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CM carbonaceous chondrite falls and their terrestrial alteration

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Abstract–The CM carbonaceous chondrites provide unique insights into the composition of the protoplanetary disk, and the accretion and geological history of their parent C-complex asteroid(s). Of the hundreds of CMs that are available for study, the majority are finds and so may have been compromised by terrestrial weathering. Nineteen falls have been recovered between 1838 and 2020, and there is a hint of two temporal clusters: 1930–1942 and 2009–2020. Falls are considered preferable to finds to study because they should be near pristine, and here this assumption is tested by investigating their susceptibility to alteration before recovery and during curation. CMs falling on the land surface are prone to contamination by organic compounds from soil and vegetation. Where exposed to liquid water prior to collection, minerals including oldhamite can be dissolved and most fluid mobile elements leached. Within days of recovery, CMs adsorb water from the atmosphere and are commonly contaminated by airborne hydrocarbons. Interaction with atmospheric water and oxygen during curation over year to decadal timescales can produce Fe-oxyhydroxides from Fe,Ni metal and gypsum from indigenous gypsum and oldhamite. Relationships between the petrologic (sub)types of pre-1970 falls and their terrestrial age could be due to extensive but cryptic alteration during curation, but are more likely a sampling bias. The terrestrial history of a CM fall, including circumstances of its collection and conditions of its curation, must be taken into account before it is used to infer processes on C-complex parent bodies such as Ryugu and Bennu.

INTRODUCTION

Carbonaceous chondrite meteorites sample primitive bodies, most probably C-complex asteroids (Burbine 2017; Bates et al. 2020). As such they contain materials that were accreted from the protoplanetary disk including presolar grains, chondrules, refractory inclusions, and organic matter (Brearley and Jones 1998). In almost all carbonaceous chondrites, many of these original constituents have been partially altered or destroyed by one or more parent body processes including radiogenic and shock heating; impact deformation; space weathering; and, most significantly, aqueous alteration (Bunch and Chang 1980; Grimm and McSween 1989; Scott et al. 1992; Rubin et al. 2007; Rubin 2012). Products of this water/rock interaction include phyllosilicates and carbonates that can provide valuable insights into the timescale of parent body processing, and the delivery of water and other volatiles to the inner planets (Fujiya et al. 2012; Alexander et al. 2012; Alexander 2017).

The records of protoplanetary disk composition and parent body evolution that are contained within carbonaceous chondrite meteorites will be potentially compromised by their time on Earth. Serendipitous meteorite finds have been exposed to the deleterious effects of liquid water, solar heating, and other agents, in most cases for an indeterminate length of time (Gooding 1986; Velbel 1988; Velbel et al. 1991; Lee and Bland 2004; Bland et al. 2006), although short-lived radionuclides can be used to approximate the duration of terrestrial residence (Nishiizumi et al. 1989; Jull 2001, 2006). The most pristine samples of C-complex asteroids should therefore be those meteorites that were seen to fall, recovered shortly afterward, and subsequently curated under controlled conditions. Nonetheless, prior to recovery and during storage, these meteorites may react with the atmosphere, hydrosphere/cryosphere, and biosphere, leading to contamination by inorganic and organic compounds that are also available to react with pre-terrestrial constituents (Velbel 2014). The damaging effects of terrestrial exposure are highlighted by several CI carbonaceous chondrite falls that are encrusted by terrestrial evaporites (Gounelle and Zolensky 2001; King et al. 2020).

Here, we have sought to evaluate the vulnerability of CM carbonaceous chondrite falls to terrestrial alteration. There is reason to believe that the CMs could be just as susceptible as the CIs because meteorites of both groups are composed mainly of phyllosilicates. Developing a good understanding of how pristine the CM falls are is important because they are likely to be the closest meteorite analogs for two C-complex asteroids that are the targets of current sample return missions. Ryugu is a Cb-type asteroid with a CM-like mineralogy (Perna et al. 2017; Kitazato et al. 2019; Bates et al. 2020) and Bennu is a B-type asteroid that has an affinity to CI/CM meteorites (Clark et al. 2011; Hamilton et al. 2019). While these missions are designed to return pristine samples, comparison with the CM meteorites can help to understand Ryugu and Bennu in the context of the wider population of C-complex asteroids.

COMPOSITION OF THE CM GROUP

There are 19 CM falls (Table 1), and in order to evaluate whether they have been terrestrially altered, we first outline the nature and properties of the group. CM meteorites have a petrologic type of less than 3 (Van Schmus and Wood 1967) showing that their original constituents (mainly anhydrous silicate, metal, and sulfide) were aqueously altered in a parent body environment (e.g., Bunch and Chang 1980; Browning and Bourcier 1998; Brearley 2006; Pignatelli et al. 2016). The main products of aqueous alteration are finely crystalline (sub-micrometer size) phyllosilicates, most of which occur in the fine-grained matrix, together with subordinate <~100 µm size grains of sulfides, carbonates, and iron oxides (Bunch and Chang 1980; Barber 1981; Howard et al. 2015).

Much work has been done to describe and understand the degree of aqueous alteration of the putative initial CM lithology (i.e., petrologic type 3) through mineralogical, chemical, and isotopic analyses of bulk samples or thin sections. Most of these studies have considered finds and falls together, and so make the tacit assumption that the CMs are not measurably affected by terrestrial alteration. The mineralogic alteration index (MAI) of Browning et al. (1996) tracks aqueous alteration through progressive changes in matrix mineralogy and chemical composition, from being dominated by cronstedtite to MgFe serpentine. These mineralogical changes lead to a reduction in the concentration of total Fe and Fe^{3+} of matrix phyllosilicates, with the ferric component being calculated from site occupancy (Velbel and Palmer 2011). The scheme of Howard et al. (2009, 2011, 2015) classifies the petrologic type of bulk CMs using their mineralogy as quantified by X-ray diffraction. Petrologic type 1.0 has a phyllosilicate fraction (volume of total phyllosilicate/anhydrous silicate + total phyllosilicate) of more than 95% and type 3.0 less than 5%. Finds have a broader range of petrologic types than falls (1.1–1.7 versus 1.2/1.4–1.5, respectively; Table 1, Fig. 1a). Rubin et al. (2007) classified CMs by petrologic subtype using a set of petrographic, mineralogical, and chemical properties: nature of mesostasis and mafic silicate phenocrysts in chondrules; abundance of matrix phyllosilicates, metallic Fe,Ni, and PCP clumps; mineralogy of sulfides and carbonates; "FeO"/SiO_2 composition of PCP. The classification scheme ranges from CM2.0 (most highly altered) to CM3.0. Meteorites of subtype CM2.0–CM2.6 were identified by Rubin et al. (2007) and CM2.7 was described subsequently (Rubin 2015), with falls have a broader range of subtypes than finds. Lentfort et al. (2020) determined the petrologic subtypes of CM chondrites using just the "FeO"/SiO_2 composition of tochilinite–cronstedtite intergrowths (TCI, formerly PCP). The ratio decreases with increasing degree of aqueous alteration, and by employing this metric, they showed that many CMs, both falls and finds, contain clasts of different petrologic subtype. Lentfort et al. (2020) identified clasts of subtypes ranging from CM2.0 to CM2.9, with falls containing clasts spanning the whole range apart for CM2.0 (Table 1).

