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Oxygen isotope evidence from Ryugu samples for early water delivery to Earth by CI chondrites

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The delivery of water to the inner Solar System, including Earth, is still a debated topic. A preferential role for hydrated asteroids in this process is supported by isotopic measurements. Carbonaceous chondrite (CC) meteorites represent our main source of information about these volatile-rich asteroids. However, the destruction of weaker materials during atmospheric entry creates a bias in our CC data. The return of surface materials from the C-type asteroid 162173 Ryugu by the Hayabusa2 spacecraft provides a unique opportunity to study high-porosity, low-density, primitive materials, unrepresented in the meteorite record. We measured the bulk oxygen isotope composition from four Ryugu particles and show that they most closely resemble the rare CI (CC Ivuna-type) chondrites, but with some differences that we attribute to the terrestrial contamination of the CI meteorites. We suggest that CI-related material is widespread among carbonaceous asteroids and a more important source of Earth's water and other volatiles than its limited presence in our meteoritic collection indicates.

Between June 2018 and November 2019, the JAXA Hayabusa2 spacecraft made detailed spectroscopic observations and measurements of the C-type asteroid 162173 Ryugu. Material from two different locations on the asteroid was collected and returned to Earth on 6 December 2020 (ref. [1]). One sample was stored in Chamber A of the return capsule and the other, collected close to an impactor-formed crater, was stored in Chamber C. Near-IR spectroscopic data obtained during orbital observations of asteroid Ryugu indicated that it was composed of material "similar to thermally and/or shock-metamorphosed carbonaceous chondrite meteorites" (refs. [2,3]), with a potential match to the CY (Yamato-type) chondrites[4]. In contrast to this interpretation, initial curation studies at the JAXA ISAS facility suggested that the returned

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samples were "most similar to CI chondrites" (ref. [1]). These contradictory classifications can only be resolved by detailed characterization studies of the Ryugu particles. In particular, high-precision oxygen isotope analysis is widely recognized as the most powerful technique for establishing the interrelationships between individual samples and well-characterized meteorite groups. The results presented here provide a firm basis for evaluating the relationship between the Ryugu samples and the carbonaceous chondrite (CC) meteorite inventory.

Results

Subsamples from four distinct Ryugu particles were analysed for their bulk oxygen isotopic compositions by laser fluorination, using a 'single-shot' technique[5,6] (Methods). Three of the four analysed samples were from Chamber C (C0014,21; C0068,21; C0087,2) and one from Chamber A (A0098,2). Sample transport, loading and analysis techniques used in this study ensured that at no time were the particles exposed to atmospheric contamination (Methods).

The four particles from which the analysed material was extracted consist predominantly of fine and coarse-grained phyllosilicates, varying between approximately 64 and 88 vol% (ref. [7]) (Figs. 1a,b). Anhydrous silicates (olivine and pyroxene) have not been observed in any of these four particles, but rare examples have been identified in other Ryugu particles[8,9]. Phyllosilicates comprise a serpentine-saponite intergrowth and have bulk compositions that fully overlap with those found in CIs[7]. Carbonate minerals, mainly dolomite, with minor Ca-carbonate and breunnerite, are present in highly variable amounts (approximately 2 to 21 vol%)[7]. Magnetite (approximately 3.6 to 6.8 vol%), as framboids, plaquettes and spherical aggregates, and sulfide minerals (approximately 2.4 to 5.6 vol%) are also present within the phyllosilicate-rich matrix (Fig. 1a,b)[7,9]. Ryugu particles have a high average porosity of 41% and consequently a low average density of 1,528 ± 242 kg m^{-3}, comparable to that of the CI chondrite Orgueil or the ungrouped primitive meteorite Tagish Lake[9].

Seven individual analyses were undertaken on material extracted from the four Ryugu particles (Methods). The mass of material analysed varied from 0.18 to 1.83 mg (Table 1). For comparison with the Ryugu particles, the CI chondrites Orgueil, Ivuna and Alais and the CY chondrites[4] Y-82162 and B-7904 were also analysed as part of this study (Table 1). Due to the wide range of Ryugu particle masses analysed in this study, a weighted average has been calculated for comparison purposes with the CY and CI chondrites (Table 1). Unless otherwise specified, in the text and figures, the weighted Ryugu composition has been compared to the unweighted averages for the CIs and CYs. Weighted and unweighted average data for all samples analysed in this study are given in Table 1.

The mean oxygen isotopic composition (weighted) of the seven Ryugu analyses are shown in Fig. 2 along with data for potentially related CC groups. The Ryugu particles have a mean oxygen isotope composition that overlaps with that of the CI chondrites, but is considerably lighter with respect to δ^{18}O than the CYs (Fig. 2). A possible match between Ryugu samples and CIs has also been suggested on the basis of bulk oxygen isotope data in two other recent studies[9,10].

δ^{18}O variation in Ryugu particles

Individual Ryugu analyses show a large range in δ^{18}O values, from 11.46 to 19.30‰ (Table 1 and Fig. 3). The largest subsample assigned for oxygen isotope analysis came from particle C0014 and had a total initial mass of 5.5 mg, permitting multiple measurements (n = 4) (total analysed mass so far 3.3 mg) (Table 1). Analyses of C0014 have δ^{18}O values that vary from 13.73 ± 0.08‰ (2 s.d.) to 19.30 ± 0.07‰ (2 s.d.). The relatively large range in δ^{18}O values displayed by Ryugu particles reflect intrinsic isotopic heterogeneity at the sampling scale involved. Note that detailed mineralogical studies[7,9] show a considerable level of heterogeneity within individual Ryugu particles (Fig. 1a,b). Analysis of individual mineral phases in Ryugu particles

