

Weathering of Chondritic Meteorites

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Meteorites are frequently not the pristine record of early solar system processes that we would like them to be. Many unique meteorites, samples that can further our understanding of the environment of the early solar system, are finds, so a knowledge of terrestrial weathering becomes a requirement to accurately interpret their preterrestrial history. This chapter outlines the weathering process in chondrites, the mineralogical changes that occur in response to weathering, the variations in weathering in different environments, the effect of weathering on O isotopes and chemistry, and how weathering varies between different chondrite groups. As the most abundant meteorite group, most studies of meteorite weathering have concentrated on ordinary chondrites, although weathering affects all classes of meteorites. In many cases, these data are relevant to other chondrite groups, as well as irons and achondrites, but there are a number of specific issues related to carbonaceous chondrites (degradation of organic C, mobilization of S, reordering of phyllosilicates) that require a separate discussion. In particular, carbonaceous chondrites appear to be uniquely susceptible to terrestrial weathering, and recent studies have shown a number of alteration effects, even in falls curated in museum collections. The terrestrial alteration of carbonaceous chondrites concludes our chapter.

1. INTRODUCTION

Meteorite weathering can be regarded as the alteration of original component phases of the meteorite to phases that are more stable at Earth's surface. On entering Earth's atmosphere, interaction with the terrestrial environment begins. Understanding the nature of this interaction is extremely important, given that modern meteoritics involves the analysis of a much higher proportion of finds, following the recovery of large populations of samples from desert accumulations, than was the norm a few decades ago. Knowing the details of the terrestrial weathering process allows those effects to be deconvolved from any asteroidal alteration that may be present (e.g., *Abreu and Brearley, 2005*). In addition, recent studies have observed a number of parallels between weathering of chondrites in Antarctica and aqueous alteration on asteroids (e.g., *Lee and Bland, 2004; Sephton et al., 2004*). There are parallels, both in the effect of alteration on organic material and in the nature of the weathering products produced. It may be that the Antarctic environment can provide an effective analog in helping us to understand asteroidal alteration.

Terrestrial weathering of a meteorite is often viewed as an obstacle to interpreting the preterrestrial history of a sample, but meteorites have value to terrestrial researchers involved in low-temperature aqueous alteration studies in that they provide a "standard sample" for tracking the effects of rock weathering. Meteorites are unique among geological materials in that they show relatively minor intragroup variations and their terrestrial ages can be established by measuring the decay of cosmogenic radionuclides by accelerator mass spectrometry. They may therefore be a potential "chronometer" of environmental conditions during their terrestrial residency, given the proper analytical approach. The processes and rates of this weathering depend on a similar group of factors to those that control the alteration of terrestrial rocks, namely the chemistry of weathering fluids and the nature of reactions at mineral and alloy surfaces. The microenvironmental conditions within a rock that regulate the rates of chemical reactions are frequently influenced by climate and include such factors as the pH and f_{O_2} of pore- and groundwaters, temperature, drainage, and microbial activity. Meteorites have been recovered from all over the world and therefore have been exposed to all types of

climatic regimes. Other factors that control the response to weathering include the composition and crystal structure of the silicate or alloy, the nature of secondary oxidation products, the degree of porosity or fracturing, and the exposure time of the sample (e.g., Gooding, 1981; Velbel *et al.*, 1991).

1.1. Falls, Finds, and Meteorite Accumulations

Prior to 1969 the total number of known meteorites was around 2100 (Grady, 2000). Many of these had been observed as fireballs as they entered Earth's atmosphere, and were then recovered later ("falls"), but around 60% were chance discoveries or "finds." Currently, meteorite samples available to researchers number in the tens of thousands, following extensive collecting programs in the Antarctic and in hot desert regions such as the Sahara and the Nullarbor of Australia. With only 1000 falls in modern collections (Grady, 2000), the ratio of falls to finds has changed dramatically since 1969. Preservation of meteorites depends mainly on climate. In temperate and tropical zones weathering is relatively rapid. However, in persistently dry climates chemical weathering is less vigorous, and this process might be slowed down to the point where meteorites begin to accumulate (Bland *et al.*, 1996a, 1998a). Meteorites may be preserved for thousands, tens of thousands, or, as in Antarctica, millions of years after their fall.

