaka 560-0043, Japan.
- 78 ³⁹Spectroscopy and Imaging, Japan Synchrotron Radiation Research Institute; Hyogo 679-
79 5198 Japan.
- 80 ⁴⁰School of Earth and Space Exploration, Arizona State University; Tempe, AZ 85281, USA.
- 81 ⁴¹Geology, University of Maryland, College Park, MD 20742, USA.
- 82 ⁴²Graduate School of Natural Science and Technology, Okayama University; Okayama 700-
83 8530, Japan.
- 84 ⁴³Earth and Planetary Sciences, University of California; Davis, CA 95616, USA.
- 85 ⁴⁴Science and Engineering, National Museum of Nature and Science; Tsukuba 305-0005,
86 Japan.
- 87 ⁴⁵Chemistry, Tokyo University of Science; Tokyo 162-8601, Japan.
- 88 ⁴⁶School of Earth Sciences and Engineering, Nanjing University; Nanjing 210023, China.
- 89 ⁴⁷Department of Earth Science, Tohoku University; Sendai, 980-8578, Japan.
- 90 ⁴⁸Department of Earth and Planetary Sciences, Kyushu University; Fukuoka 819-0395, Japan.
- 91 ⁴⁹Earth and Planetary Systems Science Program, Hiroshima University; Higashi-Hiroshima,
92 739-8526, Japan.
- 93 ⁵⁰Kanagawa Institute of Technology; Atsugi 243-0292, Japan.
- 94 ⁵¹UTokyo Organization for Planetary and Space Science, University of Tokyo; Tokyo 113-
95 0033, Japan.

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(*corresponding author: paquet@ipgp.fr, moynier@ipgp.fr)

98

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106 **Abstract** (162 words)

107

108 **Abstract (162 words)**

109 Initial analyses showed that asteroid Ryugu's composition is close to CI (Ivuna-like)
110 carbonaceous chondrites – the chemically most primitive meteorites, characterized by near-solar
111 abundances for most elements. However, some isotopic signatures (e.g., Ti, Cr) overlap with other
112 carbonaceous chondrite (CC) groups, so the details of the link between Ryugu and the CI
113 chondrites are not fully clear yet. Here we show that Ryugu and CI chondrites have the same zinc
114 and copper isotopic composition. As the various chondrite groups have very distinct Zn and Cu
115 isotopic signatures, our results point at a common genetic heritage between Ryugu and CI
116 chondrites, ruling out any affinity with other CC groups. Since Ryugu's pristine samples match
117 the solar elemental composition for many elements, their Zn and Cu isotopic compositions likely
118 represent the best estimates of the solar composition. Earth's mass-independent Zn isotopic
119 composition is intermediate between Ryugu/CC and non-carbonaceous chondrites, suggesting a
120 contribution of Ryugu-like material to Earth's budgets of Zn and other moderately volatile
121 elements.

122

123 **Main text**

124 **Introduction.** Ivuna-type (CI) carbonaceous chondrites (CCs) have elemental abundances that
125 are the closest to the composition of the solar photosphere (e.g., [1]) (the exceptions being H, C,
126 N, O, Li and the noble gases). Thus, the CIs are key reference samples for investigating how early
127 Solar System processes shaped the compositions of the planets and their building blocks. The
128 return of the Hayabusa2 spacecraft in December 2020, after two successful touchdown and
129 sampling events on the Cb-type asteroid (162173) Ryugu^{2,3}, offers the unprecedented opportunity
130 to study volatile element fractionation processes using samples unaffected by terrestrial alteration,
131 in particular water incorporation. Initial studies on bulk chemical and isotopic compositions
132 revealed similarities between Ryugu and CIs⁴⁻⁷. However, Ryugu samples exhibit slightly higher
133 $\Delta^{17}\text{O}$ than the average from other CI samples, Orgueil and Ivuna, which is interpreted in terms of
134 original heterogeneity between small samples, or contamination of the meteorites by terrestrial
135 water incorporated into the structure of the alteration minerals (e.g., phyllosilicates, sulfates, iron
136 oxides and hydroxides), not adsorbed to the surfaces⁵. Similarly, although Ti and Cr isotope
137 compositions show that asteroid Ryugu formed in the CC reservoir, it was not possible to establish
138 a clear genetic link to just one of the CC groups because the Cr and Ti isotopic compositions of
139 Ryugu overlap not only with Ivuna-like (CI) but also with the Bencubbin-like (CB)⁵, Renazzo-like
140 (CR)⁵ and High-iron (CH) groups. However, the low volatile contents of these three groups of
141 meteorites, as well as the metal-rich nature of the CB and CH chondrites, argue against any affinity
142 with Ryugu^{5,6}. Thus, because CI chondrites and Ryugu samples share the same Ti and Cr
143 nucleosynthetic signatures, as well as similar mineralogical and elemental compositions^{5,6}, it has
144 been proposed that they formed contemporaneously from the same outer Solar System reservoir^{3,5-}
145 ⁷.