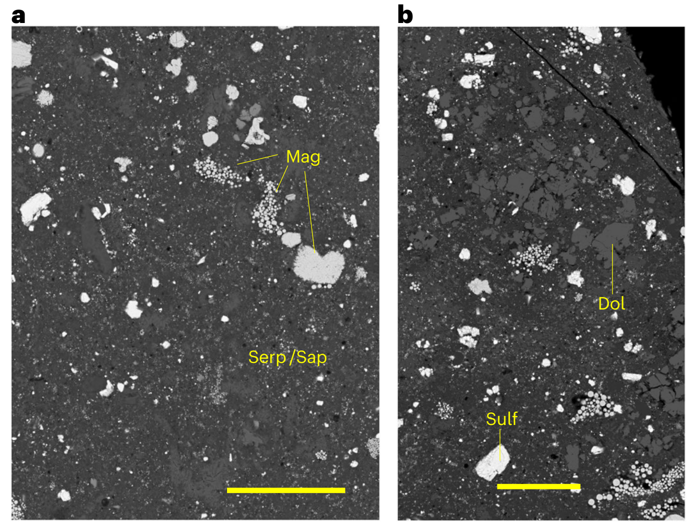

Fig. 1 | Backscattered images from two areas of particle C0068 illustrating the heterogeneous nature of the Ryugu material[7]. a, Magnetite (Mag) occurs both as clusters of framboids and spherical aggregates. The dominant, fine-grained dark material comprises intergrown serpentine and saponite (Serp/Sap). The fine-grained dark matrix represents intergrown serpentine and saponite. **b**, A large number of relatively coarse-grained dolomite crystals (Dol) are present along the right-hand edge. Sulfide crystals (Sulf) of varying grain sizes can also be seen. Scale bars, 25 μm.

by secondary ion mass spectrometry has revealed a large variation in δ^{18}O, with magnetite in the range −5.3 to 7.4‰, dolomite 25.4 to 41.6‰ and Ca-carbonate 34.2 to 39‰ (refs. [9–11]). As the dominant phase in Ryugu particles (64 to 88 vol.%), phyllosilicates are likely to have a δ^{18}O composition that is relatively close to the mean bulk value of 15.88‰ determined in this study (Table 1). This value is within the range determined for CI matrix separates[12]. In view of the heterogeneity of Ryugu particles and the wide variation in δ^{18}O displayed by different mineral phases, the range of values measured in this study is not unexpected. Where bulk oxygen isotope values have been determined on mg-sized fractions taken directly from CC meteorites, it is common to obtain a range of δ^{18}O values similar to, or exceeding, those obtained in this study (Methods).

Calculations based on measured modal data for Ryugu particles[7] and oxygen isotope analysis of Ryugu mineral phases[10] yielded bulk δ^{18}O values between 9.7 and 18.9‰ (Supplementary Information), which is close to the range determined in this study. The overall range of δ^{18}O values measured in Orgueil (14.39 to 16.62‰) and Y-82162 (20.77 to 24.47‰) is greater than would be anticipated on the basis of our measured system precision (±0.1‰). These meteorites were run as relatively coarse-grained powders to reflect the overall grain size of the Ryugu material and were not ground to very fine powders as required for complete homogenization. Intense grinding would probably modify their primary compositions, for example there is the potential for substantial changes to the water content of these hydrated samples.

The single particle analysed from chamber A (A0098) has the lowest δ^{18}O value (11.46 ± 0.12‰ (2 s.d.)) of all the Ryugu material analysed in this study, and one of the lowest Δ^{17}O values (0.56 ± 0.06‰ (2 s.d.)). However, it is also one of the smallest samples analysed here, and as such one of the most susceptible to the effects of sampling a heterogeneous mineralogy. Other small samples also show some of the largest variations, for example C0014-3 and C0087 with Δ^{17}O values of 0.54 and 0.75, respectively (Table 1). Therefore, the most likely explanation for the low δ^{18}O value of A0098 is sample heterogeneity, possibly reflecting a higher modal magnetite content.

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Ryugu analyses compared to CI and CY chondrites

Despite the large variation in $\delta^{18}O$ values displayed by the Ryugu particles, they tend to cluster close to our analyses of Orgueil and Alais in Fig. 3. Ivuna has a $\delta^{18}O$ composition that falls within the Ryugu range, but has a lower $\Delta^{17}O$ value. The Ryugu particles are isotopically distinct from the CY chondrites in respect of both $\delta^{18}O$ and $\Delta^{17}O$ (Fig. 3). The two CYs analysed in this study (Y-82162 and B-7904) have similarly high $\delta^{18}O$ values, but their $\Delta^{17}O$ values are distinct. This suggests that the CYs, as currently defined[4], are not a single homogeneous group. This finding merits further investigation but does not alter the principal conclusion of this study that Ryugu particles are closely related to CI chondrites. This similarity is well demonstrated when the weighted mean composition of the Ryugu particles (weighting by mass of O_2 gas liberated during fluorination) is compared to the mean CI value (Fig. 3), $\delta^{18}O$ being 15.88 ± 4.85‰ (2 s.d.) and 15.16 ± 4.05‰ (2 s.d.), respectively (Table 1). The mean $\Delta^{17}O$ value for the CIs (0.53 ± 0.21‰ 2 s.d.) is lower than the weighted mean value for the Ryugu particles (0.66 ± 0.09‰ (2 s.d. weighted)), but there is clearly significant overlap at the 2 s.d. level (Fig. 3).

Ryugu particles analysed in other studies[9,10] overlap, or have similar oxygen isotope compositions to those obtained here, with the notable exception of one analysis[10] that lies at the edge of the CY* field in Fig. 4. This value is not thought to reflect analytical differences with the other laboratory involved in this joint study, but instead is attributed to small scale intrinsic heterogeneity within the Ryugu regolith[10]. Previous oxygen isotope analyses of CY chondrites[13] provide additional support for the possibility that these meteorites respresent two distinct groups (Fig. 4).