The cold desert of Antarctica was the first place where concentrations of meteorites were recognized (Yoshida *et al.*, 1971). In Antarctica, meteorites survive for longer owing to relatively constant environmental conditions and low chemical weathering, and are physically concentrated by processes of ice movement and ablation. As ice moves from the center of the continent to the coast it encounters obstacles in the form of mountain ranges. While most of the ice flows around the barriers, some remains in front of the rock and is only removed gradually by wind ablation. Any meteorites that may have fallen into the ice upstream of this point will thus gradually be exposed, forming a concentration on this stranding surface (Cassidy *et al.*, 1992). In addition to Antarctica, a number of the world's other arid and semi-arid regions have proved to be prolific sources of meteorites. Large numbers of meteorites have been recovered from "bad lands" subject to wind deflation in Roosevelt County, New Mexico (Scott *et al.*, 1986; Sipiara *et al.*, 1987; Zolensky *et al.*, 1990), and many more meteorites have also been found in the stony deserts of North Africa (Wlotzka, 1989; Jull *et al.*, 1990; Schlüter *et al.*, 2002). More than 3.8 million km² of Australia is arid or semi-arid land that has also provided conditions suitable for the prolonged preservation of meteorites (Bevan, 1992a,b).

1.2. Differences Between Antarctic and Non-Antarctic Populations

Terrestrial age dating of meteorites by cosmogenic radionuclides has revealed that there are large differences in the residence time between Antarctic and hot-desert meteorites. Meteorites from Antarctica show a mean residence

time of around 10⁵ yr (Nishiizumi *et al.*, 1989; Nishiizumi, 1995), while hot desert meteorites have much younger terrestrial ages, typically between 15,000 and 20,000 yr (Jull *et al.*, 1993a, 1995; Wlotzka *et al.*, 1995). This raises the possibility that the two meteorite populations might sample different asteroidal parent bodies, since it has been suggested (Wetherill, 1986) that "meteorite streams" may be the source of many of Earth's more common meteorites, and that these streams may vary over the timescale of 10⁵–10⁶ yr (Greenberg and Chapman, 1983). Dennison *et al.* (1986) and Dennison and Lipschutz (1987) observed differences in the trace-element composition between Antarctic and non-Antarctic H5 ordinary chondrite populations, and suggested a temporal variation in the source regions from which the meteoroids derive as a cause. Benoit and Sears (1992) also observed differences in the metallographic cooling rate between Antarctic and non-Antarctic H5 chondrites. Nonetheless, while it appears likely that there may be generic differences between Antarctic and non-Antarctic meteorite populations, it is clear that many of the variations are a function of climate and its effect on the weathering of meteorites (Buchwald, 1989): A better understanding of terrestrial weathering will help in identifying generic differences between meteorite populations and make the identification of extraterrestrial alteration simpler.

1.3. Weathering Scales

In an attempt to quantify the degree of alteration that a sample has experienced, a number of qualitative weathering indices have been applied to cold- and hot-desert populations. The Meteorite Working Group at the NASA Johnson Space Center in Houston uses weathering categories "A," "B," and "C" to denote minor, moderate, and severe rustiness of Antarctic finds, and have a similar scale for degree of fracturing. A scale of weathering effects seen in polished thin sections of meteorites has been proposed by Jull *et al.* (1991) and updated by Wlotzka (1993), Wlotzka *et al.* (1995), and Al-Kathiri *et al.* (2005) for meteorites from hot deserts. In this system six categories of weathering are recognized, beginning with minor to complete oxidation of metal and then troilite (categories W1–W4) and continuing with at first minor (W5) and then massive (W6) alteration of mafic silicates. Bland *et al.* (1996a,b, 1998a, 2000a) discussed a quantitative index of alteration for ordinary chondrites, using ⁵⁷Fe Mössbauer spectroscopy to determine the relative abundance of Fe³⁺ in a sample (unweathered ordinary chondrite falls contain negligible Fe³⁺).

