




The water content of CM carbonaceous chondrite falls and finds, and their susceptibility to terrestrial contamination

Martin R. LEE ^{1*}, Lydia J. HALLIS ¹, Luke DALY ^{1,2,3}, and Adrian J. BOYCE⁴

¹School of Geographical & Earth Sciences, University of Glasgow, Glasgow, UK

²Australian Centre for Microscopy and Microanalysis, University of Sydney, Sydney, New South Wales, Australia

³Department of Materials, University of Oxford, Oxford, UK

⁴Scottish Universities Environmental Research Centre, Glasgow, UK

*Correspondence

Martin R. Lee, School of Geographical & Earth Sciences, University of Glasgow, Glasgow G12 8QQ, UK.

Email: martin.lee@glasgow.ac.uk

(Received 07 June 2023; revision accepted 26 October 2023)

Abstract—CM carbonaceous chondrites can be used to constrain the abundance and H isotopic composition of water and OH in C-complex asteroids. Previous measurements of the water/OH content of the CMs are at the higher end of the compositional range of asteroids as determined by remote sensing. One possible explanation is that the indigenous water/OH content of meteorites has been overestimated due to contamination during their time on Earth. Here we have sought to better understand the magnitude and rate of terrestrial contamination through quantifying the concentration and H isotopic composition of telluric and indigenous water in CM falls by stepwise pyrolysis. These measurements have been integrated with published pyrolysis data from CM falls and finds. Once exposed to Earth's atmosphere CM falls are contaminated rapidly, with some acquiring weight percent concentrations of water within days. The amount of water added does not progressively increase with time because CM falls have a similar range of adsorbed water contents to finds. Instead, the petrologic types of CMs strongly influence the amount of terrestrial water that they can acquire. This relationship is probably controlled by mineralogical and/or petrophysical properties of the meteorites that affect their hygroscopicity. Irrespective of the quantity of water that a sample adsorbs or its terrestrial age, there is minimal exchange of H in indigenous phyllosilicates with the terrestrial environment. The falls and finds discussed here contain 1.9–10.5 wt% indigenous water (average 7.0 wt%) that is consistent with recent measurements of C-complex asteroids including Benu.

INTRODUCTION

Meteorites derived from C-complex asteroids are likely to have been an important source of water and other volatiles for the terrestrial planets early in their history (Alexander, 2017; Alexander et al., 2012, 2018; Marty, 2012). A good understanding of the chemical and isotopic composition of these bodies is therefore crucial for modeling planetary evolution and habitability. The abundance of water and hydroxyls (OH⁻) in C-complex asteroids can be determined by infrared spectroscopy using ground-based telescopes and spacecraft, with a

focus on the ~3 μm absorption band (Praet, Barucci, Clark, et al., 2021; Praet, Barucci, Hasselmann, et al., 2021; Rivkin et al., 2003, 2015). For example, measurements of the B-type asteroid Benu by the OSIRIS-REx spacecraft found that its surface has a mean H content of 0.71 ± 0.16 wt% (equivalent to 6.35 ± 1.43 wt% water; Praet, Barucci, Clark, et al., 2021). Using data from main belt C-complex asteroids obtained by the Akari telescope, Beck et al. (2021) estimated that they host ~4.5 ± 4.0 wt% water. The abundance of asteroidal water/OH, and its H isotopic composition, can also be estimated using carbonaceous chondrite (CC)

meteorites (e.g., Boato, 1954; Eiler & Kitchen, 2004; Kerridge, 1985; Kolodny et al., 1980) and samples of C-complex asteroids that have been returned by spacecraft (e.g., the Hayabusa2 mission to the C-type asteroid Ryugu; Ito et al., 2022). Comparisons of asteroid water contents inferred from meteorites and measured by remote sensing show that CCs typically have higher concentrations of water than their putative parent bodies (e.g., Beck et al., 2021; Hiroi et al., 1996). Explanations for this discrepancy include: (i) the asteroids that have been measured spectroscopically have been broken up to expose their heated interiors, but those same parts have not been sampled by CC meteorites (Hiroi et al., 1996); (ii) the outer surfaces of asteroids have been partially dehydroxylated by space weathering (e.g., Beck et al., 2021); and (iii) the abundance of indigenous water in CC meteorites is overestimated owing to terrestrial contamination (e.g., Vacher et al., 2020). Here we test the third explanation using stepwise pyrolysis data from Mighei-like (CM) meteorites, which are the largest and petrologically most diverse group of hydrous CCs in our collections.

The CM group is spectroscopically linked to carbonaceous asteroids, and in particular the Ch class (e.g., Rivkin et al., 2015; Takir et al., 2013, 2019). Most of the indigenous “water” in these meteorites is contained within phyllosilicates (e.g., serpentine $[\text{Mg,Fe}]_3\text{Si}_2\text{O}_5[\text{OH}]_4$), sulfides (tochilinite: $2[\text{Fe,Ni,Cu}]_{1-x}\text{S}\cdot n[\text{Fe,Mg}][\text{OH}]_2$), and organic matter (Alexander et al., 2013). Hereafter these phases are collectively referred to as the “indigenous” hosts of water. The phyllosilicates and tochilinite were formed by aqueous alteration of silicates, sulfides, metal, and amorphous materials following melting of co-accreted water-rich ice (e.g., Bunch & Chang, 1980; DuFresne & Anders, 1962). The ice was D-poor ($\delta D -427 \pm 45\text{‰}$; Alexander et al., 2018) and the phyllosilicates are correspondingly isotopically light ($\delta D -200$ to -450‰ ; Alexander et al., 2012) ($\delta D = (D/H_{\text{sample}}/D/H_{\text{standard}}(\text{VSMOW}) - 1) \times 1000$, where VSMOW denotes Vienna Standard Mean Ocean Water with a D/H ratio of 155.76×10^{-6}). The abundance of hydrous minerals differs between meteorites depending on their degree of aqueous alteration, which is expressed by their petrologic subtype/type. Unaltered and nominally anhydrous CMs are classified as CM3.0/3.0, and completely altered meteorites as CM2.0/1.0 (Alexander et al., 2013; Howard et al., 2015; Rubin et al., 2007). Insoluble organic matter (IOM) in the CMs can produce water by the oxidation of H as it breaks down during laboratory analysis. At the time of its accretion, the organic matter was D-rich (up to $\delta D \sim 3500\text{‰}$; Alexander et al., 2007, 2012) but could have been subsequently isotopically modified by reaction with D-poor parent body water (Alexander et al., 2012, 2013). In addition to these indigenous sources, CM meteorites can evolve water from

one or more terrestrial carriers: (i) adsorbed molecular water; (ii) water- and OH-bearing minerals (e.g., ferrihydrite, gypsum); and (iii) organic compounds (e.g., Boato, 1954; Eiler & Kitchen, 2004; Lee, Cohen, et al., 2021; Robert & Epstein, 1982; Vacher et al., 2020).