Despite the range in petrologic (sub)types, the CM group is chemically homogeneous with regard to most elements (Brearley 2006; Rubin et al. 2007; Braukmüller et al. 2018). However, individual meteorites can be depleted/enriched in fluid-mobile elements due to terrestrial weathering (see below), and the abundances and isotopic compositions of H, C, N, and O vary considerably throughout the group (Alexander et al. 2012; Fig. 2). Intermeteorite differences in carbon mainly reflect the abundance of organic matter. The CMs are dominated by insoluble organic matter

Table 1. CM falls listed in chronological order, and their classifications.

	Fall details[a]							
	Date[b]	Location/latitude	Mass	MAI[c]	Petrologic subtype[d]	Petrologic subtype[e]	Petrologic type[f]	Petrologic type[g]
Kolang	01/08/2020	Indonesia/1°N	2.55 kg	–	–	–	–	–
Aguas Zarcas	23/04/2019	Costa Rica/10°N	27 kg	–	CM2.2 ± 0.1[l]	–	1.3–1.5[m]	–
Mukundpura	06/06/2017	India/26°N	2 kg	–	~CM2.0[k]	–	–	–
Shidian	27/11/2017	China/24°N	1809 g	–	CM2.2[j]	–	–	–
Maribo	17/01/2009	Denmark/54°N	25.8 g	–	~CM2.6[i]	CM2.6	–	–
Sayama	29/04/1986	Japan/35°N	430 g	–	–	–	–	–
Murchison	28/09/1969	Australia/36°S	100 kg	0.43	CM2.5	CM2.7-2.9	1.5	1.6
Murray	20/09/1950	USA/36°N	12.6 kg	0.57	CM2.4/2.5	–	1.5	1.5
Pollen	16/04/1942	Norway/66°N	254 g	0.53	–	–	–	–
Erakot	22/06/1940	India/19°N	113 g	–	–	–	–	–
Santa Cruz	03/09/1939	Mexico/24°N	60 g	–	–	CM2.7	1.4[h]	–
Crescent	17/08/1936	USA/35°N	78.4 g	–	–	–	–	–
Banten	24/04/1933	Indonesia/6°S	629 g	–	–	CM2.6-2.9	–	1.7
Boriskino	20/04/1930	Russia/54°N	1342 g	0.73	–	–	–	–
Haripura	17/01/1921	India/28°N	315 g	–	–	–	–	–
Nawapali	06/06/1890	India/21°N	105 g	–	–	–	–	–
Mighei	18/06/1889	Ukraine/48°N	8 kg	0.77	–	–	1.4	1.6
Nogoya	30/06/1879	Argentina/32°S	4 kg	0.97	CM2.2	CM2.2-2.5	1.2/1.4	1.1/1.6
Cold Bokkeveld	13/10/1838	South Africa/33°S	5.2 kg	1.03	CM2.2	CM2.1-2.7	1.4	1.3

This list does not include Diepenveen (27/10/1873, Netherlands/52°N, 68.4 g) that is classified as CM2-an in the Meteoritical Bulletin database, and Sutter's Mill (22/04/2012, USA/38° N, 993 g) that is classified as C (Ruzika et al. 2014), although described as a CM2 breccia (Jenniskens et al. 2012; Zolensky et al. 2014).

[a] Data from the *Meteoritical Bulletin*.

[b] Day/month/year.

[c] Mineralogic Alteration Index (Browning et al. 1996).

[d] As defined by Rubin et al. (2007).

[e] Data from Lentfort et al. (2020). The range of petrologic subtypes of the clast population in each meteorite is indicated.

[f] As defined byHoward et al. (2015)

[g] As defined by Alexander et al. (2013).

[h] Santa Cruz has a phyllosilicate fraction of 0.78 (King et al. 2019).

[i] Estimated by Haack et al. (2012).

[j] Estimated by Fan et al. (2020).

[k] Estimated by Rudraswami et al. (2019).

[l] Estimated by Martin and Lee (2020).

[m] Measured by Davidson et al. (2020).

(Alexander et al. 2007) whereas the soluble fraction comprises a wide range of compounds including carboxylic acids; aromatic, aliphatic, and polar hydrocarbons; hydroxy acids; and amino acids (Pizzarello et al. 2006). Finds analyzed by Alexander et al. (2012) have a wider range of C than the falls, which are more numerous at the high C/low $\delta^{13}C$ end (Figs. 2a and 2b). Intermeteorite variations in the concentration and isotopic composition of H mainly reflect different proportions of phyllosilicates (high H/low δD) to organic matter (low H/high δD; Alexander et al. 2012, 2013; Figs. 2c and 2d). CM finds again have a wider range than falls, the latter tending toward the high H end (Fig. 2c). The amount of bulk rock H present as water/OH was used by Alexander et al. (2013) to classify CMs by petrologic type. The samples they analyzed range in petrologic type from 1.1 (most highly hydrated and H-rich) to 1.9, with falls having a narrower range than finds (Table 1, Fig. 1b). The oxygen isotopic compositions of the CMs vary considerably: $\delta^{18}O = 22.3$ to 4.5‰; $\delta^{17}O = 23.9$ to 0.2‰; $\Delta^{17}O = 0.4$ to -4.2‰ (Clayton and Mayeda 1999; $n = 32$). Intermeteorite differences partly reflect the ratio of anhydrous silicates (isotopically light) to phyllosilicates (isotopically heavy), although there is no direct correlation between oxygen isotopic composition and petrologic (sub)type.

Identifying Terrestrial Alteration

The mineralogical, chemical, and isotopic diversity of the CM group will make identifying terrestrial alteration in finds and falls challenging. These difficulties will be exacerbated because some of the

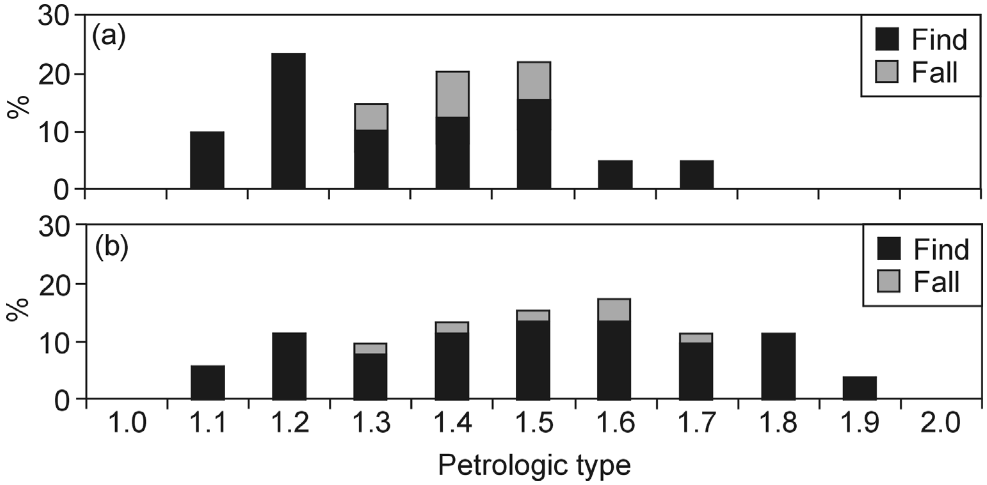

Fig. 1. The petrologic types of CM falls and finds. a) Meteorites classified using the scheme of Howard et al. (2015). Data are from Howard et al. (2015), King et al. (2017, 2019), and Lee et al. (2019a, 2019b). *n* = 33 finds, 6 falls. b) Meteorites classified by Alexander et al. (2013). *n* = 46 finds, 6 falls. Averages have been taken where the same meteorite was analyzed twice, whereas paired meteorites are treated as individuals. In both classifications, finds span a wider range than falls.