2. PROCESSES AND EFFECTS OF WEATHERING

2.1. Alteration of Ferromagnesian Silicates

Determining the differences in alteration rate between phases in a meteorite is a rather difficult analytical problem. Mössbauer spectroscopy is a useful tool in this regard. Observing how the spectral area of primary phases varies

with the total amount of oxidation allows us to determine which phases are most susceptible to weathering: An increase in a ferric component should be accompanied by a concomitant decrease in an absorption associated with a primary mineral. *Bland et al.* (1998a) compared the spectral area of “opaque” phases (Fe-Ni and troilite) and ferromagnesian silicates to total oxidation, and observed a decrease in primary phases with increasing oxidation, suggesting that all Fe-containing minerals within the meteorite are affected by weathering to some degree. A similar observation was made by *Burns et al.* (1995) on weathered Antarctic meteorites. Interestingly, although ferromagnesian silicates appear to be weathered at a constant rate, there was no detectable difference in the rate of weathering between olivine and pyroxene. Although a similar feature has been observed in studies of weathered Antarctic meteorites (*Fisher and Burns, 1992*), its interpretation remains problematic given the well-documented susceptibility of olivine to dissolution in experimental weathering studies (*Luce et al., 1972; Siever and Woodford, 1979; Grandstaff, 1978*). One explanation relates to how the O-isotopic composition of meteorites varies with weathering (discussed in more detail below). *Bland et al.* (2000b) found evidence for topotactic alteration during the incipient weathering of olivine. *Banfield et al.* (1990, 1991) note that olivine weathered in this manner is comparatively resistant to subsequent oxidation: a possible explanation for the similarity between olivine and pyroxene weathering rates in ordinary chondrites.

2.2. Rusting of Meteorites and Corrosion Products

In common with the iron meteorites, the Fe-Ni metal, troilite, and mafic silicates — the main components in ordinary chondrites — render these meteorites susceptible to rusting; ferruginous oxidation products are the most obvious weathering products on meteorites. *Gooding* (1986) recognized two forms of rust: “metallic” (Fe-Ni-S) rust, formed by weathering of primary Fe-Ni metal and troilite, and “sialic” (Fe-Si-Al) rust, which forms by the weathering of mafic silicates. In a model based on the variable ability of Fe-rich minerals in meteorites to nucleate water to ice (and so retard weathering), *Gooding* (1986) found that troilite is less susceptible to weathering than Fe-Ni metal (composed of the two polymorphs taenite and kamacite), taenite is less susceptible than kamacite, and silicates are less susceptible than sulfides and metal. Using scanning electron microscope, electron microprobe, and X-ray powder diffraction, akaganéite [β -FeOOH], goethite [α -FeOOH], lepidocrocite [γ -FeOOH], and maghemite [γ -Fe₂O₃] were identified by *Buchwald and Clarke* (1989) as constituting the bulk of the corrosion products in a suite of Antarctic meteorites. Magnetite [Fe₃O₄] was also observed by *Buchwald* (1989). *Marvin* (1963), using X-ray powder photographs, recognized akaganéite as a corrosion product of iron meteorites along with goethite and magnetite. Hematite was found in very small quantities in only one meteorite. Other studies have also tentatively identified jarosite [KFe₃(SO₄)₂(OH)₆] (*Gooding, 1981, 1986; Zolensky and Gooding, 1986*) as another

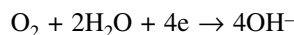
component of “metallic” rust. Metallic rust commonly occurs as mantles on Fe-Ni metal or as veins, and may act as a pore-filling cement in a similar way to diagenetic cements in sedimentary rocks (*Gooding, 1986*). The importance of Mössbauer spectroscopy as a technique in characterizing these types of mineral assemblages is evident. The presence of this suite of oxides and oxyhydroxides was confirmed in a ⁵⁷Fe Mössbauer spectroscopy study of weathered ordinary chondrites by *Bland et al.* (1998a). Only jarosite was not observed.

In an extensive study of weathering in Antarctic meteorites, *Buchwald and Clarke* (1989) found that akaganéite was the mineralogical key to an understanding of the corrosion of meteoritic metal. It has also been shown to be important in the weathering of a modern fall: Tsarev, an L5 chondrite that fell in Russia in 1922 (*Honda et al., 1993*). β -FeOOH (akaganéite) was described in detail by *Mackay* (1960, 1962); *Waychunas* (1991) describes the mineral as having a tunnel structure similar to hollandite [KAlSi₃O₈], with a variable amount of a large anion (Cl⁻ is common) occupying the alkali site. Evidence suggests that akaganéite is not stable without this extra species (*Feitknecht et al., 1973; Chambaere and DeGrave, 1984*), and as such a new and more detailed formula for akaganéite may be written [Fe₁₅Ni][O₁₂(OH)₂₀]Cl₂(OH) (*Buchwald and Clarke, 1989*), which is a variation of that proposed by *Keller* (1970).