146

147 Material akin to carbonaceous chondrites such as Ryugu and the parent body of the CIs could
148 have delivered significant fractions of the moderately and highly volatile elements present in inner
149 Solar System planets (e.g., 8-13). Because Ryugu samples have been handled carefully to avoid
150 possible contamination, they are ideally suited to estimate the solar composition and assess the
151 contribution of these outer Solar System objects to the inventory of volatile elements in the
152 terrestrial planets. Highly volatile and moderately volatile elements are defined as elements with
153 50% condensation temperatures (T_c) <665 K and 665–1135 K, respectively, under canonical
154 nebular gas conditions at 10^{-4} bar (e.g., 14). Zinc and Cu are ideal elements to investigate volatility-
155 related processes, such as volatile element loss, during planetary accretion¹⁵, and are classified as
156 moderately volatile elements (MVE) (with T_c of 726 K and 1037 K, respectively)^{14,16}.
157 Carbonaceous chondrite groups display distinct Zn and Cu isotopic mass fractionation effects (e.g.,
158 17-21), defining a trend from CIs to CKs (CK = Karoonda-like), with the CIs being the most
159 volatile-rich and isotopically heaviest (for both Zn and Cu) of the CC groups. We have measured
160 the Zn and Cu isotopic compositions of Ryugu samples to (i) verify the link between Ryugu and
161 CI chondrites for moderately volatile elements, and (ii) assess the contribution of Ryugu-like
162 material to the inventory of moderately volatile elements in Earth.

163

164 **Results.** The Zn and Cu isotope compositions for four Ryugu samples (see **Methods section**),
165 together with six CC samples [Alais (CI), Allende A and B (CV), Murchison (CM), Orgueil (CI),
166 Tagish Lake (C2-ungrouped) and Tarda (C2-ungrouped)], were determined following the same
167 analytical protocol, and on the same samples as in [5] (**Table 1**). Most of these CC samples have
168 previously been characterized for their Zn and Cu isotopic composition^{17-19,21}, except Tarda. The

169 isotopic compositions are given as the permil deviations from the JMC-Lyon Zn and NIST
 170 SRM976 Cu standards:

$$171 \quad \delta^x\text{Zn} = \left[\frac{\left(\frac{x\text{Zn}}{64\text{Zn}}\right)_{\text{Sample}}}{\left(\frac{x\text{Zn}}{64\text{Zn}}\right)_{\text{JMC-Lyon}}} - 1 \right] \times 1000 \quad (1)$$

172 where x = 66, 67 and 68.

$$173 \quad \delta^{65}\text{Cu} = \left[\frac{\left(\frac{65\text{Cu}}{63\text{Cu}}\right)_{\text{Sample}}}{\left(\frac{65\text{Cu}}{63\text{Cu}}\right)_{\text{SRM976}}} - 1 \right] \times 1000 \quad (2).$$

174 During the course of this study, the two standards gave $\delta^{66}\text{Zn}$ of 0.00 ± 0.005 ‰ (2SE; n = 163;
 175 JMC-Lyon) and $\delta^{65}\text{Cu}$ of 0.00 ± 0.02 ‰ (2SE; n = 54; NIST SRM976). Zinc isotope measurements
 176 are also corrected for mass-dependent fractionation using the exponential law²², with the
 177 normalizing ratio of $^{68}\text{Zn}/^{64}\text{Zn}$ of 0.3856²³. Zinc isotopic anomalies are quantified using the epsilon
 178 notation relative to the JMC Lyon standard, as follows:

$$179 \quad \varepsilon^{66}\text{Zn}_c = \left[\frac{\left(\frac{66\text{Zn}}{64\text{Zn}}\right)_{\text{Sample}}}{\left(\frac{66\text{Zn}}{64\text{Zn}}\right)_{\text{JMC-Lyon}}} - 1 \right] \times 10^4 \quad (3)$$

180 where c is the normalizing ratio $^{68}\text{Zn}/^{64}\text{Zn}$.