Discussion

It is clear from our oxygen isotope data that there is a much stronger case for a connection between the Ryugu particles and CIs than with CYs (Figs. 2–4). This potential relationship is also supported by detailed mineralogical and petrological studies of Ryugu material[7,9,10]. Differences between the Ryugu particles and CIs that have been identified so far probably reflect terrestrial alteration of the latter. It is well documented that Orgueil, which fell in 1864, has undergone considerable mineralogical modifications due to terrestrial weathering[14] and this would necessarily result in the incorporation of atmospheric oxygen and so pull the bulk $\Delta^{17}O$ value closer to the terrestrial fractionation line (TFL). This conclusion is in keeping with the mineralogical evidence that Ryugu particles do not contain ferrihydrite or sulfate[7,10], whereas Orgueil does[15]. There is also evidence that phyllosilicates in at least some Ryugu samples may lack interlayer water in the saponite component[10]. Stepwise pyrolysis of Orgueil has demonstrated that the interlayer water present in the meteorite is of terrestrial origin, as is also the case for the more recent CC fall Tagish Lake[16].

All the CI chondrites measured in this study have lower mean $\Delta^{17}O$ compositions (Alais 0.60 ± 0.01‰; Ivuna 0.41 ± 0.01‰; Orgueil 0.58 ± 0.08‰) than the weighted mean Ryugu value (0.66 ± 0.09‰ 2 s.d.) (Table 1). We have undertaken calculations using our analyses of Orgueil to examine the possibility that the $\Delta^{17}O$ difference between the Ryugu particles and CIs is the result of terrestrial contamination of the latter (Supplementary Information). Using either the modal composition of Orgueil[15] (method 1), or its full chemical analysis[17] (method 2), these calculations indicate that the measured $\Delta^{17}O$ value of Orgueil (0.58‰) can be fully accounted for in terms of the terrestrial contamination of material with a pre-atmospheric composition identical to that of the Ryugu particles (0.66‰). It is important to note that these calculations do not provide unequivocal confirmation that the $\Delta^{17}O$ differences between the CIs and Ryugu is solely the result of terrestrial contamination of CI chondrites and it remains possible that primary differences between these materials may also be a factor. However, the evidence that Ryugu grains lack interlayer water[10] and that such water in CIs may be of terrestrial origin[16] is consistent with this $\Delta^{17}O$ difference being the result of terrestrial contamination.

The larger $\Delta^{17}O$ difference between Ivuna and Ryugu particles compared to the other CIs may reflect local-scale heterogeneity. Studies have shown that CIs display chemical heterogeneity at sampling scales of less than 1 to 2 g (refs. [18,19]). Such large samples of these important meteorites are rarely available and homogeneous powders are normally based on 100 to 200 mg aliquots. However, despite the possibility of local-scale heterogeneities, all three CI meteorites measured here display lower $\Delta^{17}O$ values than the weighted average for Ryugu. This is consistent with the other lines of evidence indicating that they have experienced a substantial degree of terrestrial contamination. This may have important implications for the use of CI meteorite bulk compositional data as proxies for Solar System values[20]. In addition, as a result of adding a large terrestrial water component, and the realistic possibility of contamination by terrestrially-derived organic molecules, light stable isotope data (C, H, O, N) from CI meteorites needs to be carefully evaluated, as it is likely to include an important non-indigenous component. As a result of their lack of terrestrial contamination, chemical and isotopic data from Ryugu samples will provide a fresh perspective on these bulk Solar System values.

While only 5.4 g of material was collected by the Hayabusa2 spacecraft, initial spectral characterization of the returned samples indicated that they provide a good match to the global average data obtained during orbital observations of Ryugu[1]. Hence, the returned particles are probably representative of the asteroid as a whole. The close match between $\delta^{18}O$ compositions of Ryugu particles and the CIs and the likelihood that both had very similar preterrestrial $\Delta^{17}O$ values provides a firm basis for linking asteroid Ryugu to the CI chondrites.

CI chondrites are a rare group of meteorites, with only nine examples (November 2022) listed on the Meteoritical Bulletin database[21], of which four are probably members of the CY group[4]. This compares to 724 entries (November 2022) for CM2 (Mighei-like) chondrites, the most abundant group of hydrated CCs. However, the apparent paucity of CI-related material arriving on Earth may simply reflect their low-strength characteristics[22]. While CC meteorites represent only about 4% of observed meteorite falls (Meteoritical Bulletin Database)[21] they comprise 55–60% of the micrometeorite population (fragments in the size range 10 μm–2 mm sized), which accounts for most of the 40,000 ± 20,000 metric tons of extraterrestrial material accreted by the Earth each year[23]. CI-related particles have been tentatively identified within the larger-sized fraction of micrometeorites and may be more common among the less well studied, smaller-sized particles[23].

CI chondrites have very short cosmic ray exposure ages, which are generally less than 2 Myr (ref. [24]). Asteroid Ryugu is probably the product of multiple parent body disruption/resurfacing events, but is estimated to have formed in its present 'spinning-top' shape more than 8.5 Myr ago[25]. This raises the possibility that Ryugu may represent the immediate source body to the CIs, including the important meteorites Orgueil, Ivuna and Alais. Ryugu is an Apollo Earth-crossing asteroid with an aphelion of 1.419 astronomical units (AU) and a perihelion of 0.96 AU (ref. [26]). In contrast, calculations of the pre-atmospheric orbit of the Orgueil meteorite suggest an aphelion beyond the orbit of Jupiter[27]. Pre-atmospheric trajectories determined for recent CC falls all have aphelions in the outer main belt[28], consistent with the proposal that near-Earth objects are not a major source of meteorites, with most falls originating directly from the main belt[29]. It therefore seems unlikely that the known CI meteorites originated from Ryugu.