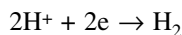
Buchwald and Clarke (1989) postulated a process of corrosion in meteorites involving anodic metal going into solution and Cl⁻ ions from the terrestrial environment moving to the reaction surface to maintain a charge balance, Cl-containing akaganéite then precipitating at the reaction surface. Over time, these initial corrosion products decompose to intimate intergrowths of goethite and maghemite, releasing Cl⁻ for further corrosive action. As suggested in this model, the Cl content is found to be highest at the interface with the metal. In temperate zones, Cl could be introduced from soil groundwater; however, in Antarctica the source is probably sea-spray, carried inland as an aerosol or gas (see below). The transition from kamacite to akaganéite appears to take place without significant volume change (the silicates remain undisturbed). The Ni content of the oxide reflects the composition of the phase from which it formed, so Fe/Ni ratios of around 16 result when akaganéite is formed from kamacite, and about 4 when formed from taenite. In a further study involving Antarctic and non-Antarctic meteorites *Buchwald* (1989) noted that in cold, dry environments (<20% relative humidity) akaganéite is stable and long lived, while in meteorites from temperate climates, akaganéite is only seen in the active corrosion zone, most of it having decomposed to the oxides goethite, maghemite, and magnetite. Using conventional half-reactions, the corrosion mechanism proposed by *Buchwald and Clarke* (1989) may be expressed as follows. Fe-Ni metal (Fe⁰) is oxidized at the anode



and O₂ is reduced at the cathode



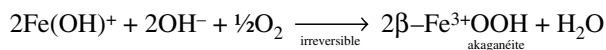
or alternatively, H is reduced at the cathode



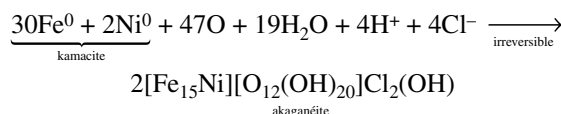
Combining the first two equations



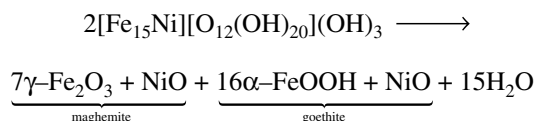
This is followed under oxidizing conditions by the formation of akaganéite



Using a modified version of the formula proposed by Keller (1970), Buchwald and Clarke (1989) express the reaction as



This reaction occurs slowly in the Antarctic environment, but more rapidly in temperate climates. Cl⁻ in akaganéite exchanges with OH⁻, and Cl⁻ is released into solution to move again to the corrosion front or to be flushed from the system. The akaganéite left behind will approach the composition [Fe₁₅Ni][O₁₂(OH)₂₀](OH)₃ and, having lost its Cl, becomes unstable. Under conditions of modest heating and/or seasonal changes in temperature and humidity, it decomposes according to a reaction such as



The recognition of a similar suite of corrosion products, including akaganéite, in meteorites recovered from a variety of climatic regimes (Bland *et al.*, 1998a) indicates that this corrosion mechanism may well be acting in the oxidation of all meteoritic metal. This is also substantiated by the presence of the mixed valence ironhydroxychloride, Fe₂(OH)₃Cl, in a number of meteorites from temperate climates (Buchwald, 1989), a mineral that may represent the initial step in the corrosion process. More recently, the suggestion that akaganéite requires Cl for stability has been questioned by Rézel and Génin (1990). These workers used Mössbauer spectroscopy and X-ray analysis to demonstrate that akaganéite retained its crystal structure following substitution of Cl⁻ ions by OH⁻. In addition, Bland *et al.* (1997) have recorded akaganéite in a meteorite weathered in the absence of Cl⁻. But whether or not akaganéite requires Cl⁻ for stability, the ubiquitous nature of Cl in the environment means that natural occurrences of akaganéite contain Cl.