181

182 The Ryugu samples span a very limited range of mass-dependent Zn isotopic compositions
 183 with $\delta^{66}\text{Zn}$ from $+0.41 \pm 0.06$ ‰ to $+0.45 \pm 0.02$ ‰ (2SD), with an average value of $+0.43 \pm 0.05$
 184 ‰ (2SD, n = 48) (**Fig. 1a and Table 1**). The $\delta^{65}\text{Cu}$ values for Ryugu samples range from $0.00 \pm$
 185 0.08 ‰ to $+0.09 \pm 0.05$ ‰, (average value of $+0.04 \pm 0.11$ ‰, n = 8, 2SD) (**Fig. 1b and Table 1**).
 186 Zinc and Cu abundances also span limited ranges from 338 ± 4 ppm to 383 ± 6 ppm (average 361
 187 ± 40 ppm, n = 4, 2SD), and from 133 ± 2 ppm to 168 ± 1 ppm (average 147 ± 37 ppm, n = 4, 2SD),
 188 respectively (**Table 2**). These values are higher than the abundances reported for any CI chondrite,

189 consistent with other element abundances for Ryugu samples relative to CIs (**Fig. 1, Table 2**),
190 although Ryugu samples have lower H₂O contents than CIs⁵. It is worth noting that the samples
191 from both landing sites show identical $\delta^{66}\text{Zn}$ values (**Fig. 1a**). In addition, the soluble organic
192 matter (SOM) extractions of samples C0107 and A0106, which was done prior to purification of
193 Zn and Cu, do not seem to have affected the Zn and Cu isotope compositions of the Ryugu samples
194 (see **Methods section**). All the CCs measured in this study have $\delta^{66}\text{Zn}$ and $\delta^{65}\text{Cu}$ values, as well
195 as Zn and Cu abundances, that are consistent with previous studies (e.g., [17-21]), except for the
196 Cu isotopic composition for Allende A, which is more similar to CO chondrites (**Figs. 1b and 2**).
197 We note, however, that until now only one other measurement of Allende has been reported in the
198 literature¹⁹, and so the difference could represent heterogeneity in the different analyzed fractions
199 of Allende in this study and in [19]. Similar sample heterogeneities for Cu isotopic compositions
200 have been reported for several fragments of Orgueil²¹. The Ryugu samples exhibit Zn and Cu
201 isotopic compositions that are similar to the Alais and Orgueil samples analyzed in this study (**Figs.**
202 **1 and 2**). This is consistent with previous work on the bulk elemental, isotopic, and mineralogical
203 properties of these samples, which reveal a genetic link between the Ryugu samples and CI
204 chondrites, implying formation from the same outer Solar System reservoir⁴⁻⁶.

205

206 **Discussion.** Earlier work has shown that bulk CC chondrites define negative correlations in
207 plots of $\delta^{66}\text{Zn}$ versus $1/\text{Zn}$ ^{17,18} and $\delta^{65}\text{Cu}$ versus $1/\text{Cu}$ ¹⁹. The variable degree of volatile element
208 depletion among the different CC groups reflect mixing of chemically and isotopically distinct
209 reservoirs during their accretion (e.g., 17-19,24,25). The CI chondrites, along with Ryugu, are the
210 least volatile depleted and isotopically heaviest (for both Zn and Cu) of the CC groups, while the
211 most volatile depleted chondrites, the CVs, are the isotopically lightest (this study; [17-21]) (**Figs.**

212 **1 and 2**). The CC trend is interpreted as the result of mixing of volatile-rich material enriched in
213 heavy Zn and Cu isotopes and volatile-poor material enriched in light Zn and Cu isotopes (e.g.,
214 [17-19]). Similar correlations are observed for other moderately volatile elements in CCs, such as
215 Te^{25} and $\text{Rb}^{26,27}$ and their associated isotope compositions, which is interpreted as mixing between
216 matrix (volatile-rich) and chondrules (volatile-poor) (e.g., [17-19,24,26]). Such mixing between
217 distinct CC reservoirs is also observed in the relationships between $\delta^{66}\text{Zn}$ and nucleosynthetic
218 isotope anomalies, such as $\epsilon^{54}\text{Cr}$ (parts per ten thousand mass-independent variations of the
219 $^{54}\text{Cr}/^{52}\text{Cr}$ ratio relative to a terrestrial standard) (**Fig. 3**) (e.g., [18]). Ryugu and the CIs have similar
220 Cu and Zn mass-dependent isotopic compositions and differ markedly from the CBs and CHs
221 (**Figs. 1, 2 and 3**). We can, therefore, exclude any genetic relationship to the CB or CH groups for
222 the Ryugu samples. A shared nucleosynthetic heritage between Ryugu and CI chondrites has been
223 established based on their identical Ti and Cr nucleosynthetic isotope anomalies^{5,6}. Our Zn and Cu
224 results show that this parentage extends to mass-dependent fractionation of moderately volatile
225 elements, strengthening the link between CI chondrites and Ryugu (**Figs. 1, 2 and 3**). The near
226 solar Zn and Cu relative abundances of the Ryugu samples, which are free of the potential
227 ambiguities of terrestrial alteration, suggests that the Zn and Cu isotopic compositions measured
228 for Ryugu and the CI chondrites most likely preserved the proto-Sun's composition¹ (**Fig. 1c**).