minerals that are produced by parent body aqueous alteration (e.g., calcite, iron oxide) can also form readily under Earth surface conditions, and the diverse inventory of indigenous organics may be hard to distinguish from natural and anthropogenic compounds. Criteria that can be used individually or in combination to help identify terrestrial alteration of a find or fall include cases where:

- Differences in chemical/isotopic composition or mineralogy within a meteorite are associated with post-fall features such as mineral veins cross-cutting the fusion crust, or compositional gradients with respect to a stone's outer surface.
- The chemical/isotopic composition or mineralogy of a set of meteorites varies in accordance with their terrestrial age.
- The chemical/isotopic composition or mineralogy of one or more meteorites is anomalous relative to most other CMs. Such meteorites may have specific terrestrial histories (e.g., finds from a similar climatic setting, falls or finds with similar curatorial histories), or post-fall circumstances such as contact with liquid water.

TERRESTRIAL WEATHERING OF CM FINDS

The weathering of CM finds can be used to predict the likely processes and products of terrestrial alteration of falls. The main visual criterion for assessing the degree of terrestrial weathering of meteorites is their degree of "rusting" (e.g., Bland et al. 1998). Rust forms by the exposure of Fe,Ni metal to water (hydrolysis and oxidation) to produce minerals including magnetite ($Fe^{3+}{}_2Fe^{2+}O_4$), goethite (α-$Fe^{3+}OOH$), lepidocrocite (γ-$Fe^{3+}OOH$), akaganéite ($Fe^{3+}[O,OH,Cl]$), and limonite (i.e., goethite plus lepidocrocite with some adsorbed water; Deer et al. 1992; Bland et al. 1998, 2006; Lee and Bland 2004). The Meteorite Working Group recognizes three categories (A, B, C) that correspond to minor, moderate, and severe rustiness, respectively. Wlotzka (1993) proposed a seven-point weathering classification for finds in thin section whereby W0–W5 describe the progressive rusting of Fe,Ni metal, and W5–W6 the subsequent breakdown of silicates. Antarctic and hot desert CM finds range in weathering category from A to C (The Meteoritical Bulletin). The rust in these meteorites occurs as finely layered rims of minerals including akaganéite and goethite on Fe,Ni metal grains, and narrow veins cross-cutting the matrix (Floyd 2019; Floyd and Lee 2020; Fig. 3). Atmospherically derived Cl catalyzes akaganéite crystallization, and hence, its supply is an important determinant of metal alteration (Floyd 2019). While these rims are readily identifiable using light and scanning electron microscopy, incipient alteration of metal at the nanoscale requires transmission electron microscopy (TEM). From a detailed TEM study of metal grains in the fine-grained rims of the Antarctic CM find Yamato 791198, Chizmadia et al. (2008) showed that the surfaces of micrometer-size grains of metal have sub-μm scale pitting that has been accompanied by the formation of $Fe(OH)_2$. Although parent body alteration was favored, Antarctic weathering could not be ruled out.

The presence of evaporites is also used to classify the weathering of meteorite finds (Velbel 1988). These

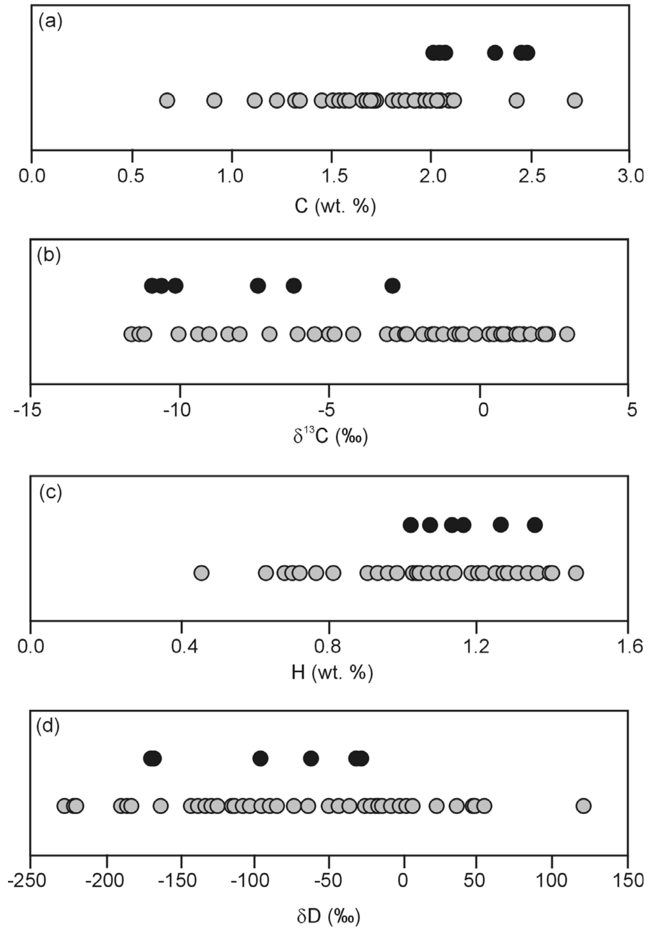

Fig. 2. Carbon a, b) and H c, d) chemical and isotopic compositions for CM finds (dark gray symbols, *n* = 45) and falls (black symbols, *n* = 6). Data from Alexander et al. (2012). Averages have been used where the same meteorite was analyzed twice whereas paired meteorites are plotted individually. In all four plots, the falls tend to lie toward one end of compositional range of the finds, which is more pronounced for C than H.

minerals comprise Ca- and Mg-sulfates including gypsum (Velbel 1988; Losiak and Velbel 2011). Evaporites are commonplace on Antarctic finds but differ in abundance between meteorite groups (Velbel 1988; Losiak and Velbel 2011). CMs are considerably more prone to evaporite formation than ordinary chondrites, probably because cations including Mg and Ca can be more readily sourced from within the meteorites themselves (e.g., by weathering of serpentine group minerals and calcite). Carbonate minerals can also form on Antarctic finds by evaporation. For example, using oxygen and carbon isotopes (δ^{18}O, δ^{13}C, ^{14}C), Jull et al. (1988) showed that nesquehonite had grown since 1950 by the interaction of Lewis Cliff 85320 (H5 chondrite) with meltwater and atmospheric CO_2. Using the same isotope systems, Tyra et al. (2007) identified terrestrial carbonates in the Antarctic CM Elephant Morraine (EET) 96006 and its pairs. A proportion of the carbonate C was atmospherically derived, and its oxygen isotope ratios reflected interaction of the meteorite with meltwater.