While the formation of the majority of iron oxides and oxyhydroxides found in ordinary chondrites may be explained by the process detailed above, the presence of magnetite as a widespread corrosion product in meteorites from

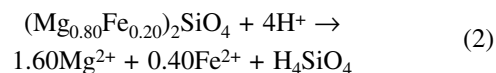
temperate climates (Buchwald, 1989; Bland *et al.*, 1998a,b; Verma and Tripathi, 2004) suggests that an additional mechanism may be involved. The correlation between spectral areas associated with Fe-Ni metal and magnetite in Mössbauer spectra from an artificially weathered sample and from meteorites recovered from hot and cold desert regions (Bland *et al.*, 1997, 1998a) indicate that magnetite is formed as a direct result of the dissolution of Fe-Ni metal. A possible explanation for the stability of a mixed Fe²⁺-Fe³⁺ oxide in this environment may be that in a situation of rapid dissolution and oxidation of Fe-Ni metal, porewaters around metal grains become saturated with Fe²⁺ ions. While some proportions are oxidized to Fe³⁺, others remain in solution, the combination of both species allowing the formation of magnetite. A similar process was suggested by Faust *et al.* (1973) to explain oxidation observed in the Wolf Creek meteorite.

Although Buchwald and Clarke (1989) only examined the corrosion of meteoritic metal, similar reactions occur for the dissolution of other Fe-containing meteoritic minerals

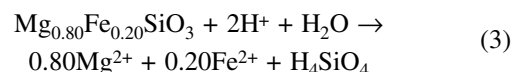
(i) troilite (FeS):



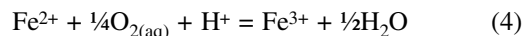
(ii) olivine (Fo_{0.80}Fe_{0.20}):



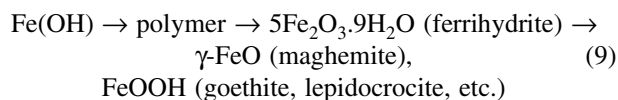
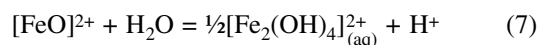
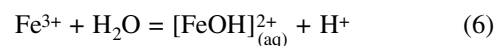
(iii) pyroxene (En_{0.80}Fs_{0.20}):



Once dissolution of Fe²⁺ ions from primary meteoritic minerals has occurred, oxidation of dissolved Fe²⁺ ions and hydrolysis of Fe³⁺ ions takes place



where the overall hydrolysis reaction (equation (5)) involves intermediate steps, including



In the above reactions, Mg²⁺ and other nonferric cations may be incorporated in carbonates, as traces in some iron oxides/oxyhydroxides, in clay minerals, or removed from the system entirely.

Ferrihydrite is the mineral name identifying poorly crystalline to amorphous hydrous iron oxides with chemical composition near $5\text{Fe}_2\text{O}_3 \cdot 9\text{H}_2\text{O}$. It is formed abundantly as the initial product of the hydrolysis and precipitation of dissolved Fe in a variety of geological systems (Waychunas, 1991). It is likely that this is also true for the oxidation of meteorites. In ^{57}Fe Mössbauer spectra of a sample of ALlegan weathered in the laboratory, Bland et al. (1997) observed ferrihydrite as one of the suite of corrosion products that formed. The artificially accelerated oxidation and dissolution of this sample makes it likely that a poorly crystalline, amorphous iron oxide such as ferrihydrite would be the initial weathering product.

The weathering of ordinary chondrite meteorites therefore appears to involve a combination of a number of processes. The initial product of the weathering of Fe-Ni metal is akaganéite (or magnetite where dissolution is exceptionally rapid). For other primary meteoritic minerals (troilite, olivine, and pyroxene), ferrihydrite may be the first iron oxide mineral following the dissolution of these phases. Over time, the initial corrosion products undergo an aging process that converts them to goethite, maghemite, and lepidocrocite. Magnetite may remain as a metastable reaction product.

2.3. Differences in Weathering Between Cold and Hot Deserts

In Mössbauer studies of weathered meteorites, two classes of alteration phases are readily distinguished: paramagnetic weathering minerals, which include akaganéite, small-particle goethite, lepidocrocite, and phyllosilicates; and magnetically ordered phases, which include magnetite and maghemite. Bland et al. (1998a) observed that the ratio of paramagnetic to magnetically ordered ferric species varies widely in weathered ordinary chondrites, with a high abundance of magnetically ordered phases typically associated with more humid environments. It was noted that magnetite was extremely uncommon in Antarctic samples.

In a TEM study of Antarctic and hot desert finds, Lee and Bland (2004) observed cronstedtite, a Fe-rich phyllosilicate common in CM chondrites in the heavily weathered Antarctic finds ALHA 78045 and 77002. This mineral is rare on Earth, and was not observed in hot desert samples. An unidentified hydrous Si-Fe-Ni-Mg mineral or gel had also partially replaced taenite in ALHA 78045. In addition to Fe-rich weathering products, “hot” desert meteorites contain sulfates, Ca-carbonate, and silica, minerals that were largely absent from Antarctic finds. As preterrestrial cronstedtite is abundant in CM2 carbonaceous chondrites, the Antarctic environment may be a powerful analog for aqueous alteration in the asteroidal parent bodies of primitive meteorites.