229

230 Our study also provides evidence for mass-independent Zn isotope variations ($\epsilon^{66}\text{Zn}$) in
231 Ryugu samples (**Fig. 4 and Table 1**). These Zn isotopic anomalies are consistent with previous
232 observations^{28,29}. While non-carbonaceous chondrites (NCs) display negative $\epsilon^{66}\text{Zn}$ (ordinary
233 chondrites: -0.21 ± 0.04 ‰, 2SE, $n = 12$, [28,29]; enstatite chondrites: -0.19 ± 0.08 ‰, $n = 8$,
234 [28,29]), the Ryugu samples and all CCs exhibit identical positive $\epsilon^{66}\text{Zn}$ within error ($+0.33 \pm 0.04$

235 ‰, 2SE, with $n = 7$ for Ryugu (**Table 1**) and $+0.39 \pm 0.07$ ‰, $n = 7$ for CCs, respectively) with
236 the value previously reported for CC of $+0.28 \pm 0.04$ ‰ (2SE, $n = 11$) [28,29]. It is worth noting
237 that the first replicate of sample C0108 (measured at 100 ppb Zn) has an $\epsilon^{66}\text{Zn}$ of -0.21 ± 0.17
238 ‰, whereas the second C0108 replicate (measured at 250 ppb Zn) has an $\epsilon^{66}\text{Zn}$ of $+0.35 \pm 0.10$
239 ‰ similar to all other Ryugu samples (see **Methods section**): the first replicate is thus considered
240 an outlier as it was analyzed at the lower concentration of 100 ppb and was excluded from the
241 mean value reported here. The reference geological material BHVO-2 and the Zn standard solution
242 IRMM 3702 measured during the first and second sessions have $\epsilon^{66}\text{Zn}$ (-0.07 ± 0.15 ‰, 2SE, n
243 $= 12$; $+0.02 \pm 0.11$ ‰, 2SE, $n = 7$, respectively), consistent within error with estimates for bulk
244 Earth ($+0.015 \pm 0.075$ ‰, 2SE, $n = 4$ [28] and -0.07 ± 0.013 ‰, 2SE, $n = 3$ [29]). There are no
245 known terrestrial processes which can mass-independently fractionate Zn isotopes. The positive
246 $\epsilon^{66}\text{Zn}$ values in the Ryugu samples, therefore, reinforce their genetic link with the CCs (**Fig. 4**).
247 Thus, the difference between the CCs and NCs, originally identified O and Cr isotope
248 compositions³⁰, and later with Ti, Ni and Mo anomalies³¹⁻³⁶, appears to also hold for Zn isotopes.

249

250 Because meteorites show a large variability of isotope anomalies³⁷ and planetary accretion
251 is stochastic³⁸⁻³⁹, it is likely that Earth's composition does not reflect accretion from a single type
252 of material, both in terms of isotopic and elemental compositions. Although enstatite chondrites
253 are isotopically closest to the Earth⁴⁰, their chemical signatures are extreme and deviate
254 substantially from the bulk composition of Earth. Possible mixtures of primitive and thermally
255 processed meteorites or their components (e.g., chondrules; [6, 41-43]) have been proposed to
256 explain the chemical and isotope composition of the Earth^{32,37,44-48}. In particular, the mass-
257 independent isotopic composition of Zn of the Earth appears intermediate between CCs and NCs.