Ryugu is classified as a Cb-type asteroid, possibly derived from either the Eulalia or Polana asteroid families[30]. Bennu, the target asteroid of the OSIRIS-REx mission, is also probably derived from one of these two asteroid families[31]. CI chondrites have been spectrally matched to C, Cb and B-type asteroids[32], which make up about half of all the C-complex bodies in the main belt[30]. These types are also well represented in the inner main belt and probably supply a notable fraction

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Table 1 | Oxygen isotopic compositions of Ryugu samples and carbonaceous chondrites

Sample	Mass (mg)	µg O_2	Percentage Yield	Method	N*	$\delta^{17}O$	±2SD	$\delta^{18}O$	±2SD	$\Delta^{17}O$	±2SD
Hayabusa2 data											
C0014-1	0.78	132	nd	B	60	10.72	0.16	19.30	0.07	0.68	0.14
C0014-2	0.52	89	nd	M	40	8.60	0.11	15.23	0.19	0.68	0.03
C0014-3	0.18	31	nd	M	60	7.69	0.04	13.73	0.08	0.54	0.02
C0014-4*	1.83	311	nd	B	40	9.23	0.09	16.47	0.03	0.67	0.09
C0068	0.5	85	17.00	M	30	7.07	0.06	12.38	0.09	0.63	0.01
C0087	0.29	50	nd	M	60	8.85	0.11	15.57	0.22	0.75	0.03
A0098	0.26	44	nd	M	40	6.52	0.11	11.46	0.12	0.56	0.06
TOTAL	**4.36**	**742**									
Mean value (unweighted)						**8.38**	**2.84**	**14.88**	**5.29**	**0.64**	**0.14**
Mean value (weighted)						**8.92**	**2.58**	**15.88**	**4.85**	**0.66**	**0.09**
CY Chondrites											
Y-82162,82 *	1.03	104	10.09	B	40	12.50	0.06	23.06	0.05	0.50	0.07
Y-82162,82 *	0.93	95	10.24	B	40	13.24	0.12	24.47	0.18	0.52	0.05
Y-82162,82 *	0.80	104	13.07	B	40	12.29	0.10	22.97	0.07	0.34	0.09
Y-82162,82 *	0.71	94	13.17	B	40	11.68	0.18	21.56	0.26	0.46	0.09
Y-82162,82 *	0.56	85	15.29	B	40	11.25	0.10	20.77	0.11	0.45	0.11
Y-82162,82 *	0.63	65	10.25	M	20	12.51	0.04	23.12	0.06	0.49	0.02
Y-82162,82 *	0.45	56	12.56	M	20	12.60	0.03	23.38	0.01	0.44	0.04
TOTAL	**5.11**	**603**									
Mean value (unweighted)						**12.29**	**1.30**	**22.76**	**2.45**	**0.46**	**0.12**
Mean value (weighted)						**12.28**	**1.32**	**22.75**	**2.48**	**0.46**	**0.13**
B-7904	**2.16**	**495**	**22.90**	**B**	**40**	**11.08**	**0.030**	**21.37**	**0.030**	**-0.03**	**0.020**
CI Chondrites											
Orgueil*	1.30	301	23.24	B	40	8.81	0.05	15.78	0.03	0.61	0.03
Orgueil*	1.58	364	23.05	B	40	9.23	0.01	16.59	0.02	0.61	0.01
Orgueil*	1.10	261	23.79	B	40	9.14	0.04	16.44	0.03	0.59	0.04
Orgueil*	1.44	321	22.29	B	40	8.00	0.03	14.39	0.02	0.52	0.03
Orgueil*	0.60	221	36.83	B	40	8.34	0.05	14.79	0.03	0.64	0.05
Orgueil*	0.60	132	22.00	M	40	8.12	0.12	14.56	0.04	0.55	0.12
Orgueil*	1.74	394	22.64	B	30	8.93	0.03	16.14	0.02	0.54	0.03
Orgueil**	2.38	695	29.20	B	30	9.24	0.02	16.62	0.02	0.60	0.01
Orgueil**	2.33	553	23.73	B	30	8.41	0.02	15.04	0.01	0.59	0.02
TOTAL	**13.06**	**3242**									
Mean value (unweighted)						**8.69**	**0.97**	**15.59**	**1.81**	**0.58**	**0.08**
Mean value (weighted)						**8.78**	**0.94**	**15.77**	**1.76**	**0.58**	**0.07**
Ivuna	2.06	270	13.09	B	40	8.55	0.03	15.67	0.01	0.41	0.02
Ivuna	2.18	451	20.74	B	40	9.52	0.02	17.51	0.03	0.42	0.03
TOTAL	**4.24**	**721**									
Mean value (unweighted)						**9.04**	**1.36**	**16.59**	**2.60**	**0.41**	**0.01**
Mean value (weighted)						**9.17**	**1.32**	**16.82**	**2.52**	**0.41**	**0.01**
Alais	2.03	377	18.58	B	40	7.60	0.04	13.46	0.01	0.60	0.05
Alais	2.27	471	20.73	B	40	7.08	0.03	12.46	0.05	0.60	0.01
TOTAL	**4.30**	**848**									
Mean value (unweighted)						**7.34**	**0.73**	**12.96**	**1.42**	**0.60**	**0.01**
Mean value (weighted)						**7.31**	**0.72**	**12.91**	**1.41**	**0.60**	**0.01**
CI mean value* **						**8.42**	**1.96**	**15.16**	**4.05**	**0.53**	**0.21**

NOTES All analyses this study except *from Ito et al.[7] and ** from Greenwood et al.[6]. $mass was estimated based on measured 17% yield for particle C68 - see methods for further details. BA = Bellows analysis; M = microvolume. N* = Total number of sample/reference gas comparisons. Quoted errors are 2SD based on the compositions calculated for each block of 10 sample to reference gas comparisons. *** CI mean calculated is the average of the weighted mean values for Orgueil, Ivuna and Alais.