Here we have used stepwise pyrolysis to quantify the abundance and H isotopic composition of indigenous and terrestrial water in bulk CM samples. These results have also been integrated with pyrolysis data from the literature. To help understand the rate of terrestrial contamination we have analyzed meteorites that have been in contact with the Earth’s environment for precisely known lengths of time (i.e., falls). The sample set chosen spans 180 years from Cold Bokkeveld (fell 13 October 1838) to Winchcombe (fell 28 February 2021). Data from the falls are also compared with published stepwise pyrolysis results from Antarctic CM finds to test the assumption that the latter are more highly contaminated. Outcomes of this work can help to inform the interpretation of spectroscopic data from C-complex asteroids, and protocols for determining the water content of CC lithologies including fresh meteorite falls and returned samples.

MATERIALS AND METHODS

The quantity and H isotopic composition of water evolved from the CM falls Aguas Zarcas, Kolang, Murchison, and Shidian was measured by stepwise pyrolysis. Results from these four meteorites are described together with previously reported data from the CM falls Cold Bokkeveld (Lee, Cohen, et al., 2021) and three samples of Winchcombe (King et al., 2022) that were obtained using the same technique and in the same laboratory. The dates and locations of fall of these meteorites, and their petrologic classifications, are listed in Table 1 and Table S1, and their curatorial backgrounds are described in Table S2. Stepwise pyrolysis results from 12 Antarctic CM finds are included in the Results and Discussion sections to compare with the falls’ data. Those finds were analyzed in the same laboratory as the falls (Lee, Cohen, et al., 2021), and results are listed in Table S3. The table also identifies those meteorites that have undergone post-hydration heating (which lowers their indigenous water contents) and were recorded to host evaporites (a terrestrial weathering product).

Stepwise pyrolysis analyses were undertaken at the Scottish Universities Environmental Research Centre (SUERC). The methodology was described in Lee, Cohen, et al. (2021) and is summarized below. A chip of each sample, a few tens of mg in size, was put into a degassed platinum crucible in an all-glass vacuum line, pumped down overnight, then incrementally heated using

TABLE 1. Meteorites analyzed by stepwise pyrolysis.

| Meteorite | Fall date (dd/mm/yyyy), location | Petrologic classifications | | | |
|-----------------------------|----------------------------------|----------------------------|-------------------|----------------------|-------------------|
| | | Subtype ^a | Type ^b | Type ^c | Type ^d |
| Winchcombe ^e | 28/02/2021, England | CM2.0–2.6 ^e | — | 1.1–1.2 ^e | 1.3–1.7 |
| Kolang | 01/08/2020, Indonesia | CM2.2 ^f | — | 1.3 ^f | 1.4 |
| Aguas Zarcas ^g | 31/05/2019, Costa Rica | CM2.2–2.8 ^h | — | — | 1.6 |
| Shidian | 21/11/2017, China | CM2.2 ⁱ | — | — | 1.6 |
| Murchison | 28/09/1969, Australia | CM2.5 | 1.6 | 1.5 | 1.7 |
| Cold Bokkeveld ^j | 13/10/1838, South Africa | CM2.2 | 1.3 | 1.4 | 1.7 |

— denotes no classification is available.

^aClassification scheme of Rubin et al. (2007).

^bClassification scheme of Alexander et al. (2013).

^cClassification scheme of Howard et al. (2015).

^dClassified in the present study using the protocol of Alexander et al. (2013).

^eStepwise pyrolysis data and classifications published in King et al. (2022).

^fKing et al. (2021).

^gA post-rain sample.

^hKerraouch et al. (2021).

ⁱFan et al. (2020).

^jStepwise pyrolysis data published in Lee, Cohen, et al. (2021).

a temperature-controlled resistance furnace (25–700°C) and temperature-controlled induction furnace (>700°C). Seven steps were used: 100, 200, 300, 400, 500, 700, 1100°C ($\pm 5^\circ\text{C}$), and samples were held at each temperature for at least 30 min. The released gases were first stored in a cold trap at -196°C using a liquid N_2 bath. It was replaced by a mixture of dry ice and acetone at -78°C , trapping water but releasing other gases. The yield of the gas not trapped at -78°C was measured using an Edwards PR10-C Pirani vacuum pressure gauge. The dry ice and acetone bath was removed from the cold trap, gently heated, and the water released was reduced to H_2 by exposure to Cr powder at 850°C (Donnelly et al., 2001). For each step the H yield was measured using the Pirani vacuum gauge, and H_2 was collected using a mercury Toepler pump into a small borosilicate vessel with Teflon-sealed high-vacuum valves. The vessels were taken immediately for measurement of H isotopes using a VGI Optima mass spectrometer. In-run repeat analyses of water standards (international standards GISP, VSMOW and internal standard LT-STD) gave a reproducibility of better than $\pm 2\%$ for δD . In those cases where the yield from a step was too small to measure (i.e., $<0.15 \mu\text{mol mg}^{-1} \text{H}_2$) it was added to the subsequent step. Pyrolysis results for all of the fall samples are listed in Table S4.

The carriers of water can be constrained using the temperature at which it is evolved and its H isotopic composition. The yield from the 100 and 200°C steps is typically inferred to be from molecular water that has been adsorbed from the Earth's atmosphere (e.g., Boato, 1954; Kolodny et al., 1980; Robert & Epstein, 1982; Vacher et al., 2020); however, oxidation of volatile

organic matter and the dehydration/dehydroxylation of terrestrially formed minerals may also contribute. The organic matter could be indigenous or terrestrial, and with regard to the latter there is evidence for rapid contamination of CM falls by atmospheric aerosols (Cronin & Pizzarello, 1990). The most likely mineral source of low temperature water is gypsum, which dehydrates at $95\text{--}170^\circ\text{C}$ (Strydom et al., 1995) and could be indigenous or terrestrial. Irrespective of the nature of its carriers, a terrestrial origin for most of the $<200^\circ\text{C}$ yield is consistent with oxygen isotopic analyses of water from a bulk sample of Murchison (CM2) (Baker et al., 2002). They found that the water evolved at $<200^\circ\text{C}$ has a terrestrial $\Delta^{17}\text{O}$ signature (i.e., $\sim 0\%$) whereas that produced at higher temperatures has a negative, and so non-terrestrial, $\Delta^{17}\text{O}$. Most of the water that is evolved from CM chondrites above 200°C is assumed to originate from the dehydration/dehydroxylation of indigenous hydrous minerals (i.e., serpentine, tochilinite) and the oxidation of H from indigenous organic matter (e.g., Kolodny et al., 1980; Lee, Cohen, et al., 2021; Robert & Epstein, 1982; Vacher et al., 2020). There may also be a small contribution from terrestrial Fe-oxyhydroxides (e.g., goethite), which dehydroxylate at $>200^\circ\text{C}$ (Garenne et al., 2014; Gualtieri & Venturilli, 1999).