258 Thus, our new data show that CC-like materials, potentially akin to Ryugu, have likely contributed
259 to the delivery of Zn and more generally the volatile elements to the Earth. Thus, following the
260 same approach as in [28,29], and using the average $\epsilon^{66}\text{Zn} = +0.33 \pm 0.04 \text{‰}$ for Ryugu, $+0.35 \pm$
261 0.13‰ for CI [this study, 28, 29], $-0.20 \pm 0.04 \text{‰}$ (2SE, $n = 20$) for NC (ordinary, enstatite from
262 [28,29]) and $-0.02 \pm 0.04 \text{‰}$ for the BSE (2SE, $n = 7$, [28,29]), the mass fraction of Ryugu- or
263 CI-derived Zn in the BSE is estimated 33.5% or 32.2%, respectively. Thus, we find that ~30% of
264 the terrestrial Zn derives from outer Solar System material, while the NC reservoir contributes to
265 ~70% to the terrestrial Zn. Then, to account for the Zn abundances of the accreting materials by
266 Earth, we estimate the mass fractions of NC and Ryugu-like or CI-like bodies accreted by Earth
267 using the Zn abundance of the BSE of $53.5 \pm 2.7 \text{ ppm}$ [49], and the $[\text{Zn}]_{\text{Ryugu}}$ of $361 \pm 40 \text{ ppm}$ [this
268 study] and $[\text{Zn}]_{\text{CI}}$ of $309 \pm 43.8 \text{ ppm}$ [this study, 17,18,21]. Thus, up to ~5% of Ryugu-like material
269 (or ~6% of CI-like material) might be needed to account for Earth's Zn isotopic composition,
270 consistent with estimations from previous studies on Zn isotopic anomalies^{28,29} and representing a
271 substantial contribution to the terrestrial budget of moderately volatile elements^{11,28,29}.

272

273 **Methods**

274 **Major and trace elements.** Zinc and copper isotopic compositions were measured in four
275 samples from the asteroid (162173) Ryugu [C0108, C0107, A0106-A0107 and A0106]. Fragments
276 A0106-A0107 and A0106 are coming from the first touchdown site, and C0108 and C0107 from
277 the second touchdown site⁴⁻⁶. Samples A0106-A0107 and C0108 were pristine samples, whereas
278 A0106 and C0107 were treated for Soluble Organic Matter (SOM) extraction before chemical
279 purification (see **Supplementary Table 1**). In addition, six CCs [Alais, Allende A, Allende B,
280 Murchison, Orgueil, Tagish Lake and Tarda] were processed following the exact same protocol as
281 the Ryugu samples and were analyzed as controls. For each sample, ~25 mg of powder of all the

282 samples were dissolved at Tokyo Institute of Technology. Elemental abundances were determined
283 using Inductively-Coupled-Plasma Mass-Spectrometry (ICP-MS): major and trace elements for
284 A0106-A0107 and C0108 samples are from [5]. After chemical analysis, the same sample
285 solutions were used to determine Zn and Cu isotopic compositions: Zn fractions were pre-
286 separated, as well as 3% of the bulk rock dissolution for Cu purification which represent about 80-
287 100 ng of Cu.

288
289 **Zinc and copper purification.** All the CC meteorites (except Tarda) have previously been
290 measured for Cu and Zn isotopic compositions and were analyzed as controls. Further chemical
291 purifications of Zn and Cu on the same sample aliquots were conducted at the Institut de Physique
292 du Globe de Paris, using the procedure described by [50] for Zn, and by [51,52] for Cu. For Zn,
293 samples were loaded in 1.5 mol.L⁻¹ HBr on 50 µL of AG1-X8 (200-400 mesh) anion exchange
294 resin in home-made PTFE columns. Matrix elements were washed by further addition of 2 mL of
295 1.5 mol.L⁻¹ HBr, and Zn was eluted using 2 mL of 0.5 mol.L⁻¹ HNO₃. The collected samples were
296 then evaporated to dryness. For Cu, samples were loaded in 1 mL of 7 mol.L⁻¹ HCl on home-made
297 PTFE columns filled with 1.6 mL of AG-MP1 resin. After washing the resin with 8 mL of 7 mol.L⁻¹
298 ¹ HCl, the Cu was collected with 16 mL of 7 mol.L⁻¹ HCl. Both procedures were repeated twice
299 to ensure clean Zn and Cu fraction. Procedural blank is < 0.3 ng of Zn, and 0.6 ng of Cu which is
300 negligible relative to the amount of Zn and Cu in the sample mass analyzed for the Ryugu samples
301 and the CCs.