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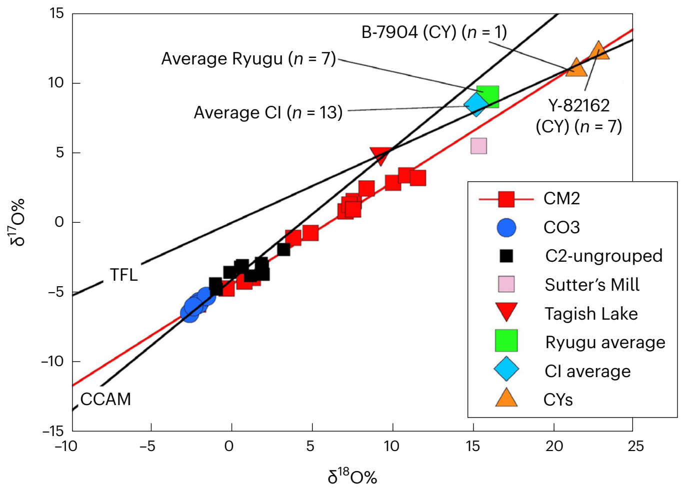

Fig. 2 | Oxygen isotopic composition of the weighted average of the seven Ryugu particles (green square) analysed in this study shown in relation to other relevant CC groups. The figure shows clearly that the Ryugu particles have a mean oxygen isotope composition that is close to that of the CIs (blue diamond), but distinct from the CYs (brown triangles). Data for CIs (Alais, Ivuna and Orgueil) and CYs (B-7904, Y-82162) are given in Table 1. Other data are CO3 chondrites[57] (blue circles), CM2 (red squares) and C2 ungrouped (black squares)[13,58–61], along with analyses for Tagish Lake (inverted red triangle) and Sutter's Mill (pink square) (Supplementary Information). The red line is the best fit line through CM2 (finds and falls) data only. CCAM, CCs anhydrous minerals line[13]. Value *n* refers to the number of individual aliquots of material that were independently run on the laser fluorination line for each sample (Table 1).

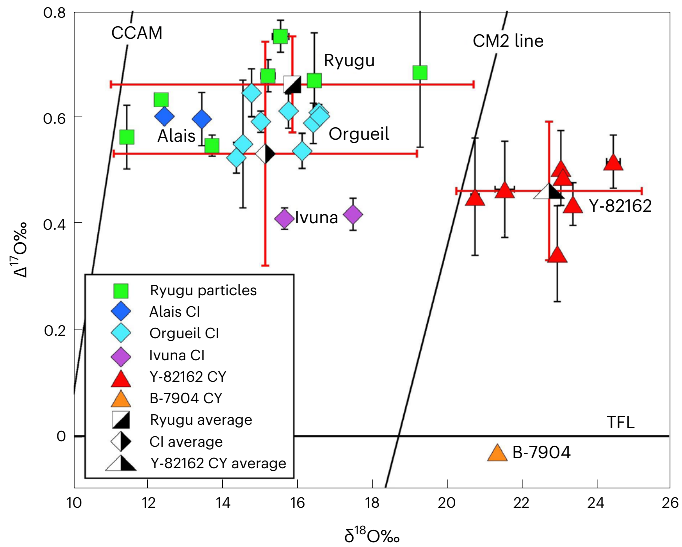

Fig. 3 | Oxygen isotope composition of the samples analysed in this study. Ryugu particles (green squares) show a wide variation in $\delta^{18}O$ values, which reflects intrinsic isotopic heterogeneity at the sampling scale involved (see text for further discussion). CI chondrites (coloured diamonds) and CY chondrites (coloured triangles) were analysed for comparison purposes with the Ryugu particles. In terms of their $\delta^{18}O$ values, the weighted average value for the Ryugu particles and the mean value for the CIs (Alais, Ivuna and Orgueil) are very close in composition (Table 1). In contrast, the mean value for the CY chondrite Y-82162 is substantialy displaced to higher $\delta^{18}O$ values compared to either the Ryugu particles or CIs. CM2 Line is the extension of the best fit line through CM2 falls and finds shown in Fig. 2. Error bars ±2 s.d.

of the extraterrestrial material delivered to Earth[30]. Confirmation of a CI-like composition for asteroid Ryugu provides additional evidence that this material is widespread in the main belt. The likelihood must be that the bulk of CI-like material delivered to Earth is too friable to withstand atmospheric entry and so does not show up in the meteorite record. This is potentially important for the delivery of volatiles to the inner Solar System, as CI chondrites are the most hydrated of all the CC meteorites[17]. Even after subtraction of the potentially contaminated interlayer water, CI chondrites have water contents higher than CM2s (ref. [17]). While CM chondrites show a strong link to Ch asteroids, they also have affinities to all other C-complex classes[32]. A mix of CI and CM chondrites seems a strong possibility for hydration of the inner Solar System, with Ryugu data pointing to a more important role for CIs than their paucity as meteorites might suggest.

How the Earth gained its water remains an outstanding issue in planetary science[33]. While a small fraction may have been inherited from the protosolar nebula, modelling and isotopic studies suggest this may only be about 1%, with the remaining 99% delivered by CCs during the main phase of Earth's accretion[33–35]. CIs, as well as CMs, have H and N isotopic compositions close to that of the bulk Earth, whereas cometary sources are isotopically far removed from terrestrial values[35]. CIs are the only meteorite group that shows a close match to Earth's nucleosynthetic Fe isotope composition ($\mu^{54}Fe$)[36]. On the basis of the evidence from Ryugu discussed above, the uncontaminated $\Delta^{17}O$ composition of CIs of 0.66‰ makes them the hydrated CC group with the closest oxygen isotopic composition to Earth. However, there remains considerable uncertainty about how much water is present on Earth and it is possible that other meteorite groups contributed to its water budget. Enstatite chondrites have been proposed as a potential source of Earth's water[37]. However, indigenous water in enstatite chondrites is substantially lower than in CCs[17]. So, while enstatite chondrites could have supplied up to three ocean masses (one ocean mass 1.38×10^{21} kg), this would be at the lower end of terrestrial water estimates, which might be up to 18 ocean masses[38], with experimental evidence indicating considerable amounts of hydrogen partitioned into Earth's core[39].