RESULTS

Stepwise Pyrolysis Profiles of the CM Falls

Results from the fall samples are plotted in Figure 1 and listed in Tables S1 and S4. The lowest water yields from all samples were in the 100°C step (below detection

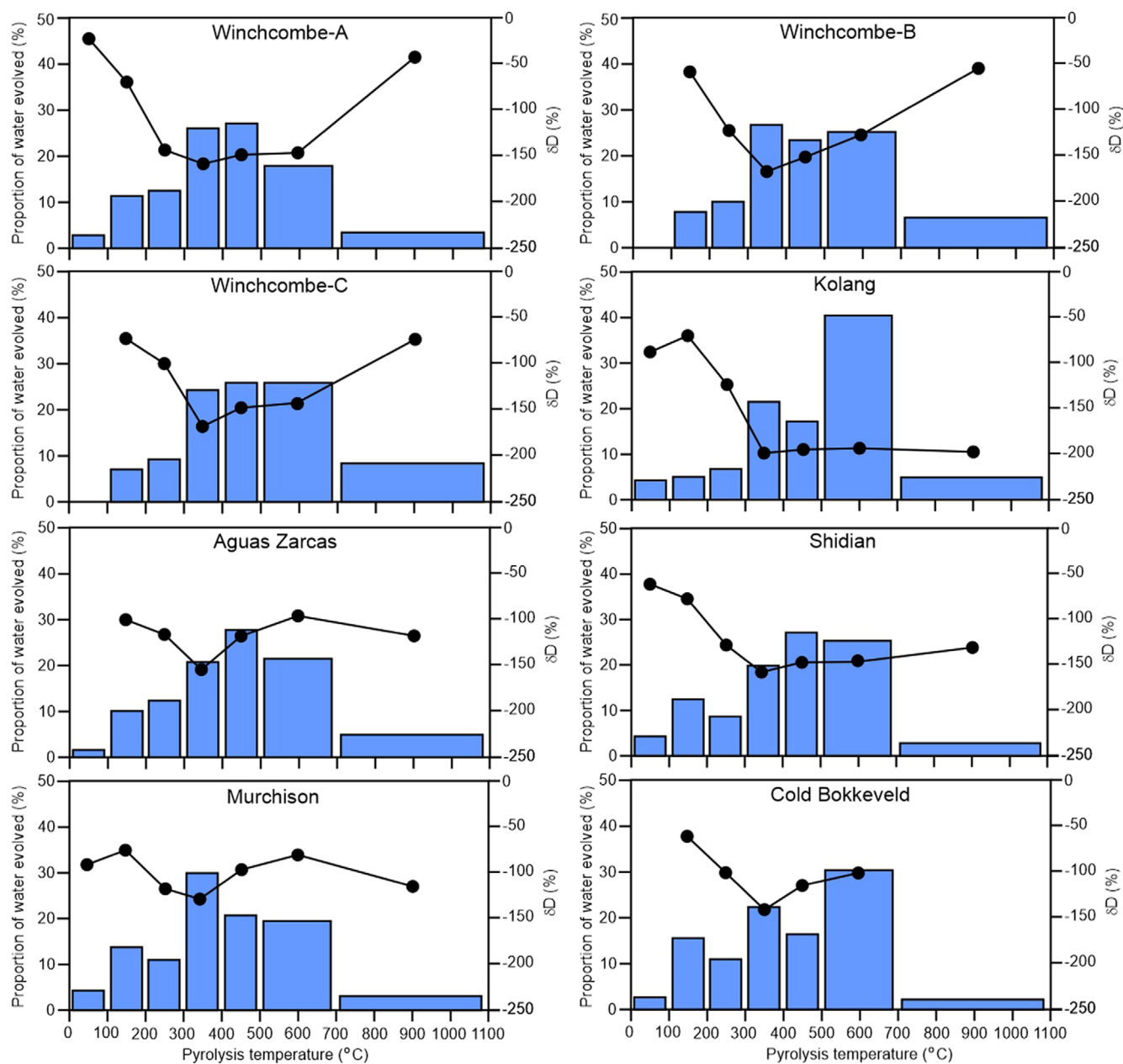


FIGURE 1. Stepwise pyrolysis profiles for the CM falls. Water yield is plotted as the blue columns and its H isotopic composition as black circles. The raw data are in Tables S1 and S4. Analyses of Cold Bokkeveld and the three Winchcombe samples are in Lee, Cohen, et al. (2021) and King et al. (2022), respectively. (Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com))

to 4.3% of total) and 1100°C step (2.0%–8.4% of total) whereas the 400, 500, or 700°C steps gave the highest yields. Two samples have distinctive profiles: Kolang evolved 40% of its water in the 700°C step, and Murchison yielded 30% in the 400°C step. The highest δD values ($> -100\text{‰}$) were typically in the 100 or 200°C step, with subsequent steps falling to between -100 and -200‰ . The three Winchcombe samples differ from the other falls in that their 1100°C step has a relatively high δD of -74 to -43‰ .

In order to facilitate description and interpretation of these results, the data are divided into five fractions (Table S1): (i) Water evolved in the 100°C step (“weakly bound-1”); (ii) water evolved in the 200°C step (“weakly bound-2”); (iii) water evolved in the 100 plus 200°C steps (“weakly bound-total”); (iv) water evolved in the 300–1100°C steps (“indigenous”); and (v) all water evolved over the whole temperature range (i.e., weakly bound-total + indigenous; “Bulk”). Stepwise pyrolysis data from the Antarctic CM finds are also expressed as the five fractions in Table S3.

Weakly Bound Fractions

The falls' weakly bound-1 fraction constitutes up to 0.50 wt% water (Table S1). Although yield does not correlate with the time between when the meteorite fell and when it was analyzed (hereafter "elapsed time"), two samples of the most recent fall (Winchcombe B and C) evolved insufficient water to measure (note that they are subsamples of the same chip; Table S2). The falls' weakly bound-2 fraction comprises 0.60–1.71 wt% water and again does not relate to elapsed time, although the highest yields were from Murchison and Cold Bokkeveld (Figure 2a). Also plotted in Figure 2a is the weakly bound-2 fraction of the finds. They show no systematic difference to the falls, yet the highest yields were from two of the Antarctic CMs that were recorded to contain evaporites (PCA 91084 and SCO 06043). The average H isotopic composition of the weakly bound-total fraction for the falls and Antarctic finds is $\delta D -71 \pm 10\text{‰}$ and $-82 \pm 10\text{‰}$, respectively. The falls show some relationship between δD and elapsed time in that values decrease overall between the three Winchcombe samples and Murchison, although increase for Cold Bokkeveld (Table S1, Figure 2b).