Terrestrial weathering can alter bulk chemical compositions of meteorite finds including the CMs. Hot desert weathering may cause enrichment in elements, for example, Ca, Sr, Ba, Li, and U in the case of the CM Jbilet Winselwan (Friend et al. 2018; King et al. 2019). Antarctic CMs are less affected, although some are depleted in highly fluid mobile elements including K, Na, and Rb (Braukmüller et al. 2018). As noted above, the abundance of evaporites in Antarctic CMs suggests that Mg, Ca, C, and S can be leached during cold desert weathering.

EVIDENCE FOR TERRESTRIAL ALTERATION OF CM FALLS

CM falls are mineralogically and chemically less diverse than the finds, as shown by their narrower range of petrologic (sub)types and elemental/isotopic compositions (Table 1; Figs. 1 and 2). Below we describe and evaluate evidence for terrestrial alteration of CM falls in the context of four interrelated processes and products, including (1) organic contamination, (2) leaching and dissolution by liquid water, (3) hydrolysis and oxidation, (4) terrestrial overprinting of pre-terrestrial aqueous alteration.

Organic Contamination

The potential for contamination of falls by organic compounds has been a focus of much attention owing to the variety and cosmochemical significance of indigenous organics. One terrestrial source of organic compounds is the site where the meteorite fell. For example, Pizzarello and Yarnes (2018) identified amino acids and lactic acid in samples of Mukundpura, which are likely to have come from the agricultural ground where the stones were recovered. Glavin at al. (2020) found that Aguas Zarcas had also been contaminated by amino acids from local soil and vegetation.

Falls are readily contaminated by hydrocarbon aerosols, and the susceptibility of carbonaceous chondrites is probably due to the insoluble carbonaceous phases that they contain (Cronin and Pizzarello 1990). *n*-alkanes have been detected on CM falls including Cold Bokkeveld, Murchison, and Murray, and interpreted to have been derived from petroleum (Oró et al. 1968; Cronin and Pizzarello 1990; Sephton et al. 2001). Kerridge (1985) recorded differences in carbon concentrations of Cold Bokkeveld

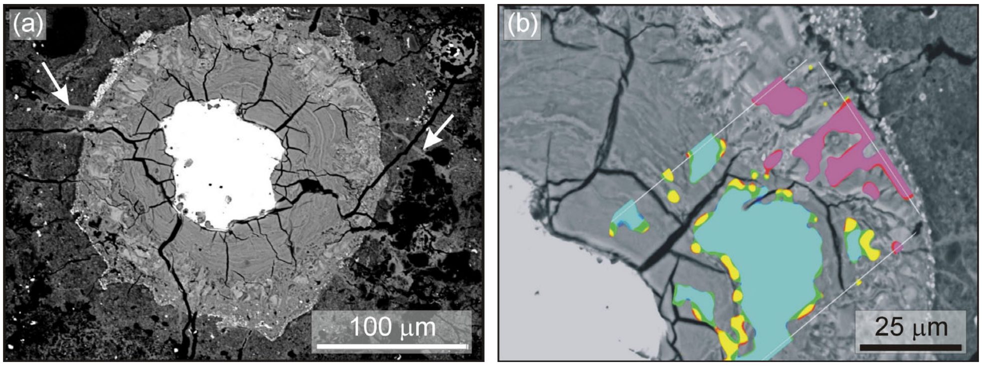

Fig. 3. Images of a weathered grain of Fe,Ni metal in the Antarctic CM find Lewis Cliff (LEW) 85311. a) Backscattered electron SEM image. A grain of kamacite (white) is enclosed by an ~70 μm thick concentrically layered rim of weathering products (gray). Narrow veins of weathering products cross-cut the matrix (arrowed). b) Backscattered electron SEM image of the upper right hand side of the rim in (a). Overlain on part of the image is a map showing the mineralogy of different parts of the rim as determined by Raman spectroscopy. Pink denotes tochilinite, blue denotes akaganéite, and yellow is goethite (Floyd and Lee 2020). Tochilinite was the first mineral to form, by parent body aqueous alteration of Fe,Ni metal (Palmer and Lauretta 2011). The tochilinite may have partially protected the Fe,Ni metal from subsequent terrestrial hydrolysis and oxidation (Floyd 2019; Floyd and Lee 2020). (Color figure can be viewed at wileyonlinelibrary.com.)

samples from different meteorite collections, which they interpreted as being due to terrestrial contamination. The contamination of falls by leaded fuel is demonstrated by the high concentration and isotopic composition (^{207}Pb/^{206}Pb) of Pb in Cold Bokkeveld, Nogoya, and Mighei (the three oldest falls) relative to other CM falls and finds (Braukmüller et al. 2018). Cronin and Pizzarello (1990) noted that vacuum pumps and rock saws used in sample preparation are another source of *n*-alkanes. Organic contaminants can also include microorganisms, as shown by Oró and Tornabene (1965), who cultured 6000 bacteria per g from Murray ~15 years after its fall. Although differences in the abundance and isotopic composition of C between CM falls and finds are most likely due to indigenous organic matter (Figs. 2a and 2b), there could also be an anthropogenic overprint on the falls from C contamination during curation or sample preparation (e.g., the use of acetone; Alexander et al. 2012).

Carbonaceous chondrites can be contaminated very rapidly. Han et al. (1969) identified *n*-alkanes on the surfaces of samples of Allende (CV3) that had been collected just 7 days after falling in 1969. Although they are most abundant on the outer surfaces of stones, organic contaminants readily penetrate into their interior. Cronin and Pizzarello (1990) recorded phthalates from 1.2 to 2.5 cm below the outer surface of Murray, and concluded that although pristine indigenous hydrocarbons can be extracted from the interiors of stones, great care must be taken to isolate samples from airborne contaminants and those sourced from sample preparation. Thus, the analysis of organic matter must be undertaken as soon as possible after the fall of a CM and ideally using samples extracted from the interior of a stone.

Leaching and Dissolution by Liquid Water Before Recovery

The contact of carbonaceous chondrite falls with liquid water prior to recovery can have a significant impact on their mineralogy and composition, as highlighted by Tagish Lake and Sutter's Mill. Tagish Lake (C2-ung) fell in 2000. Some stones were collected promptly from the surface of a frozen lake, whereas others were recovered several months later, during which time they had been variously degraded by interaction with meltwater (Brown et al. 2000). The degraded samples had lost water-soluble elements (Friedrich et al. 2002), with Na, Ca, K, Sr, and Ba being depleted relative to median CM compositions (Braukmüller et al. 2018). Tagish Lake was also contaminated by amino acids from the meltwater (Kminek et al. 2002). Despite chemical alteration, there is little evidence that exposure to meltwater had affected the mineralogy of the degraded samples. Izawa et al. (2010) found that there had been some loss of saponite and serpentine by physical separation to give the degraded samples a higher density, and suggested that the absence of gypsum could be due to dissolution of pre-terrestrial sulfates.