CI chondrites, and CI-related asteroids such as Ryugu, are highly altered (Fig. 1), having experienced extensive parent body hydrothermal processing, such that only traces remain of their original silicate mineralogy[7–9,15,18,19]. Despite this, CIs are chemically the most primitive CC group, with a bulk composition close to that of the solar photosphere for most elements[18–20]. Although local-scale heterogeneities are present in CIs[18,19], their bulk chemical composition is essentially unfractionated, suggesting that aqueous alteration took place isochemically, potentially under static fluid conditions[40]. In contrast, modelling studies point to major fluid migration and hence open-system behaviour[41]. Recent measurements made on trapped fluid in a Ryugu pyrrhotite crystal indicate that it contains halogens, nitrogen, sulfur, CO_2 and dissolved organic compounds[42]. As in terrestrial systems[43], high solute concentration may have been important in controlling fluid flow by reducing the density contrast with the enclosing silicate material. In addition, the primordial Ryugu parent body may have been small, potentially no more than about 20 km in diameter[11]. Models of CC alteration are generally based on much larger bodies, for example of 50 km radius[44]. Alteration on a small asteroid in which the fluid was stagnant may resolve the contradictions between studies that favour isochemical alteration[18,19,39] and the open-system behaviour predicted by numerical simulations[40].

It has been proposed that Ryugu may be of cometary origin[9,45], as has also been suggested for Orgueil[27]. Direct measurement of the gaseous atmosphere of Comet 67P/Churyumov-Gerasimenko by the Rosetta spacecraft gave a $\delta^{18}O$ value of close to 120‰ (ref. [46]). While this measurement is subject to very large errors, it suggests that cometary ice is very ^{16}O-depleted. Matrix materials, termed cosmic symplectites, from the primitive ungrouped chondrite Acfer 094 have $\delta^{18}O$ values that are slightly higher than the Comet 67P measurements and are considered representative of the composition of primordial ice[47,48]

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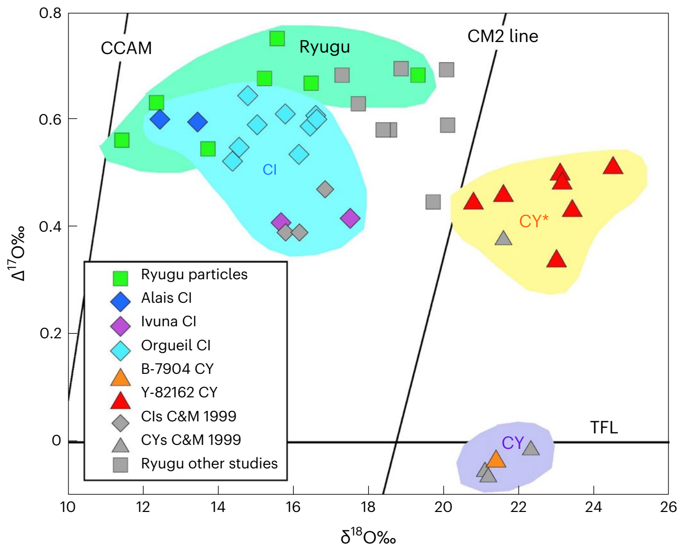

Fig. 4 | Plot of δ^{18}O versus Δ^{17}O for samples analysed in this study (coloured symbols) compared to the results obtained in other studies (grey symbols). Fields for Ryugu particles (green), CI (blue) and CY (yellow and mauve) chondrites are based only on the analyses obtained in this study (Fig. 3). Ryugu particles analysed in other studies (grey squares)[9,10] overlap, or are close to, those obtained here, with the exception of one analysis that lies at the edge of the CY* field[10]. Also shown are earlier analyses of CI and CY chondrites[13] (grey diamonds and triangles). CY chondrites appear to represent two distinct groups with similar δ^{18}O values, but distinct Δ^{17}O compositions. As a consequence, the respective CY fields have been labelled CY and CY*. CM2 Line is the extension of the best fit line through CM2 falls and finds shown in Fig. 2.

(Fig. 5). In contrast, the final fluids on the CI/Ryugu parent body/bodies would have evolved to more ^{16}O-rich values, due to protracted exchange with ^{16}O-rich solids[8,9]. There is little evidence that such extensive levels of aqueous alteration have taken place within cometary nuclei, with particles from Comet 81P/Wild 2 sampled during the Stardust mission dominated by a ^{16}O-rich, high temperature assemblage[49]. The evidence from Ryugu and CIs is that their parental sources were early-formed asteroids[7,9,11] that underwent extensive aqueous alteration, in response to the decay of short-lived radionuclides, such as ^{26}Al ($t_{1/2}$ = 0.73 Myr)[43].

Orgueil contains 10.8 wt% structurally-bound water[17] which, on the basis of the results of stepwise pyrolysis[16], is likely to be of extraterrestrial origin. As discussed earlier, the magnitude of Earth's water inventory is poorly constrained[33,37]. Taking a median estimate of ten ocean masses[33] requires a CI contribution of 2.1% to Earth's mass, equivalent to 54× the mass of the asteroid belt. There is debate about when such material was added to Earth[50]. Modelling studies suggest that water could have been added to Earth throughout its formation, with smaller bodies involved in the earlier stages and a few larger, late-accreting bodies, delivering the bulk of the water during the final stages of terrestrial formation[50]. O, Ru and Mo isotopic evidence suggests that CC material was not added any later than the Moon-forming giant impact[51,52],. On the basis of Mo isotopic evidence, it has been proposed that much of Earth's water was delivered during the Moon-forming event by an impactor of CC composition[53].

Water delivery to the inner Solar System, including Earth, appears to have taken place in three, probably overlapping, stages: (1) early nebular ingassing[32,38], (2) delivery by small asteroidal bodies, possibly in response to giant planet migration[50] and finally, (3) effects of giant protoplanets, possibly of CC composition, during the final stages of Earth's accretion[50,52]. It would have been during stage 2 that CI-related bodies, which were small and probably originated in the outer Solar System[7,54], would have made their most important contribution to the hydration of the inner Solar System, including Earth.

Methods

Oxygen isotopic analysis was undertaken at the Open University (Milton Keynes, UK) using an infrared laser-assisted fluorination system[6]. Four distinct Ryugu samples were transported to the Open University in two sealed, nitrogen-filled FFTC (facility-to-facility transport containers). One of the two FFTC contained grains from the initial Hayabusa2 touchdown collection (particle A0098,2, five grains), the other FFTC contained three sets of particles from the second, post-impactor collection: C0014,2 one particle 5.5 mg; C0068,2 one particle 0.5 mg and C0087,2 approximately ten grains, 0.8 mg. Both holders were stored at the Open University in a dedicated cabinet with a continuously purged nitrogen atmosphere.