Indigenous Fraction

The falls evolved 8.98–11.44 wt% water from indigenous sources (i.e., minerals and organic matter) with a weighted δD of -191 to -108‰ . Excluding those finds that have undergone post-hydration heating, most of the Antarctic CMs evolved less indigenous water than the falls (4.61–9.29 wt%) and have a wider range of H isotopic composition ($\delta D -236$ to 145‰). The four post-hydration heated finds are relatively water-poor (2.67–7.90 wt%; Tables S1 and S3).

Petrologic Type of the Falls and Finds

Differences in indigenous water yields between the falls and Antarctic finds are most likely due to contrasts in phyllosilicate abundance, which is determined by their degree of parent body aqueous alteration and can be further modified by post-hydration heating. However, this suggestion cannot be readily confirmed because none of the published petrologic classifications include all of the falls and finds discussed here. In addition, several of the falls are breccias containing clasts with different degrees of aqueous alteration (i.e., Aguas Zarcas, Kerraouch et al., 2021; Cold Bokkeveld, Lentfort et al., 2021; Winchcombe, King et al., 2022; Suttle et al., 2022). Thus, milligram-size samples of the same meteorite may differ significantly in phyllosilicate abundance. Owing to these limitations, the petrologic type of all of the samples was calculated via the scheme of Alexander et al. (2013)

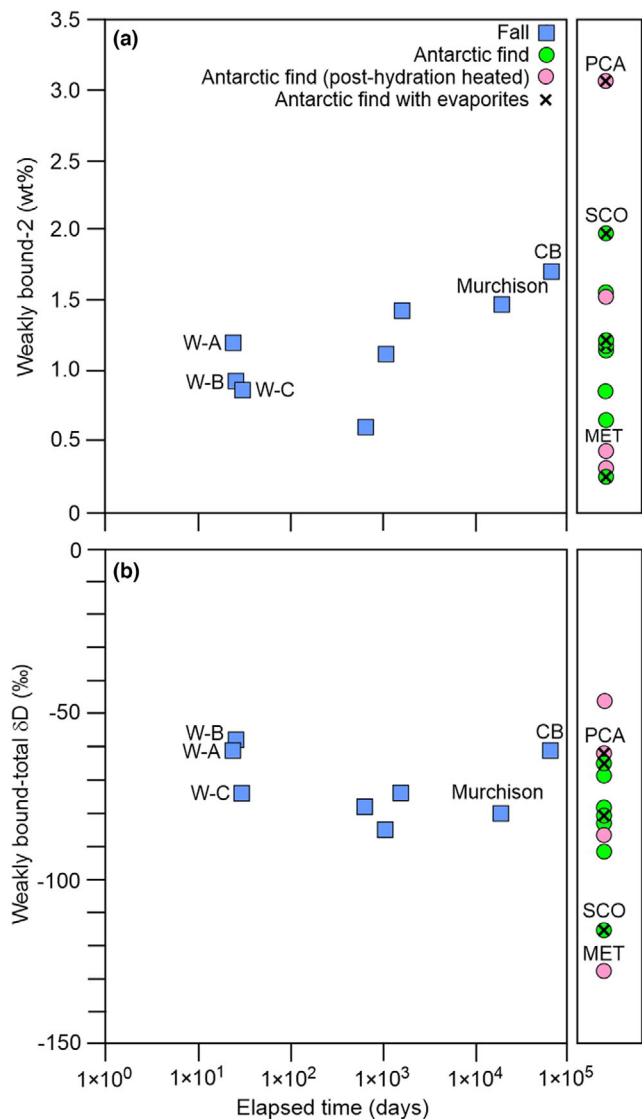


FIGURE 2. (a) The concentration of the weakly bound-2 fraction plotted against elapsed time for the eight fall samples. To the right of the graph is the same fraction for the 12 Antarctic finds. The finds span a wider range of water yields, and the two samples that evolved most water are evaporite bearing. (b) The H isotopic composition of the weakly bound-total fraction of the eight fall samples plotted against elapsed time. To the right of the graph is the same fraction for the 12 Antarctic finds. The falls show a decrease overall in δD between the three Winchcombe samples and Murchison. The finds span a wider range than the falls, and two of them have a significantly lower δD than any of the other samples. Labeled meteorites are: Cold Bokkeveld (CB), MET 01075-a (MET), SCO 06043 (SCO), PCA 91084 (PCA), and Winchcombe (W-A, W-B, W-C). The legend is the same for both graphs. (Color figure can be viewed at wileyonlinelibrary.com)

and using their amount of indigenous water as listed in Tables S1 and S3.

Prior to calculating petrologic type, it was necessary to subtract water derived from the oxidation of IOM-derived

H from each sample's indigenous fraction. By making such a correction the classifications will better reflect each sample's degree of aqueous alteration (i.e., petrologic type will be calculated using only water that was sourced from minerals that formed during parent body water/rock interaction, mainly phyllosilicates and tochilinite). These IOM-adjusted indigenous water contents (hereafter "corrected indigenous") will also be more directly comparable to the spectroscopically measured water/OH contents of C-complex asteroids. The IOM-sourced water was subtracted by assuming that it has a H/C ratio of 0.055 by weight (Alexander et al., 2012, 2013) and taking bulk C contents from the literature (Table S5). As some of the C in each sample is hosted by carbonate minerals, their contribution to bulk C values was removed using data on the amount of carbonate-hosted C in each meteorite (Alexander et al., 2015). The newly calculated petrologic types of the falls and finds are listed in Table 1 and Table S5. It should be noted that there will be a degree of uncertainty in these petrologic type classifications of $\sim\pm 0.1$ – 0.2 because bulk C and carbonate C data were obtained from different samples to those used in the pyrolysis experiments.

The falls, and those finds that have not undergone post-hydration heating, show a good correlation between newly calculated petrologic type and the H isotopic composition of their indigenous water (Figure 3). This correlation means that samples of a lower petrologic type have a greater proportion of isotopically light phyllosilicate relative to isotopically heavy IOM. The petrologic types of Cold Bokkeveld (1.7) and Murchison (1.7) may seem anomalous given that other classifications find Cold Bokkeveld to be the more highly aqueously altered of the two (Table 1). However, when expressed to two decimal places Cold Bokkeveld (type 1.66) is indeed more altered than Murchison (type 1.72), and we are confident in these classifications because both samples plot very close to the regression line in Figure 3. It may be that the chips of Cold Bokkeveld and/or Murchison that were used for stepwise pyrolysis measurements were atypical in their degree of alteration, for example due to the presence of clasts of a different petrologic type. The post-hydration heated finds plot to the right of the regression line in Figure 3 because they have a high petrologic type for their δD (i.e., their petrologic type is over estimated owing to dehydroxylation during heating after aqueous alteration).