2.4. Formation of Evaporites and Carbon Content

Weathering of meteorites involves not only the formation of rust and oxidation of metal but also hydrolysis of silicates, hydration, carbonation, and solution. Clay mineraloids have been identified compositionally (Gooding, 1986, 1981)

but are often insufficiently crystalline to allow identification by XRD (Velbel and Gooding, 1988). An integral part of the process of meteorite weathering is the production of a variety of terrestrial evaporitic carbonates and sulfates (Velbel et al., 1991; Jull et al., 1988; Grady et al., 1989a). Miyamoto (1989, 1991) used infrared spectroscopy to analyze a suite of ordinary chondrites including modern falls and Antarctic and non-Antarctic finds, and found that hydrated carbonates were ubiquitous in Antarctic samples and common in non-Antarctic meteorites. X-ray diffraction studies of Antarctic chondrites (Marvin, 1980; Gooding et al., 1988) have shown that the specific mineral constituents of evaporitic carbonates and sulfates include nesquehonite [$\text{Mg}(\text{HCO}_3)(\text{OH}) \cdot 2\text{H}_2\text{O}$], hydromagnesite [$\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 4\text{H}_2\text{O}$], starkeyite [$\text{MgSO}_4 \cdot 4\text{H}_2\text{O}$], epsomite [$\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$], and gypsum [$\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$]. Measurements of $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ from nesquehonite in one Antarctic H5 chondrite indicate that it formed at near 0°C by reaction with terrestrial water and CO_2 (Grady et al., 1989a). Carbon-14 dating of this phase showed that it formed in the last 40 years (Jull et al., 1988). An analysis of the stable-isotopic ratios in Antarctic ordinary chondrites (Grady et al., 1989b; Socki et al., 1991) has confirmed that carbonates originate from a reaction between atmospheric CO_2 and the meteorite, while the source of the cations in evaporites (Na, Mg, K, Ca, Rb) is probably meteoritic (Velbel et al., 1991). The primary mineral likely to react to produce a hydrous magnesium carbonate would be olivine (Velbel et al., 1991) via decomposition with water and CO_2 .

Total C content, and macromolecular C, may also be affected by weathering. Studies of the Holbrook meteorite, which fell in 1915 in Arizona and has been collected at various times over a period of 60 years, have shown that weathering of the meteorite has been accompanied by an increase in the C content (Gibson and Bogard, 1978). A study of the unequilibrated ordinary chondrite, Roosevelt County 075, has shown that this meteorite has a higher C content than any other bulk ordinary chondrite (Ash and Pillinger, 1992a). Stepped combustion experiments suggest that about 50% of the C is from terrestrial organic material (Ash and Pillinger, 1992a). Further work has shown some correlation between the degree of C contamination and terrestrial residence time in other meteorites from Roosevelt County (Ash and Pillinger, 1993).

2.5. Oxygen Isotopes and Weathering

Although the oxidative interaction of terrestrial porewaters with extraterrestrial meteoritic minerals must necessarily involve the incorporation of terrestrial O into the stable crystal lattices of secondary alteration minerals within the meteorite, very little has been done to systematically quantify the effect of weathering on the bulk O-isotopic composition of meteorites. Clayton et al. (1984) recognized that a major drawback in an O-isotopic study of Antarctic meteorite finds is the possibility of contamination by O derived from the polar ice. In the case of ice from the Allan Hills region, from which a large number of meteorites have been recovered, Fireman and Norris (1982) found a $\delta^{18}\text{O}$ of