Sutter's Mill is officially classified a C carbonaceous chondrite (Ruzicka et al. 2014), although is also described as a CM breccia (Jenniskens et al. 2012; Zolensky et al. 2014). It fell in 2012, and samples were recovered both before and after heavy rainfall.

Comparison of the two sets of stones demonstrated that liquid water had affected both mineralogy and chemical composition. Oldhamite (CaS) was present in the pre-rain samples but absent from those collected post-rain (Jenniskens et al. 2012). One of the pre-rain stones analyzed contained acetate (700 ppm), formate (80 ppm), sulfate (1300 ppm), and chloride (262 ppm), whereas the interior of a post-rain stone had ~100 ppm acetate and only traces of the other compounds (Jenniskens et al. 2012). Furthermore, water had redistributed Re, and to a lesser extent Os, thus compromising the Re-Os isotopic system (Walker et al. 2018). The rapid interaction of Sutter's Mill with water may have been facilitated by its porosity of 31.0 ± 1.4% (Jenniskens et al. 2012), which is within the range of the CMs (15.0–36.7%) although higher than the average of 24.7% (Macke et al. 2011). However, the interconnectivity of pores in Sutter's Mill was not quantified, and as the finely crystalline matrices of carbonaceous chondrites have a very low permeability (Bland et al. 2009), ingress of water must have been facilitated by fractures and other macroscale interconnected pores.

Other CM falls have been exposed to liquid water prior to recovery. Fragments of Murray, Murchison, and Aguas Zarcas were collected both before and after rainfall (Horan 1953; Jarosewich 1971; Davidson et al. 2020). Pieces of Cold Bokkeveld are recorded to have fallen onto moist ground (MacLear 1839) and Sayama was water soaked (Yoneda et al. 2001). Haack et al. (2012) noted that Maribo had been disaggregated in situ, which they interpreted to have been caused by penetration of water into the stone followed by freeze–thaw; this water could also have altered the meteorite's chemical composition or mineralogy. One of the Kolang stones was collected from a rice paddy, but it is unclear whether it had been in contact with liquid water (Meteoritical Bulletin). Although at least a third of the 19 CMs were exposed to liquid water before recovery (i.e., Aguas Zarcas, Cold Bokkeveld Maribo, Murchison, Murray, Sayama), the only evidence for associated alteration is the possible leaching of Rb from Cold Bokkeveld (Mittlefehldt and Wetherill 1979) and Na, K, and Ca from Murray (Rubin et al. 2007). The contact of carbonaceous chondrite falls with liquid water can also lead to the precipitation of new minerals, for example, calcite on the CV3 Vigarano (fell 1910, Italy; Abreu and Brearley 2005). One of the stones was recovered after a month on the ground surface, during which time it had interacted with calcium-bearing soil water to precipitate calcite on the fusion crust and extending into the interior of the stone as narrow veins. Given the sensitivity of carbonaceous chondrites to liquid water, a systematic study of the falls that were so exposed is warranted, particularly in cases where fragments were recovered both before and after rainfall.

Hydrolysis and Oxidation from Atmospheric Exposure

Exposure to the Earth's atmosphere prior to recovery and during curation is another source of water that can potentially alter CM falls. Jarosewich (1990) chemically analyzed six CMs and found that they had 6.54–12.05 structural water (H_2O^+) and 1.33–2.69 wt% hydration water (H_2O^-, determined by weight loss at 110 °C). Thus, 11–41% of the water hosted by these CMs is likely to be terrestrial; the two falls analyzed, Banten and Murchison, have 18% and 22% hydration water, respectively. These results are consistent with experiments by Vacher et al. (2020) showing that ~10–30% of the hydrogen inventory of CMs (falls and finds) is loosely bound and lost by heating at 120 °C for 48 h. The constituents of the CMs that are most susceptible to alteration by this adsorbed water are Fe,Ni metal and S-rich phases.

Fe,Ni Metal

Previous work on ordinary chondrite falls has shown that adsorbed water can alter Fe,Ni metal, by hydrolysis and oxidation, over curatorial timescales. Lee and Bland (2004) examined two L6 falls, Barwell (fell 1965) and New Concord (fell 1860). Fe,Ni metal in Barwell is pristine (weathering classification W0) whereas within ~105 years, New Concord kamacite had been altered to form ~15 μm thick rust rims (i.e., W1; Lee and Bland 2004). As regards CM falls, the effects of terrestrial hydrolysis and oxidation should be most evident in meteorites of higher petrologic (sub)types that contain the most metal (e.g., up to ~1 vol%; Rubin et al. 2007; Howard et al. 2015). Murchison and Murray are both of a high (sub)type (Table 1) and were classified as W1–2 by Rubin et al. (2007) owing to the presence of rims of limonite on Fe,Ni metal grains. Rust rims had thus formed in less than ~38 years (Murchison) and ~57 years (Murray). There is some evidence from Murchison that rust can form on shorter timescales. Fuchs et al. (1973) observed millimeter-size patches of hydrated iron oxide on freshly broken and fusion crusted surfaces, and Mittlefehldt and Wetherill (1979) also noted samples with flecks of rust. Given the dates of these publications, the samples of Murchison studied (fell in 1969) had been exposed to the atmosphere for <4 years and <10 years, respectively. Although the presence of rust rims on Fe,Ni metal grains indicates terrestrial alteration, the reaction is likely to be limited by the lack of Cl in curatorial environments. Chlorine plays a key role in catalyzing the alteration of metal (Buchwald and Clarke 1989) as demonstrated by the presence of Cl-bearing akaganéite in Antarctic finds (Floyd and Lee 2020; Fig. 3).

S-Rich Phases

Sulfate evaporites may be expected to form by terrestrial alteration of CM falls because these minerals are abundant in CI falls including Orgueil (fell 1864) and Ivuna (fell 1938; Gounelle and Zolensky 2001; King et al. 2020). In addition, CM finds are susceptible to evaporite formation during Antarctic weathering (Velbel 1988). The CI evaporites are interpreted to have been produced by the hydrolysis and oxidation of sulfides during curation, and possibly also by the dissolution–reprecipitation of pre-terrestrial sulfates. They could have appeared within 2 weeks of the fall of Orgueil (Gounelle and Zolensky 2001), and King et al. (2020) found that epsomite/blödite had developed on a polished sample of Ivuna in less than 6 years.