Correlation Between the Weakly Bound and Indigenous Fractions

There is a good correlation between the total amounts of weakly bound water and indigenous water that the falls evolve (Figure 4a). Those with more indigenous water yield less of the weakly bound fractions,

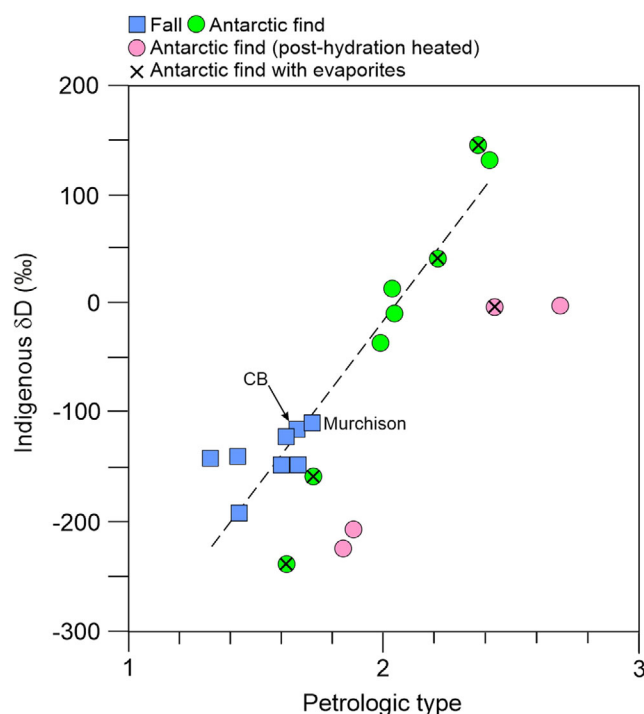


FIGURE 3. The newly calculated petrologic type of the eight fall and 12 find samples plotted against the H isotopic composition of their indigenous water. The dashed black regression line was plotted using the eight falls, and the eight finds that have not undergone post-hydration heating (R^2 0.85). Petrologic types and the data used to calculate them are listed in Table S5. Note that petrologic type expressed to two decimal places was used to plot the graph. CB denotes Cold Bokkeveld. (Color figure can be viewed at wileyonlinelibrary.com)

and this relationship is highlighted by the three Winchcombe samples that plot close to the regression line in Figure 4a. Most of the finds lie parallel to the falls' regression line but offset due to lower indigenous water contents (Figure 4a). Two Antarctic CMs plot away from the find's regression line as they have an anomalously high weakly bound fraction relative to their indigenous fraction (PCA 91084 and SCO 06043). There is no correlation between the δD of the indigenous and weakly bound-total water of the falls, which is consistent with the two fractions coming from isotopically distinct sources (e.g., adsorbed and phyllosilicate-bound, respectively). However, two Antarctic finds with the lowest δD of their weakly bound-total fraction also have a low δD of their indigenous water (Figure 4b).

DISCUSSION

The goal of this study is to understand the rate and magnitude of contamination of CM CCs with terrestrial "water", be it from adsorption of molecular water, introduction of organic matter, or the growth of water/OH-bearing minerals. We start by evaluating the nature

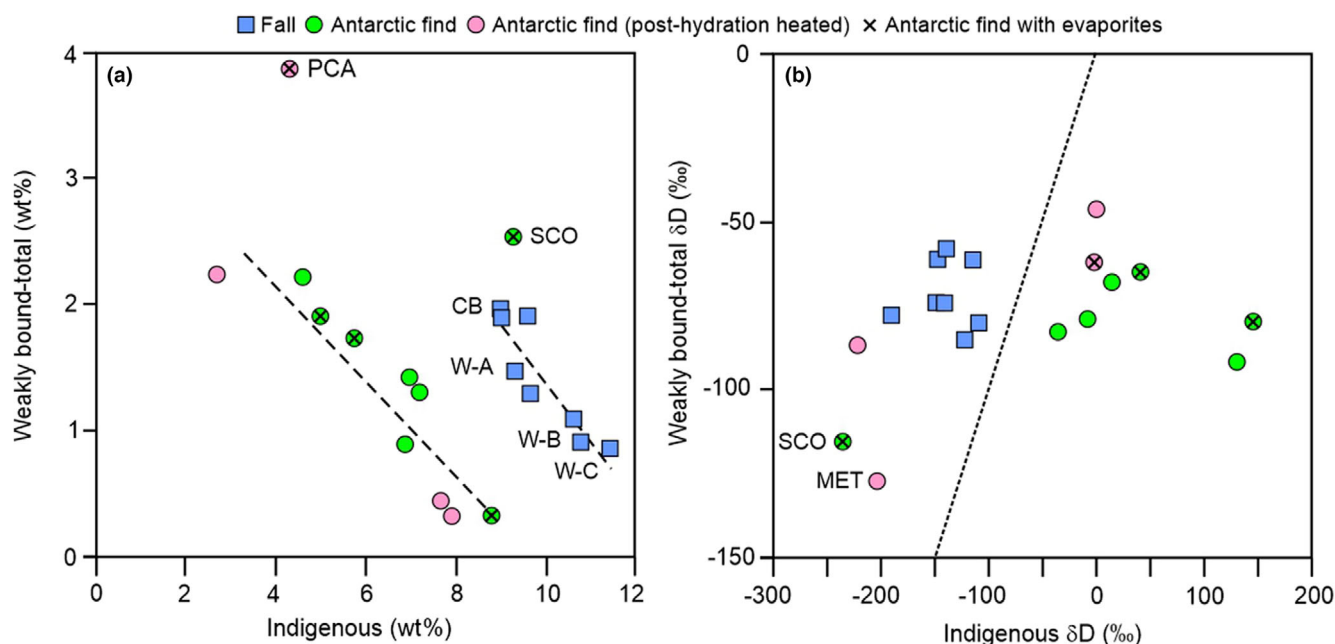


FIGURE 4. (a) The amount of water in the weakly bound-total fraction of the CM falls and finds plotted against their indigenous fraction. The dashed black regression line for the falls has a R^2 of 0.81 and for the finds (excluding PCA 91084 and SCO 06043) has a R^2 of 0.83. (b) The H isotopic composition of the weakly bound-total fraction plotted against that of the indigenous fraction. Most samples have a weakly bound-total δD of -50 to -100 ‰, but it is lower in MET 01075-a and SCO 06043, and the indigenous fraction of those two samples also has a low δD . The yield from ALH 83100 was insufficient for the H isotopic composition of its weakly bound fraction to be measured. The dashed line is a 1:1 line between the weakly bound-total and indigenous fractions. Labeled samples are: Cold Bokkeveld (CB), MET 01075-a (MET), PCA 91084 (PCA), SCO 06043 (SCO), and Winchcombe (W-A, W-B, W-C). (Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com/doi/10.1111/jmap.14099))

and origin of the hosts of the weakly bound fraction. We then assess the rate at which CMs are contaminated, and how meteorite properties control their interaction with the terrestrial environment. Finally, by using this new understanding of telluric contamination we define a range and an average indigenous water content for the CMs and compare it to spectroscopic data from C-complex asteroids.