302
303 **Zinc and Cu measurements.** Zinc and Cu isotope compositions were determined using a
304 Neptune Plus Multi-Collector Inductively-Coupled-Plasma Mass-Spectrometer (MC-ICP-MS) at

305 IPGP, using sample-standard bracketing for instrumental mass bias correction as in [50] for Zn
306 and [51,52] for Cu. Each replicate was analyzed 6–8 times for Zn and 1–5 times for Cu depending
307 on the amount of Cu available for each sample, and the reported errors are the two standard
308 deviations (2SD) of these repeated measurements. For the Zn measurements, the samples were
309 analyzed in two sessions with different sample solution concentrations: one at 100 ppb of Zn and
310 a second one at 250 ppb of Zn, with an uptake of 100 $\mu\text{L}\cdot\text{min}^{-1}$. For the Cu measurements, the
311 samples were analyzed in one session at the concentration of 30 ppb of Cu, with the same uptake.
312 The high purity of the final Zn fraction is needed to remove isobaric and non-isobaric interferences
313 from the signal. Interference on ^{64}Zn by ^{64}Ni is corrected by measuring the intensity of the ^{62}Ni ,
314 assuming natural abundances of Ni isotopes ($^{62}\text{Ni} = 3.63\%$; $^{64}\text{Ni} = 0.93\%$). No N_2 was used during
315 the measurements, as this results in high background on mass 68 from ArN_2 . No interference on
316 mass 68.5 from Ba^{2+} was detected during the sessions. The reference geological material BHVO-
317 2 and the Zn standard solution IRMM 3702 measured during the first and second sessions give
318 values consistent with the literature (e.g., [18,20]). However, during the first session, the Zn
319 fractions were measured at 100 ppb of Zn. All the Ryugu and CC samples had similar positive
320 $\epsilon^{66}\text{Zn}$, except for the first replicate of sample C0108 which had a negative value. In other words,
321 all the samples plot below the mass-dependent equilibrium fractionation line in a $\delta^{68}\text{Zn}$ against
322 $\delta^{66}\text{Zn}$ plot, whereas the C0108 replicate falls above it (**Supplementary Figure 1a**). This motivated
323 our second session of measurements on replicates at higher concentrations (250 ppb of Zn) to
324 ensure that the observed $\epsilon^{66}\text{Zn}$ were not analytical artifacts. The second replicate of sample C0108,
325 analyzed at 250 ppb of Zn, shows the same isotopic signature as the rest of the Ryugu sample set
326 and plots below the mass-dependent fractionation line with a positive $\epsilon^{66}\text{Zn}$ (**Supplementary**

327 **Figure 1b**). In the discussion and associated figures, only the second replicate of sample C0108 is
328 considered and represented.

329

330 **Data availability**

331 All data referred to in this article can be found in the tables or source data.

332

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340

341 **Author Contributions**

342 F.M., M.P. and T.Y. designed the project. H.Y. and T.Y coordinated the isotopic analyses of the
343 samples among members of the Hayabusa2-initial-analysis chemistry team. M.P. and T.Y.
344 processed the samples and separated the Zn and Cu from the matrix. M.P. measured the Zn and
345 Cu isotopic compositions. M.P. and F.M. wrote the first draft of the manuscript, with contributions
346 from T.Y., W. D., Y. H., Y. A., J. A., C. M. O'D. A., S. A., Y. A., K. B., M. B., A. B., R. W. C.,
347 M. C., B.-G. C., N. D., A. M. D., T. D. R., W. F., R. F., I. G., M. K. H., Y. H., H. Hi., H. Ho., P.
348 H., G. R. H., K. I., T. I., T. R. I., A. I., M. I., S. I., N. K., N. T. K., K. K., T. K., S. K., A. N. K.,
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351 Q.-Z. Y., S. Y., E. D. Y., H. Y., A.-C. Z., T. N., H. N., T. N., R. O., K. S., H. Y., M. A., A. M., A.
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353