Gypsum has been identified in the CM falls Cold Bokkeveld, Nogoya, Banten, Mighei, and Murchison (Fuchs et al. 1973; Burgess et al. 1991; Lee 1993; Howard et al. 2009; Labidi et al. 2017). The Cold Bokkeveld gypsum was interpreted to have formed during the waning stage of parent body aqueous alteration (Lee 1993). Evidence for this pre-terrestrial origin included the presence of multiple generations of veins indicating two or more episodes of fracturing and crystal growth. TEM proved particularly useful for illustrating of cross-cutting relationships between gypsum, calcite, and serpentine within the fine-grained matrix (Lee 1993). The oxygen isotopic composition of water-soluble sulfate extracted from six CM falls further supports a pre-terrestrial origin (Airieau et al. 2005). If the sulfate had formed post-fall, its Δ^{17}O composition should plot on the terrestrial fractionation line (TFL), as is the case for Orgueil and Ivuna (Δ^{17}O -0.12 and -0.08, respectively; Airieau et al. 2005). Of the six CM falls that were analyzed by Airieau et al. (2005), only Nogoya is within error of the TFL (Δ^{17}O -0.04). A parent body origin for CM evaporites is also consistent with good correlations between sulfate Δ^{17}O and two measures of aqueous alteration of the host meteorites (MAI and petrologic subtype; Fig. 4). These correlations thus suggest that sulfates crystallized from fluids whose oxygen isotopic compositions evolved with progressive aqueous alteration. This model is supported further by the sulfur isotopic composition of CM sulfates, which indicate that they formed by oxidation of S^0 in a parent body environment (Labidi et al. 2017). A parent-body origin for gypsum could be supported in future work if veins were found to be restricted to individual clasts of meteorite breccias.

Despite this evidence for a pre-terrestrial origin of CM sulfates, a proportion of them may also have formed post-fall. Fuchs et al. (1973) described gypsum plates on Murchison hand specimens. They interpreted the sulfate to have been created by redeposition of a pre-terrestrial vein-filling phase, which would have taken less than 4 years given that Murchison fell in 1969. Fuchs et al. (1973) also described gypsum on surfaces of pieces of Cold Bokkeveld that may have formed during curation. Oldhamite is a potential indigenous source of S given that it is highly reactive in the presence of liquid water (Jenniskens et al. 2012). The potential for rapid mobilization of S is important to consider when characterizing samples of Ryugu and Bennu that will be small and so potentially highly reactive. If samples are exposed to the terrestrial atmosphere, mineralogical and chemical characterization should be undertaken quickly (Velbel 2014).

ALTERATION CHRONOLOGY OF CM FALLS

Nineteen CM falls were recovered between 1838 (Cold Bokkeveld) and 2020 (Kolang); 21 if Diepenveen (CM2-an) and Sutter's Mill (C) are included. The distribution of fall dates hints at the possibility of two clusters ~80 years apart, namely (1) 1930–1942 (six in 12 years); (2) 2009–2020 (six in 11 years; Table 1, Fig. 5). Eighteen of the 21 falls were recorded between April and November (Fig. 5a), which is consistent with most being recovered in the northern hemisphere where they are more likely to be observed at those times of year (Fig. 5b). There is no evidence that members of each of the two putative clusters have fallen at specific times of year as might be expected if they were delivered by a meteoroid stream. The average mass of the falls is 3.4 kg (excluding Murchison), with the 1930–1942 cluster averaging 0.4 kg/fall and the 2009–2020 cluster 5.7 kg/fall (Fig. 5c).

The disequilibrium admixture of phases within most CMs (e.g., olivine/pyroxene chondrules and a phyllosilicate-rich matrix) is conventionally interpreted as due to the partial parent body aqueous alteration of anhydrous materials accreted from the protoplanetary disk (e.g., Browning and Bourcier 1998; Bunch and Chang 1980). However, it has also been proposed that CM falls could have been terrestrially modified so that the abundance and composition of aqueous alteration products reflect both pre- and post-fall processes (Benedix and Bland 2004; Bland et al. 2006). Key evidence for this suggestion is a close correlation between the terrestrial ages of the CM falls and their MAI ($R^2 = 0.91$, $n = 7$; i.e., older falls are more altered because they had longer to interact with the terrestrial environment) (Fig. 6a). Benedix and Bland (2004) and Bland et al. (2006) suggested that terrestrial mobilization of sulfur may have contributed to the age/

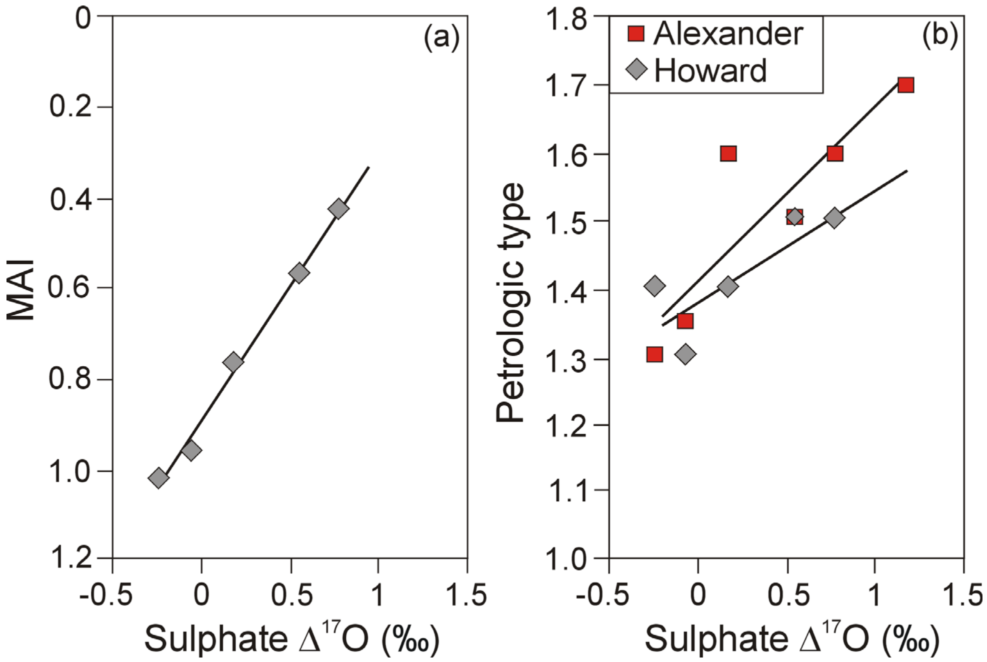

Fig. 4. The oxygen isotopic composition of water soluble sulfate in six CMs: Cold Bokkeveld, Nogoya, Mighei, Banten, Murray, Murchison. Data from Airieau et al. (2005). a) $\Delta^{17}O$ versus mineralogical alteration index (MAI; $n = 5$; $R^2 = 0.99$). b) $\Delta^{17}O$ versus petrologic type as determined by Alexander et al. (2013; $n = 6$; $R^2 = 0.77$; two analyses of Nogoya averaged) and Howard et al. (2015; $n = 5$; $R^2 = 0.66$; two analyses of Nogoya averaged). (Color figure can be viewed at wileyonlinelibrary.com.)