Rate of Terrestrial Contamination of CM Falls

As the three Winchcombe samples were analyzed within a month of their fall, they can help to constrain the rate of terrestrial contamination. Their weakly bound-1 fraction had little water, with Winchcombe A yielding the most (0.29 wt%), whereas all three samples had a larger bound-2 fraction (0.86–1.20 wt%). At 0.86–1.49 wt%, the weakly bound-total fraction of the three samples constitutes 7%–14% of their bulk water yield.

The weakly bound-1 fraction comprises adsorbed terrestrial water. Its relatively low abundance may be due to partial dehydration while the samples were held under vacuum prior to the pyrolysis (the three Winchcombe samples were under vacuum for between 2 and 6 days; Table S2). The source of the weakly bound-2 fraction is

also likely to be adsorbed molecular water, although hydrous sulfates may have made a minor contribution because very small quantities of terrestrially formed gypsum and bassanite ($\text{CaSO}_4 \cdot 0.5\text{H}_2\text{O}$) were described by Jenkins et al. (2023) from a different piece of Winchcombe that was recovered a week after its fall. Our conclusions from the stepwise pyrolysis data are supported by thermogravimetric analysis (TGA) results from a sample of Winchcombe that was analyzed 5 days after it fell showing a mass loss between 20 and 200°C (inferred to be mainly due to water) of 3.0 wt% (King et al., 2022). The total amounts of adsorbed water measured by pyrolysis (0.86–1.49 wt%) and TGA (3.0 wt%) differ significantly, which at least in part reflects contrasts in sample preparation (i.e., the TGA work used a powder that would be more prone to adsorbing water from the atmosphere than a chip), but results from the two techniques agree that Winchcombe was rapidly contaminated. Such fast acquisition of terrestrial water is also consistent with findings of Boato (1954) that the H isotopic composition of water adsorbed by a suite of meteorites including several CM falls differed between the summer and winter months reflecting seasonal variations in the H isotopic composition of local precipitation. It is quite possible that rock and mineral

samples start to be contaminated on the order of minutes because Clark et al. (1993) suggested that they can adsorb atmospheric water while being transferred between instruments in a laboratory. This point also highlights the potential effects of the nature of sample storage on water adsorption; for example, a sample that is kept under stable conditions may be contaminated to a different extent relative to one that is repeatedly moved between different curatorial settings.

Excluding Winchcombe samples, the falls had a weakly bound-2 fraction of 0.60–1.71 wt%. The largest yield was from Cold Bokkeveld (Figure 2a), which most likely represents dehydration of gypsum. This hydrous sulfate has been described from Cold Bokkeveld by Fuchs et al. (1973), Lee (1993), Airieau et al. (2005), and Howard et al. (2015), and the latter study measured 0.8 vol% by X-ray diffraction. Cold Bokkeveld evolved 1.71 wt% water in the 200°C step, which is 0.24 and 0.28 wt% greater than the two next most water-rich falls (Shidian and Murchison). Out of that difference 0.17 wt% can be accounted for by dehydration of 0.8 vol% gypsum. Although its provenance is debated (Lee, Daly, et al., 2021), a terrestrial origin for Cold Bokkeveld's gypsum is consistent with it being the terrestrially oldest CM fall. As gypsum has also been described from Murchison (Airieau et al., 2005; Fuchs et al., 1973; Garenne et al., 2014) the sulfate may also be responsible for some of that meteorite's weakly bound fraction.

Terrestrial Contamination of Antarctic Finds

Antarctic CMs might be expected to be more contaminated than the falls given their much longer terrestrial residence during which time they have been encased in ice, subaerially exposed, and sometimes in direct contact with liquid water (Harvey, 2003). However, the average amount of water in the weakly bound-total fraction of the 12 finds (1.60 wt%) is close to that of the eight fall samples (1.44 wt%) and would be lower than the falls (i.e., 1.40 wt%) if it were not for PCA 91084 whose weakly bound-total fraction is anomalously large (3.85 wt%). If the unexpectedly low weakly bound-total fraction of the finds was due to partial loss of adsorbed water during curation, a correlation may be expected between the size of their weakly bound fractions and date of meteorite recovery (i.e., the length of time that samples had been stored in the curation facility). However, the finds show no such relationship (Table S3).

Although the finds do not necessarily have more weakly bound water than the falls, there is some evidence for Antarctic-specific contamination. The weakly bound-total fraction of MET 01075-a and SCO 06043 has an anomalously low δD (Figure 4b), which could be due to an adsorbed component or weathering product

containing isotopically light H derived from Antarctic meltwater (δD –100 to –400‰ depending on location; Masson-Delmotte et al., 2008). The indigenous fraction of those two finds also has a low δD , which might be explained by Antarctic meltwater contributing to the formation of Fe-oxyhydroxides and/or clays or substituting for water in indigenous hydrous minerals. Nonetheless, most Antarctic finds have a higher indigenous δD than the falls showing that their phyllosilicates have retained their original (parent body) H (Figures 3 and 4a).

The high yield of weakly bound water from PCA 91084 may be attributed to terrestrial evaporites, which were recorded on interior surfaces of the meteorite during its initial characterization (Score & Lindstrom, 1993). The mineralogy of these evaporites was not determined, but will likely include one or more of nesquehonite, hydromagnesite, and gypsum, which are common weathering products of Antarctic CMs (Losiak & Velbel, 2011; Velbel, 1988). Labidi et al. (2017) found that PCA 91084 contains 1.3 wt% S hosted by sulfate, which if solely in gypsum would correspond to 7 wt% of the mineral. With regard to the four other evaporite-bearing Antarctic finds, SCO 06043 also has a high weakly bound-2 fraction (and with a low δD) whereas the three other samples do not (Figure 2a). This lack of a consistent correlation between high concentrations of weakly bound water and the recorded presence of evaporites could be explained by the patchy distribution of these weathering products (i.e., they were absent from some of the tens of mg size subsamples that were analyzed by stepwise pyrolysis).

Evidence for contamination of Antarctic CMs by terrestrial H or O has been described by Fujiya (2018). The study compared CM falls with finds and found that most of the falls show a good correlation between their bulk O isotopic composition and the amount of H in H₂O/OH (as measured by Alexander et al., 2012 and approximately equivalent to the indigenous fraction). The Antarctic finds have a similar relationship but offset from the falls' correlation by a ~30% higher H or ~3‰ lower $\delta^{18}\text{O}$. This offset was interpreted as due to contamination of the finds with Antarctic H or O. However, there is no correlation between the bulk O isotopic compositions of the falls or finds described in the present study and their water contents as determined by stepwise pyrolysis. A caveat is that bulk O isotopic compositions are available for only four of the 12 Antarctic finds, and can differ markedly between subsamples of the same meteorite owing to brecciation. For example, bulk $\delta^{18}\text{O}$ values of different subsamples of EET 96029 differ by 8‰ ($\delta^{18}\text{O}$ 2.40–10.82‰; Lee et al., 2016). Thus, although the findings of Fujiya (2018) cannot be tested in the present study, as the Antarctic finds discussed here are no more