Sample loading was undertaken in a nitrogen 'glove box' with monitored oxygen levels below 0.1%. A new Ni sample holder was fabricated for the Ryugu analysis work that consisted of just two sample wells, one for the Ryugu particles and the other for the internal obsidian standard. During analysis, the sample well containing the Ryugu material was overlain by a 1-mm-thick, 3-mm-diameter internal BaF_2 window to retain the sample during laser reaction. The flow of BrF_5 to the sample was maintained by gas mixing channels scribed into the Ni sample holder. The sample chamber configuration was also modified so that it could be removed from the fluorination line under vacuum and then opened within the nitrogen-filled glove box. The two-part chamber was made vacuum tight using a compression seal with a copper gasket and quick-release KFX clamp[6]. A 3-mm-thick BaF_2 window at the top of the chamber allowed simultaneous viewing and laser heating of samples. Following sample loading, the chamber was reclamped within the filled nitrogen glove box and then reattached to the fluorination line. Before analysis, the sample chamber was heated overnight under vacuum to a temperature of about 95 °C to remove any adsorbed moisture. Following overnight heating, the chamber was allowed to cool to room temperature and then the flexi section that had been brought up to atmosphere during the sample transfer process was purged using three aliquots of BrF_5 to remove any moisture. The oxygen isotope composition of these 'flexi' blanks were analysed using the MAT 253 micro-volume facility. These procedures ensured that the Ryugu samples were never opened to the atmosphere or contaminated with moisture from those parts of the fluorination line that had been brought up to atmosphere during the sample loading procedure.

All Ryugu samples were run in modified single-shot mode[5]. This procedure involved a single 5 min chamber blank to reduce and eliminate any residual moisture adsorbed on to the sample chamber walls. The oxygen isotope composition of this blank was analysed using the MAT 253 micro-volume facility. Following this blank analysis, the sample itself was run. Sample heating in the presence of BrF_5 was carried out using a Photon Machines Inc. 50 W infrared CO_2 laser (10.6 µm) mounted on an X-Y-Z stage. Reaction progress was monitored by means of an integrated video system. After fluorination, the released O_2 was purified by passing it through two cryogenic nitrogen traps and over a bed of heated KBr to remove any excess fluorine. The isotopic composition of the purified oxygen gas was analysed using a Thermo Fisher MAT 253 dual inlet mass spectrometer with a mass resolving power of approximately 200.

For five of the seven Ryugu samples, the amount of O_2 gas liberated during reaction was much less than 140 µg, the approximate limit for using the bellows facility on the MAT 253 mass spectrometer. In these cases, analysis was undertaken using the micro-volume. For monitoring purposes, a post-reaction blank was then run and its oxygen isotope composition was also determined. Finally, the internal obsidian standard was fluorinated and analysed. The gas liberated during the 5 min 'prereaction' blank procedure invariably had a composition close to the TFL indicating that it was predominantly composed of residual adsorbed atmospheric moisture.

The NF^+ fragment ion of NF_3^+ can cause interference with the mass 33 beam ($^{16}O^{17}O$). To eliminate this potential problem all samples were treated using a cryogenic separation procedure. This was either done in the forwards sense before analysis on the MAT 253, or as a second

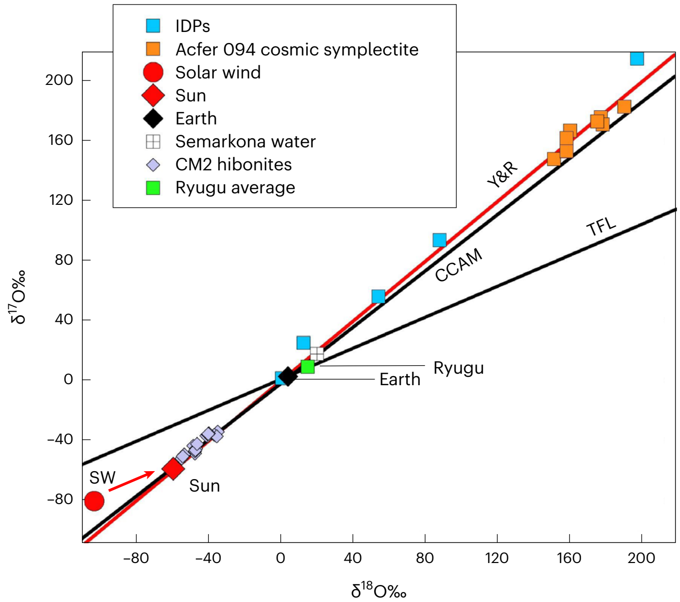

Fig. 5 | Oxygen three-isotope plot showing the average composition of Ryugu particles in relation to the principal Solar System oxygen isotope reservoirs. The composition of Sun and Solar Wind (SW)[62], refractory solids[63] plot at the bottom left of CCAM/Y&R slope 1 lines. Matrix materials from Acfer 094, known as cosmic symplectites[47,48] and some interplanetary dust particles (IDPs)[64] plot at higher $\delta^{18}O$ values along the slope 1 lines. On the scale of this diagram analyses from Ryugu, Earth and the inferred water composition from the Semarkona ordinary chondrite[65] plot close to the intersection of the TFL and slope 1 lines. Acfer 094 cosmic symplectites[47,48] analyses are thought to be representative of the oxygen isotope composition of primordial water ice. The composition of the Ryugu particles can be explained in terms of relatively extensive closed system exchange between ^{16}O-poor fluids interpreted to have a composition similar to matrix materials from Acfer 094 (refs. [47,48]) and ^{16}O-rich solids such as those commonly found in CC meteorites. This evidence appears to be at odds with the proposal that asteroid Ryugu is of cometary origin[44] (see main text for further discussion). Mixing lines are CCAM[13] and Y&R[66], which is the Young and Russell Line.

analysis with the already analysed gas pulled back onto a dedicated molecular sieve and then rerun after cryogenic separation. Cryogenic separation involved taking the gas onto the molecular sieve at liquid nitrogen temperature and then releasing it to the main molecular sieve by raising the temperature of −130 °C. Extensive tests have shown that NF^+ is retained on the first molecular sieve and that little or no fractionation results from the use of this technique.