354 **Competing Interests Statement**

355 The authors declare no conflicts of interest.

356 **Tables**

357 Table 1. Zinc and copper stable isotope of Ryugu samples and carbonaceous chondrites.

Sample	Type	n ^a (Zn)	$\delta^{66}\text{Zn}$ (‰)	2SD ^b	$\delta^{67}\text{Zn}$ (‰)	2SD ^b	$\delta^{68}\text{Zn}$ (‰)	2SD ^b	$\epsilon^{66}\text{Zn}$	2SE ^b	n ^a (Cu)	$\delta^{65}\text{Cu}$ (‰)	2SD ^b
Ryugu													
C0108 ^c		<u>8</u>	<u>0.44</u>	<u>0.05</u>	<u>0.70</u>	<u>0.10</u>	<u>0.91</u>	<u>0.09</u>	<u>-0.21</u>	<u>0.17</u>	5	0.00	0.08
Rpt		6	0.44	0.05	0.60	0.05	0.79	0.09	0.35	0.10			
C0107		8	0.42	0.06	0.57	0.10	0.75	0.09	0.37	0.11			
Rpt		6	0.41	0.05	0.60	0.10	0.76	0.13	0.29	0.11			
<i>Average site C</i>		20	0.43	0.05	0.62	0.13	0.81	0.17	0.34	0.06	5	0.00	0.08
A0106-A107		8	0.45	0.02	0.62	0.06	0.80	0.05	0.42	0.10	3	0.09	0.05
Rpt		6	0.41	0.06	0.59	0.08	0.75	0.11	0.33	0.03			
A0106		8	0.43	0.03	0.63	0.07	0.80	0.06	0.25	0.14			
Rpt		6	0.41	0.06	0.58	0.07	0.76	0.11	0.31	0.05			
<i>Average site A</i>		28	0.43	0.04	0.61	0.08	0.78	0.08	0.33	0.05	3	0.09	0.05
<i>Average Ryugu</i>		48	0.43	0.05	0.61	0.11	0.79	0.14	0.33	0.04	8	0.04	0.11
Carbonaceous chondrites													
Orgueil	CI1	8	0.52	0.06	0.67	0.11	0.91	0.06	0.62	0.15			
Rpt		8	0.52	0.09	0.71	0.09	0.96	0.17	0.33	0.14			
<i>Average Orgueil</i>		16	0.52	0.08	0.69	0.11	0.94	0.14	0.52	0.12	2	0.06	0.11
Alais	CI1	8	0.45	0.07	0.62	0.11	0.82	0.08	0.35	0.19			
Rpt		6	0.44	0.07	0.62	0.10	0.80	0.12	0.35	0.09			
<i>Average Alais</i>		14	0.45	0.07	0.62	0.10	0.80	0.10	0.35	0.11	2	0.17	0.003

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Tagish Lake	C2- ung	8	0.43	0.02	0.55	0.12	0.74	0.06	0.53	0.08			
Rpt		6	0.41	0.08	0.54	0.08	0.73	0.15	0.41	0.07			
<i>Average Tagish Lake</i>		14	0.42	0.06	0.55	0.10	0.73	0.11	0.48	0.06			
Tarda	C2- ung	8	0.46	0.06	0.58	0.09	0.80	0.06	0.50	0.12			
Rpt		6	0.46	0.09	0.64	0.14	0.83	0.15	0.41	0.08			
<i>Average Tarda</i>		14	0.46	0.07	0.60	0.13	0.81	0.11	0.46	0.08	1	-0.40	
Murchison	CM2	8	0.38	0.03	0.52	0.09	0.70	0.06	0.25	0.13			
Rpt		6	0.37	0.08	0.52	0.09	0.67	0.15	0.35	0.16			
<i>Average Murchison</i>		14	0.38	0.05	0.52	0.09	0.69	0.10	0.29	0.10	2	-0.58	0.12
Allende A	CV3	5	0.21	0.03	0.26	0.09	0.37	0.08	0.28	0.17			
Rpt		6	0.22	0.10	0.28	0.16	0.35	0.20	0.47	0.15			
<i>Average Allende A</i>		11	0.22	0.07	0.27	0.13	0.36	0.15	0.38	0.12	2	-0.80	0.08
Allende B	CV3	8	0.24	0.07	0.30	0.14	0.43	0.09	0.23	0.12			
Rpt		6	0.23	0.06	0.31	0.12	0.38	0.18	0.32	0.13			
<i>Average Allende B</i>		14	0.24	0.06	0.31	0.13	0.41	0.14	0.27	0.09			
Reference materials													
IRMM3702		7	0.22	0.04	0.26	0.05	0.43	0.06	0.02	0.11			
BHVO2		12	0.30	0.07	0.46	0.08	0.58	0.10	-0.07	0.15			

^an is the number of measurements

^b2SD is 2 x standard deviation; 2SE is 2 x standard error

^cValue excluded from the averages for Ryugu

Numbers in italic represent averages for Ryugu and the carbonaceous chondrites for the Zn data