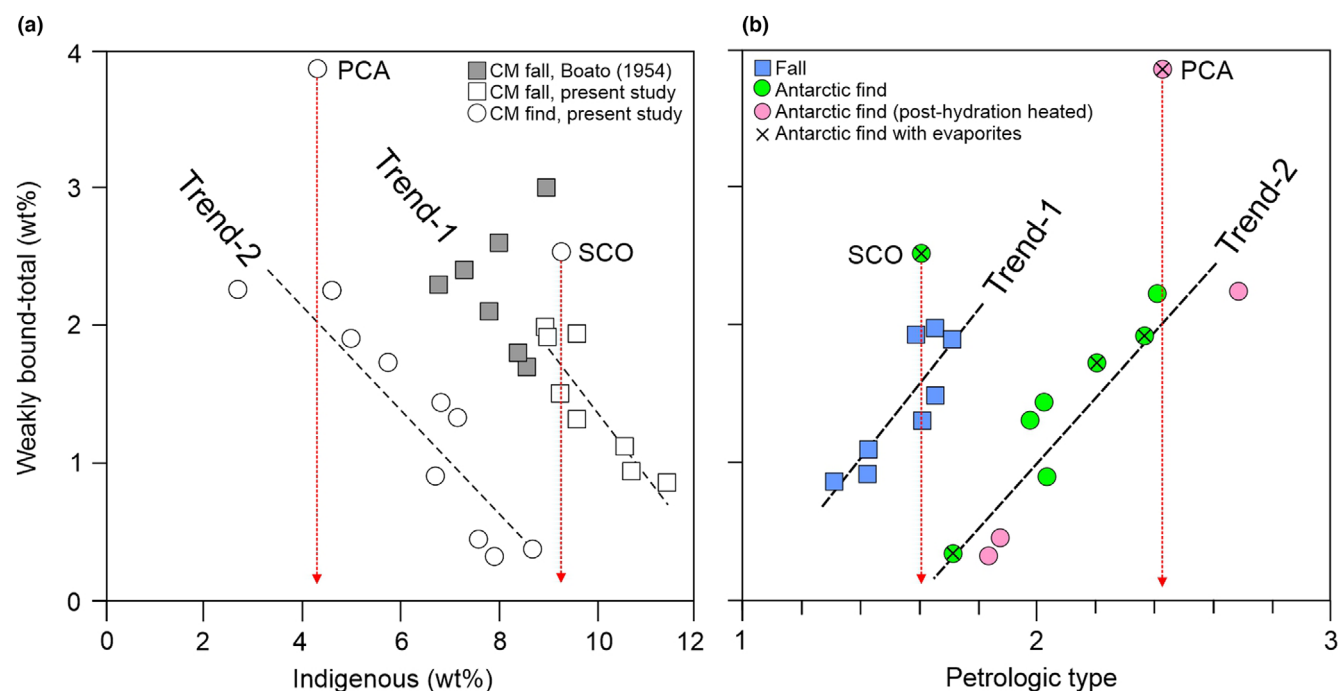


FIGURE 5. (a) Plot of the data in Figure 4a, here shown as white squares (falls) and white circles (finds). Their respective dashed black regression lines define Trends 1 and 2, respectively. The datapoints for PCA 91084 (PCA) and SCO 06043 (SCO) would move somewhere along their respective red lines if evaporite-derived water was subtracted from their weakly bound-total fraction. Also plotted are data from seven samples of CM falls analyzed by Boato (1954) using stepwise combustion and with comparable temperature steps to the present study (Mighei, Murray, Santa Cruz, Haripura, Nawapali, and two samples of Cold Bokkeveld). Only two of those seven samples do not plot close to Trend 1 (Nawapali and one of the Cold Bokkeveld samples). (b) Weakly bound-total fraction plotted against newly calculated petrologic type for the CM falls and finds. Trends 1 and 2 are defined by dashed black regression lines ($R^2 = 0.74$ and 0.85 , respectively). The datapoints for PCA 91084 (PCA) and SCO 06043 (SCO) would move somewhere along their respective red lines if evaporite-derived water was subtracted from their weakly bound-total fraction. (Color figure can be viewed at wileyonlinelibrary.com)

contaminated by terrestrial water than the falls (Figure 2), the differences between falls and finds reported by Fujiya (2018) are more likely to reflect contamination by Antarctic O than by H. This conclusion is also consistent with most of the Antarctic finds having a higher indigenous δD than the falls despite the finds being exposed to low δD Antarctic water.

Relationship Between Terrestrial Contamination and Parent Body Aqueous Alteration

The falls show a negative correlation between the total abundance of weakly bound water and indigenous water, and most of the Antarctic finds have the same relationship although offset to lower indigenous water contents (Figure 4a). Hereafter these correlations are referred to as Trend 1 and Trend 2, respectively (Figure 5). We are confident that the two correlations are not analytical artifacts because five of the seven CM falls that were analyzed by Boato (1954), also using stepped combustion, plot close to Trend 1 (Figure 5a).

Furthermore, TGA data from Antarctic finds in Garenne et al. (2014) show a negative correlation between weight loss at $<200^\circ\text{C}$ and $200\text{--}770^\circ\text{C}$ (i.e., broadly equivalent to the weakly bound and indigenous fractions, respectively), although absolute concentrations of water differ to those measured by stepwise pyrolysis (Figure 6).

Two of the Antarctic finds do not plot on Trend 2: PCA 91084 and SCO 06043. Both have a larger weakly bound-2 fraction than the other finds, which is interpreted as being due to the liberation of water from terrestrial evaporites (Figure 2). As most of the weakly bound fraction of the other samples plotted in Figure 5 is interpreted to come from adsorbed molecular water, PCA 91084 and SCO 06043 cannot be directly compared with them without subtraction of their evaporite-derived water. As the amount of water produced from the evaporites is unknown, PCA 91084 and SCO 06043 could replot anywhere along the two dashed red lines in Figure 5a. Notably, however, PCA 91084 and SCO 06043 would fall close to either end of Trend 2 if, for example, evaporites accounted for ~ 2.0 and ~ 2.5 wt% of their