Overall system precision in bellows mode, as defined by replicate analyses of our internal obsidian standard (n = 38), is: ±0.05‰ for $\delta^{17}O$; ±0.10‰ for $\delta^{18}O$; ±0.02‰ for $\Delta^{17}O$ (2 s.d.)[55]. Overall system precision in micro-volume mode is slightly lower than in bellows mode due to the reduced amount of gas being measured (<140 µg). Oxygen isotopic analyses are reported in standard δ notation, where $\delta^{18}O$ has been calculated as:

$$\delta^{18}\mathrm{O} = [(^{18}\mathrm{O}/^{16}\mathrm{O})_{\mathrm{sample}}/(^{18}\mathrm{O}/^{16}\mathrm{O})_{\mathrm{VSMOW}} - 1]1000\ (‰)$$

and similarly for $\delta^{17}O$ using the $^{17}O/^{16}O$ ratio. VSMOW is the international standard, Vienna Standard Mean Ocean Water, $\Delta^{17}O$, which represents the deviation from the TFL and has been calculated as: $\Delta^{17}O = \delta^{17}O - 0.52\delta^{18}O$.

The glove box used for sample loading was a Plas-Labs continuous nitrogen flow model. This achieved low moisture and oxygen levels (<0.1 wt% O_2). Weighing the samples during loading proved problematic due to pressure fluctuations compromising the balance. A normalization procedure has therefore been adopted using the 0.5 mg value for particle C0068 obtained during initial sample preparation at the SPring-8 synchrotron facility before dispatch to the UK. C0068 was measured in its entirety as single measurement and gave a 17% yield (Table 1). A value of 17% is reasonable in view of the average 12.1% achieved on the measured CYs (Y-82162,82) and 25.2% measured on the CIs (Orgueil) (Table 1).

Blank correction procedure

Blank correction data for all the samples analysed in this study are given in the Supplementary Information. The relatively small size of the Ryugu samples available for oxygen isotope analysis meant that it was necessary to apply a blank correction to all the samples analysed in this study[56]:

$$\delta_s = (n_T\delta_T - n_b\delta_b)/(n_T - n_b)$$

where n_T = total amount measured and is equal to $n_s + n_b$

n_b = amount of blank

n_s = amount of sample

δ_T = delta total amount

δ_b = delta blank

δ_s = delta sample

The values of n_b and δ_b were determined by loading a Ryugu tray with only an obsidian standard present. The 'flexi' blanks were carried out as normal. A 5-min sample chamber blank was then run and the 4 µg of O_2 that was evolved during this procedure was run on the MAT 253 micro-volume. The results obtained were: $\delta^{17}O$ = −5.15‰; $\delta^{18}O$ = −9.95‰ and $\Delta^{17}O$ = 0.02‰. As the amount of time that this blank was run for was greater than our usual lasering time of about 2 min, the blank correction applied was reduced to 2.4 µg of O_2. Details of the blank correction applied to each analysis are given in the Supplementary Information.

$\delta^{18}O$ variation in unhomogenized primitive CCs

In most cases, when determining the bulk oxygen isotope composition of a meteorite a relatively large chip of typically between 100 to 200 mg of the sample is crushed and homogenized. Aliquots of about 2 mg are then taken from this homogenized powder and analysed by laser fluorination. The aim is to determine a representative bulk composition for the meteorite. However, for some studies much smaller mg-sized fractions have been removed from primitive meteorites and analysed by laser fluorination. The range in $\delta^{18}O$ obtained from these small subsamples often matches or exceeds the range observed in this study. Thus, in the case of NWA 7891 $\delta^{18}O$ measurements ranged from: −15.42 to −2.39‰; NWA 8781: −6.09 to 1.22‰; NWA 11961: −2.48 to 6.43‰; Telakoast 001 −3.15 to 2.15‰ and Tarda: 15.94 to 21.97‰ (Supplementary Information). In keeping with the results from the present investigation, the results from these meteorite studies indicat that where a sample displays a substantial level of inherent isotopic heterogeneity between mineral phases, mg-sized samples, without previous homogenization of a larger fraction of material, will show a large degree of $\delta^{18}O$ heterogeneity.

$\delta^{18}O$ variation related to modal variations in Ryugu particles

As discussed in the main text, Ryugu particles analysed in this study show a significant range in $\delta^{18}O$ values (11.46 to 19.30‰). Calculations show that this variation can be fully explained in terms of the heterogeneous distribution of the main oxygen-bearing phases, phyllosilicate, magnetite and dolomite (Supplementary Information).

Calculations undertaken to model the influence of terrestrial contamination on the $\Delta^{17}O$ composition of CI chondrites

As discussed in the main text, the small difference between the $\Delta^{17}O$ compositions of the Ryugu particles and CI chondrites is most probably related to terrestrial contamination of the CI meteorites. Two distinct approaches have been used to model the contamination of the CIsl (Supplementary Information). Both sets of calculations give essentially identical results.

Data availability

All of the data relevant to this publication are available in Table 1 and in the Supplementary Information. All images and data used in this study are available at the JAXA Data Archives and Transmission System (DARTS). Data for Hayabusa2 samples and other data from the mission are available at the DARTS archive at https://www.darts.isas.jaxa.jp/curation/hayabusa2 and https://www.darts.isas.jaxa.jp/planet/project/hayabusa2/, respectively.

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Author contributions
The initial draft of the manuscript and all revisions were written by R.C.G. Sample handling, loading and analysis was undertaken by R.C.G., R.F. and J.A.M. The blank correction procedure was developed by I.A.F., R.F. and R.C.G. M.I., A.Y. and other members of the Kochi team undertook sample selection, curation and loading of samples into the sealed FFTC containers. All authors contributed to data interpretation and editing of the initial manuscript.

Competing interests
The authors declare no competing interests.

Additional information

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