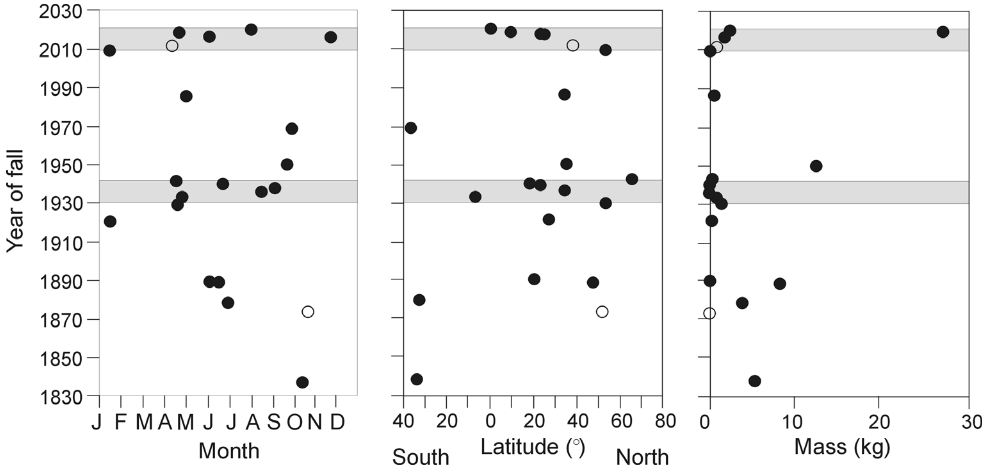

Fig. 5. The year of fall of the CMs plotted against the month of fall, latitude of the recovery site, and size (Murchison [100 kg] is not shown). Filled circles are the 19 falls listed in Table 1, with Diepenveen and Sutter's Mill plotted as open circles. Eighteen CMs fell between mid April and late November, and 17 in the northern hemisphere. There is a hint that 12 of the falls are concentrated in two temporal clusters, which are shaded: 1930–1942 and 2009–2020.

alteration correlation because the calculation of MAI uses sulfur concentrations as measured by electron probe to correct for any sulfide-hosted Fe. In addition, oxidation of sulfides can liberate sulfuric acid that is available to consume anhydrous silicates (Velbel 2014). Indeed, Velbel (2014) found that meteoritic olivine grains tens of micrometers in size can dissolve under ambient conditions and over fractions of a year to decadal timescales. Thus, several processes could have operated independently or in combination to increase the MAI of CM falls over time.

This hypothesis can be tested by comparing the ages of falls with their degree of aqueous alteration as determined using different mineralogical and chemical

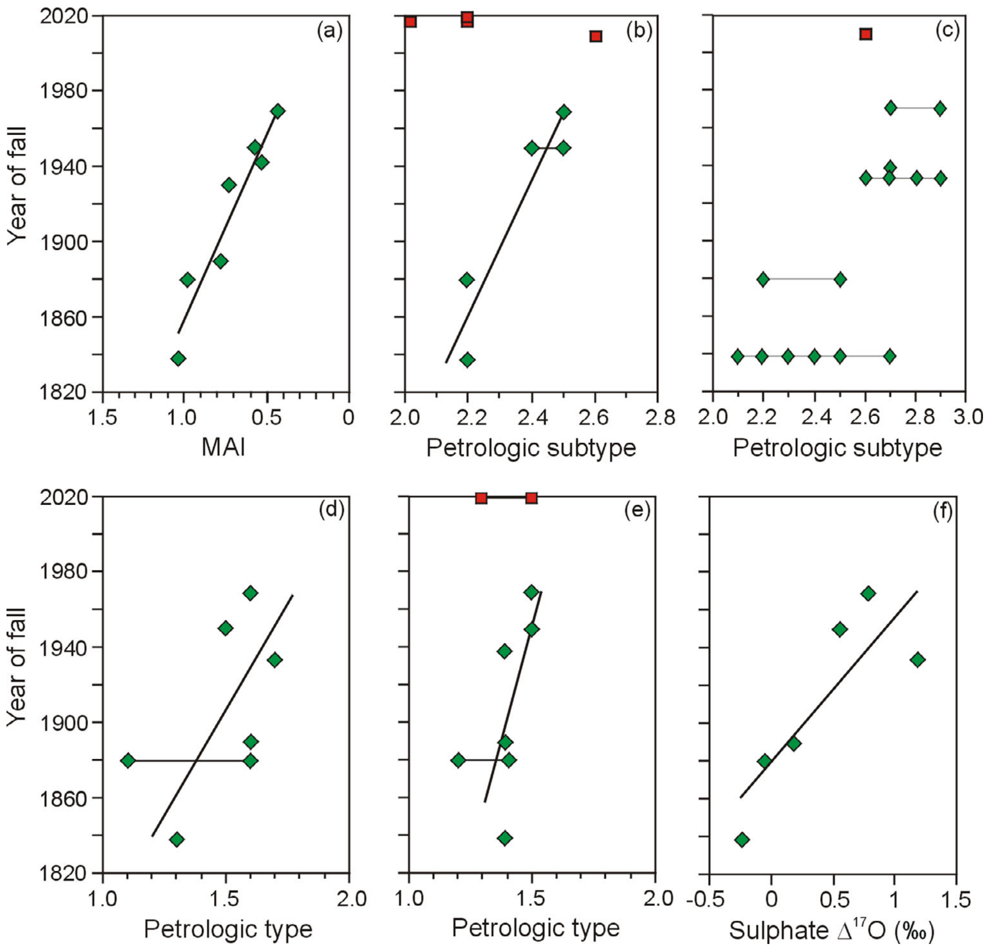

Fig. 6. The year of fall of CMs plotted against measures of their degree of aqueous alteration. The pre-1970 falls are plotted as green diamonds, and post-1970 as red squares. The regression lines and R^2 values refer only to the pre-1970 falls. All data are listed in Table 1. a) Year of fall versus the MAI of Browning et al. (1996; MAI); $R^2 = 0.91$, $n = 7$. b) Year of fall versus petrologic subtype of Rubin et al. (2007); $R^2 = 0.93$, $n = 4$. The upper and lower limits in classification of Murray are joined by a line, and the average was used in fitting the regression line. Data for the pre-1970 samples are from Rubin et al. (2007) whereas petrologic subtypes of the four post-1970 samples are as follows: Maribo (Haack et al. 2012), Shidian (Fan et al. 2020), Mukundpura (Rudraswami et al. 2019), Aguas Zarcas (Martin and Lee 2020). c) Year of fall versus petrologic subtype determined by Lentfort et al. (2020). The subtypes of all lithologies identified in each meteorite are joined by a line. Owing to the wide range in subtypes of each meteorite, a regression line has not been plotted. d) Year of fall versus petrologic type using the scheme of Alexander et al. (2013); $R^2 = 0.51$, $n = 6$. Two analyses of Nogoya are joined by a line and the average was used in plotting the regression line. e) Year of fall versus petrologic type using the scheme of Howard et al. (2015); $R^2 = 0.47$, $n = 6$. Two analyses of Nogoya are joined by a line and the average was used in plotting the regression line. The range of petrologic types of Aguas Zarcas (Davidson et al. 2020) is shown by two red squares joined by a line. f) Year of fall versus the oxygen isotopic composition of water-soluble sulfate (data from Airieau et al. 2005); $R^2 = 0.70$, $n = 6$. (Color figure can be viewed at wileyonlinelibrary.com.)