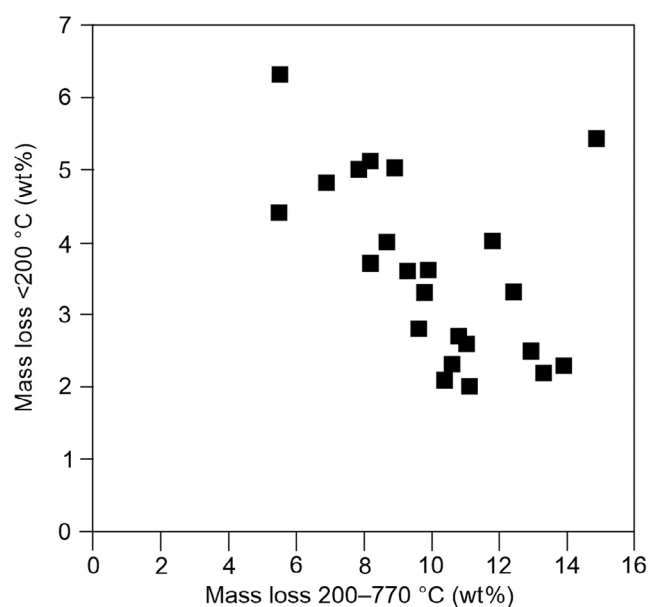


FIGURE 6. TGA data from 22 Antarctic CM finds and one CM fall in Garenne et al. (2014). The mass of water lost at less than 200°C is plotted against the mass lost at 200–770°C (i.e., broadly equivalent to the weakly bound-total and indigenous fractions, respectively). The data show a similar trend to the pyrolysis results in Figure 5a, although note the differences in scales between the two plots.

weakly bound water, respectively. The former figure is close to 1.46 wt% water that would be evolved from PCA 91084 if it contained 7 wt% gypsum as suggested by data in Labidi et al. (2017).

There is a good positive correlation between weakly bound-total fraction and petrologic type for both the falls and finds (Figure 5b), which mirrors the correlation between the weakly bound-total and indigenous fractions (Figure 5a). This close similarity between Figures 5a and 5b is expected since petrologic type has been calculated using the amount of indigenous water in each sample. Nonetheless, Figure 5b highlights that within each trend those samples of lower petrologic type (i.e., that have experienced more intense parent body water/rock interaction) are less able to adsorb terrestrial water. Although the underlying mechanism is unknown, one possibility is that CM serpentine crystals are hydrophobic so that the more altered and phyllosilicate-rich samples attract less water. Alternatively, the more altered samples may have a lower porosity and permeability such that water vapor is less able to access their interior.

It is notable that Trends 1 and 2 comprise falls and finds, respectively. This separation could just be a product of sample selection because most of the falls are of a relatively low petrologic type and the finds a higher petrologic type. However, it is hard to understand why there would be two distinct but parallel relationships

between the weakly bound-total fraction and indigenous fraction/petrologic type if water adsorption was controlled just by mineralogy and/or petrophysical properties (i.e., a single trend would be more likely given that all of the meteorite samples are CM chondrites, and most are similar in mineralogy). We therefore propose that the capacity of a CM sample to adsorb water is also affected by its terrestrial age and/or the environment where it has spent most of its time on Earth. For example, an Antarctic CM of petrologic type 1.6 can adsorb much less water than a fall with a comparable degree of aqueous alteration (Figure 5b).

The Indigenous Water Content of CM Lithologies

The 20 CM fall and find samples discussed have 1.9–10.5 wt% (average 7.0 ± 2.4 wt%) indigenous water (corrected for IOM; Table S5). The lowest water content is from a post-hydration heated find, and most of the falls are at the higher end of the range in accordance with their lower petrologic types. It is notable that the average of 7.0 wt% determined here is very close to the average of 6.95 wt% water equivalent (range 4.2–9.1 wt% water) obtained from 10 CM samples by Vacher et al. (2020). They excluded terrestrial water by preheating their powdered samples (120°C under vacuum for 48 h) prior to analysis.

The water content of the regoliths of C-complex asteroids is quantified by calibrating spectra acquired via remote sensing against laboratory spectra from meteorite samples with known water/OH contents. Terrestrial water needs to be removed from the meteorite samples prior to their measurement (Takir et al., 2013). By using such an approach, Praet, Barucci, Clark, et al. (2021) found that the surface of B-type asteroid Bennu has 6.35 ± 1.43 wt% equivalent water, and Beck et al. (2021) calculated that main belt C-complex asteroids have an equivalent water content of 4.5 ± 4 wt%. The asteroid data are thus within error of the average of the CM falls and finds, although the meteorite samples considered here extend up to 10.5 wt%. These differences at the higher end of the range may be due to: (i) scarcity of low petrologic type and water-rich lithologies within the C-complex asteroid population, or (ii) partial dehydroxylation of the regoliths of asteroids that are analyzed by remote sensing (whereas the properties of most meteorites reflect the composition of their interiors).

CONCLUSIONS

After falling to Earth, CM CCs rapidly adsorb wt% concentrations of water from the atmosphere. The amount of water that they adsorb does not gradually increase with time, but rather reaches a plateau of ~2 wt% or less, probably within days to weeks. CM meteorites do

not have an equal ability to adsorb water but differ depending on the abundance of phyllosilicate, porosity and permeability, or another physical property that varies with their petrologic type. Those properties that affect water adsorption also vary with a meteorite's terrestrial age and/or the environment in which it spent most of its time when on Earth (i.e., curated fall vs. Antarctic find). As the H isotopic composition of indigenous water in most Antarctic finds is higher than that of the falls, their phyllosilicates cannot have significantly exchanged H with low δD Antarctic water. Some finds, and falls that have been on Earth for decades, can however contain telluric water in evaporites that have formed during terrestrial weathering/curation. This work shows that great care must be taken to isolate newly arrived CC lithologies from the terrestrial environment, although even the most quickly collected and carefully curated falls such as Winchcombe cannot escape terrestrial contamination. Where telluric water is taken into account, CM meteorites have comparable water contents to C-complex asteroids including Benu.

Acknowledgments—We thank Alison McDonald (SUERC) for analytical assistance. We are grateful to the Natural History Museum London, Mendy Ouzillou (Skyfall Meteorites), and ANSMET for providing the samples used. US Antarctic meteorite samples are recovered by the Antarctic Search for meteorites (ANSMET) program which has been funded by NSF and NASA, and characterized and curated by the Department of Mineral Sciences of the Smithsonian Institution and Astromaterials Acquisition and Curation Office at NASA Johnson Space Center. This work was funded by the UK Science and Technology Facilities Council through grants ST/T002328/1, ST/T506096/1, and ST/W001128/1, and the UK Natural Environment Research Council through NEIF Facility award 2406.0231. We are very grateful to Ashley King and Lionel Vacher for their constructive and knowledgeable reviews of the original manuscript.

Conflict of Interest Statement—The authors has a conflict of interest in relation to this manuscript.

Data Availability Statement—The data that support the findings of this study are available in the supplementary material of this article.

Editorial Handling—Dr. Josep M. Trigo-Rodríguez

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SUPPORTING INFORMATION

Additional supporting information may be found in the online version of this article.

Table S1. Yield and H isotopic composition of water evolved from CM falls organized by fraction.

Table S2. The sources and curatorial backgrounds of the falls analyzed.

Table S3. Yield and H isotopic composition of water evolved from Antarctic CM finds listed by fraction.

Table S4. Stepwise pyrolysis results for the eight CM carbonaceous chondrite falls.

Table S5. Data used to subtract organic matter-derived water from indigenous water and to calculate petrologic type.