358

359 Table 2. Major and trace element compositions of Ryugu samples and carbonaceous chondrites.

Sample	Type	Zn (ppm)	2SD	Cu (ppm)	2SD	Mg (ppm)	2SD	Mg/Zn	Mg/Cu
Ryugu									
C0108*		352	4	156	2	104222	1153	296	668
C0107		383	6	168	1	98823	2890	258	589
<i>Average site C</i>		<i>368</i>	<i>45</i>	<i>162</i>	<i>17</i>	<i>101523</i>	<i>7635</i>	<i>277</i>	<i>628</i>
A0106-A107*		338	4	133	2	106866	1250	316	804
A0106		369	5	130	2	112899	3191	306	870
<i>Average site A</i>		<i>354</i>	<i>45</i>	<i>132</i>	<i>5</i>	<i>109883</i>	<i>8533</i>	<i>311</i>	<i>837</i>
<i>Average Ryugu</i>		<i>361</i>	<i>40</i>	<i>147</i>	<i>37</i>	<i>101509</i>	<i>11700</i>	<i>294</i>	<i>733</i>
Carbonaceous chondrites									
Orgueil	CI1	288	4	131	1	91158	2706	3.0	696
Alais	CI1	298	3	127	1	84683	2489	2.9	667
Tagish Lake	C2-ung	204	5	105	3	107210	1275	1.2	1021
Tarda	C2-ung	201	3	110	2	111296	2388	2.1	1012
Murchison	CM2	174	2	117	1	111130	1257	1.1	950
Allende A	CV3	110	2	97	2	141784	2461	1.7	1459
Allende B	CV3	121	1	105	1	157609	4882	3.1	1501

*Abundances from [5]

Numbers in italic represent averages for Ryugu, and each of Ryugu sample site

360

361

362 **Figures captions**

363 **Figure 1. Zinc and copper elemental and isotopic compositions for Ryugu and carbonaceous**
364 **chondrites samples.** (a) $\delta^{66}\text{Zn}$ vs Mg/Zn , (b) $\delta^{65}\text{Cu}$ vs Mg/Cu and (c) Zn/Mg vs Cu/Mg that we
365 measured for the Ryugu samples (diamonds) and carbonaceous chondrites (large circles with
366 abbreviations: Or=Orgueil, Als=Alais, Tag=Tagish Lake, Tar=Tarda, Mur=Murchison,
367 All=Allende). Small circles are from the literature [17-21] for Zn and Cu isotope compositions
368 (and references therein for major and trace element compositions). The color identifies the type of
369 chondrite as described in the legend. The purple star in panel c represents the CI chondrite
370 composition from [1]. Data are presented as mean values with 2SD error bars, reported in Table
371 1.

372
373 **Figure 2. $\delta^{66}\text{Zn}$ vs $\delta^{65}\text{Cu}$ for Ryugu samples and carbonaceous chondrites.** Literature data are
374 from [17,19]. Same symbols as in Figure 1 for the samples analyzed in this study. Other chondrite
375 groups from the literature are reported directly on the figure. Data are presented as mean values
376 with 2SD error bars, reported in Table 1. For clarity, only the error bars of our measurements are
377 displayed. Error bars for literature data are not shown.

378
379 **Figure 3. $\delta^{66}\text{Zn}$ (this study) vs $\epsilon^{54}\text{Cr}$ [5,30,53-55] for Ryugu samples and carbonaceous**
380 **chondrites.** Literature data are from [17,18,20] for Zn isotope compositions, and from [56] for Cr
381 isotope compositions. The dark and light blue shaded areas correspond to the $\epsilon^{54}\text{Cr}$ ranges for
382 site A and site C, respectively, from [6]. Same symbols as in Figure 1 for the samples analyzed in
383 this study. Other chondrite groups from the literature are reported directly on the figure. Data are
384 presented as mean values with 2SD error bars, reported in Table 1.

385
386 **Figure 4. Variations of $\epsilon^{66}\text{Zn}$ among different groups of meteorites.** For comparison purposes,
387 only Ryugu (diamond) and CI (purple circles) samples measured in this study are represented
388 here. Literature data for carbonaceous chondrites ([28] (large symbols), [29] (small symbols)),
389 ordinary chondrites [28,29], enstatite chondrites [28,29], NC and CC iron chondrites [29],
390 ureilites [29] are shown with gray symbols. Bulk Silicate Earth: $+0.015 \pm 0.075 \text{‰}$, 2SE, $n = 4$
391 [28] and $-0.07 \pm 0.013 \text{‰}$, 2SE, $n = 3$ [29]. Data are presented as mean values with 2SE error
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