properties (Fig. 6). The correlation between terrestrial age and MAI is based on pre-1970 falls, and so in the following discussion, we differentiate between falls recovered before and after 1970. Of the four pre-1970 falls that were classified by Rubin et al. (2007), the older two, Cold Bokkeveld and Nogoya, have a lower petrologic subtype than the younger two, Murray and Murchison ($R^2 = 0.93$, $n = 4$; Fig. 6b). Four of the five pre-1970 falls analyzed by Lentfort et al. (2020) contain clasts of different petrologic subtypes. All but one of the clasts in the two older falls is of a lower petrologic subtype than in three younger pre-1970 falls and their constituent clasts (Fig. 6c). The petrologic types of the six pre-1970 falls that were classified by Alexander et al. (2013) and the six that have been classified using the scheme of Howard et al. (2015) generally decrease with increasing age ($R^2 = 0.51$ and 0.47, respectively; Figs. 6d and 6e). The oxygen isotopic composition of pre-terrestrial sulfate correlates well with the terrestrial ages of pre-1970 falls ($R^2 = 0.70$, $n = 6$; Fig. 6f). In summary, a variety of criteria show that the degree of aqueous alteration of CM falls generally increases with terrestrial age, although the recent work by Lentfort et al. (2020) suggests that results from the other classification schemes should be viewed with caution as the samples analyzed may have contained one or more clasts that had been aqueously altered to different extents. Nonetheless, the age/alteration correlations in Fig. 6 could be interpreted as consistent with alteration of pre-1970 falls having taken place both within their parent body and during curation.

Six CM falls have been recovered since 1970 and so can be used to test the pre-1970 age/alteration correlation. If there is a causal link between terrestrial age and degree of aqueous alteration, the post-1970 falls should have a higher petrologic (sub)type than Murray and Murchison. Furthermore, CM finds, most or all of which will have fallen prior to 1838, should be more altered than Nogoya and Cold Bokkeveld, especially given that most of them will have been exposed to environmental conditions harsher than those in curatorial settings. The petrologic (sub)types of four of the post-1970 falls have been measured or estimated (Table 1, Fig. 6). Three of them are more altered than Murray and Murchison and so are inconsistent with an extrapolation of the pre-1970 age/alteration correlation. Furthermore, petrographic descriptions of the other two post-1970 falls whose (sub)type has not been classified, Sayama (Takaoka et al. 2001; Noguchi et al. 2002) and Kolang (Meteoritical Bulletin), suggest that both are highly altered. The petrologic (sub)types of CM finds are also inconsistent with an extrapolation of the age/alteration correlation because they vary over a wide range (Fig. 1) and include some of the least altered samples yet described, such as Yamato 791198 (Howard et al. 2015), Paris (Rubin 2015), and Lewis Cliff 85311 (Lee et al. 2019a).

The absence of a systematic age/alteration relationship shows that the petrographic, mineralogical, and chemical properties of CM falls have not been measurably overprinted by terrestrial processes. It is nonetheless conceivable that MAI contains a terrestrial signal because it quantifies relatively subtle changes in mineral chemistry (e.g., Fe valence) and correlates very well terrestrial age (Fig. 6a). The most likely explanation for the age/alteration correlation of the pre-1970 falls is a sampling bias because each of the classification schemes has used a fairly small proportion of the 13 meteorites, and all of them include Cold Bokkeveld, Nogoya, and Murchison. Nonetheless, it would be worthwhile to determine the MAI and petrologic (sub)type of the unclassified pre-1970 CM falls, with care taken to identify and characterize any clasts. If these falls plot on the age/alteration line, a reexamination of the potential for a terrestrial overprint would be justified.

IMPLICATIONS FOR COLLECTION AND CURATION

The CMs are contaminated rapidly once they are exposed to the Earth's atmosphere so that prompt recovery and curation of falls is vital. As shown by Sutter's Mill, exposure to liquid water is particularly damaging and can significantly alter the chemical composition and mineralogy of carbonaceous chondrites (Jenniskens et al. 2012). Identification of any contaminant organic matter can be assisted by the collection of air, soil, and vegetation samples from the recovery site and at the same time as the meteorite. The emphasis of subsequent storage and curation should be on isolating the sample from the atmosphere, and stopping or slowing any reactions that may have started. Samples that are curated at the Johnson Space Center, including those returned by the Apollo missions and meteorites collected from Antarctica by ANSMET, are stored in a nitrogen atmosphere (Allen et al. 2011). With regard to the Apollo samples, the amount of water vapor and oxygen in the nitrogen is regulated at ~5 ppm. For volatile-rich meteorites such as the carbonaceous chondrites, Herd et al. (2016) suggest storage in argon, and at <10°C. While curation in an inert atmosphere would be desirable for all carbonaceous chondrite samples irrespective of their terrestrial history, there is evidence that once they have been terrestrially contaminated, alteration can continue even under controlled conditions. For example, King et al. (2020) found that terrestrial evaporites were abundant in the CI carbonaceous chondrite Ivuna despite being stored in a nitrogen atmosphere for at least 10 years prior to analysis. Previous work on ordinary chondrites has shown that Fe metal is significantly altered when stored under ambient conditions over decadal timescales (e.g., Lee and Bland 2004). While these observations show that falls are susceptible to alteration, experiments would provide a clearer understanding of the mechanisms and rates of alteration during brief subaerial exposure and subsequent curation. Given that gypsum and iron oxide could have formed on Murchison in less than 4 years (Fuchs et al. 1973), reaction rates will be sufficiently rapid that alteration effects should be readily detectable within months.

SUMMARY AND CONCLUSIONS

Nineteen CM falls were recovered between 1838 and 2020 (21 including Diepenveen and Sutter's Mill), and the distribution of fall dates hints at two temporal clusters. These meteorites are not pristine samples of C-complex asteroids, and a chronology of terrestrial alteration can be recognized:

- Within days of fall carbonaceous chondrites can be contaminated by organic compounds including amino acids from water, soil, and vegetation. Water vapor is readily adsorbed from the atmosphere, and stones are contaminated by airborne hydrocarbons. Exposure to liquid water before recovery can lead to leaching of fluid-mobile elements, and dissolution of minerals such as oldhamite.
- Within a few years, the falls can be microbially colonized, and organic compounds can penetrate into meteorite interiors. Fe-hydroxides may start to form by the hydrolysis and oxidation of Fe,Ni metal, and gypsum by the dissolution–reprecipitation of indigenous S-rich phases.
- Formation of Fe-oxyhydroxides from Fe,Ni metal will continue slowly over decadal timescales.

There is no evidence that the mineralogical, chemical, and isotopic effects of parent body aqueous alteration can be terrestrially overprinted to a sufficient extent to lower the petrologic (sub)type of a CM fall. However, fragments of some of the CMs that are regarded as exemplars of the group (i.e., Cold Bokkeveld, Murray, and Murchison) could have been exposed to liquid water prior to recovery. By analogy with Tagish Lake and Sutter's Mill, the mineralogy and chemical composition of these falls could have been significantly modified and these potential effects should be investigated. Our findings show that in order to confidently interpret analyses of samples returned from the asteroids from Ryugu and Bennu, their exposure to the terrestrial atmosphere must be minimized. In the event of exposure, the provenance of organic compounds, and inorganic phases including Fe-oxyhydroxides and sulfates, should be interpreted with care.

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