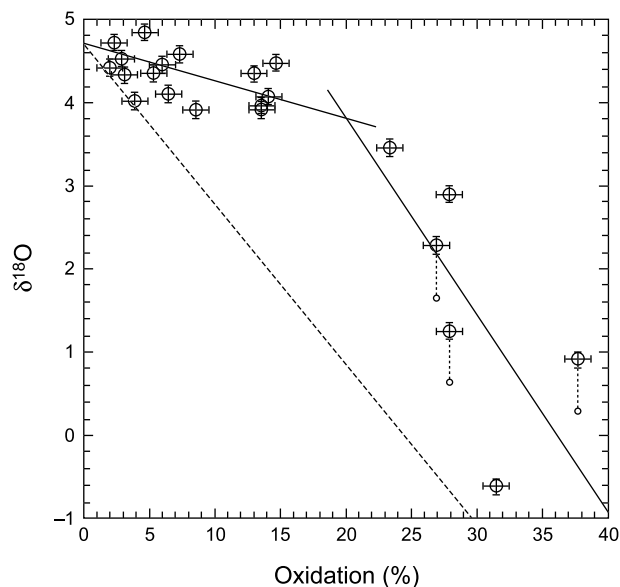


Fig. 1. Ferric oxidation (percentage of total iron now present as ferric iron, derived from ^{57}Fe Mössbauer spectroscopy analyses) plotted against $\delta^{18}\text{O}$ in L-chondrite finds from Antarctica. The dashed line shows the expected trend; solid lines are modeled trends assuming surface-mediated alteration initially and solution-mediated alteration after 20% oxidation.

about -40% relative to SMOW, i.e., even a small contamination should lead to a large displacement of the isotopic composition of the meteorite to lower $\delta^{18}\text{O}$ values (Clayton *et al.*, 1984). What is surprising is that significant amounts of alteration are required before any isotopic effect is observed. Bland *et al.* (2000b) studied how O isotopes varied in L-group ordinary chondrites weathered in Antarctica, using Mössbauer spectroscopy to quantify the degree weathering. The data indicate that alteration is a two-stage process, with an initial phase producing only a negligible isotopic effect. L-group falls contain no Fe^{3+} detectable using Mössbauer spectroscopy. Even though these meteorites begin with a $\delta^{18}\text{O}$ value of 4.7% (Clayton *et al.*, 1991), and are being altered by water at a $\delta^{18}\text{O}$ of -40% , there is no discernible shift in O away from fall values until $>25\%$ of the Fe in the meteorite has been converted to Fe^{3+} (Fig. 1). Although surprising, an explanation may be found in the alteration of terrestrial silicates. Numerous studies report pervasive development of channels a few to a few tens of nanometers wide in the incipient alteration of silicates (Eggleton, 1984; Smith *et al.*, 1987; Banfield *et al.*, 1990; Hochella and Banfield, 1995). A similar texture is observed in the samples studied by Bland *et al.* (2000b). Alteration involves a restructuring to clay minerals along these narrow channels, in which access of water is restricted. The clay shows a topotactic relationship to the primary grain, suggesting either epitaxial growth of the clay using the silicate as a substrate, or inheritance of the original O structure by the clay. The data presented by Bland *et al.* (2000b) indicate the latter: With extensive inheritance of structural polymers by the

weathering product, the bulk O-isotopic composition is comparatively unaffected. This work shows that the relationship between O isotopes and terrestrial alteration is not straightforward. It has significance in the area of asteroidal aqueous alteration in that substantial modification of chondritic materials may occur without a pronounced isotopic effect.

2.6. Elemental Mobilization During Weathering

The literature on this topic is sparse, although studies involving both ordinary chondrites and carbonaceous chondrites have been carried out. A discussion of mobilization of aqueous species during carbonaceous chondrite weathering is included at the end of this chapter.

In the case of ordinary chondrites, Gibson and Bogard (1978) observed a decrease in SiO_2 and MgO contents, and no change in the elemental concentrations for Ti, Ca, Al, P, and Mn, in the weathering of Holbrook. Bland *et al.* (1998a) also observed a decrease in Si and Mg (and minor decreases in Na and Ca) with increasing weathering. No significant changes were observed in elemental concentrations for Fe, Ti, Al, Mn, K, or P. These data are consistent with the dissolution products of ferromagnesian silicates (Si and Mg) being flushed from the system as weathering proceeds, while Fe is bound in hydrated ferric oxides, a scenario that is similar to that proposed by Velbel *et al.* (1991) for the mineral alteration and element mobilization involved in the growth of Mg-carbonates on Antarctic meteorites. As noted above, Velbel *et al.* (1991) found that Mg, Na, K, Ca, and Rb were mobilized from within an Antarctic H5 chondrite to form evaporate efflorescence on the surface. More recently, Al-Kathiri *et al.* (2005) have observed depletion in S in meteorites weathered in Oman, and also mobilization of Ni and Co, related to breakdown of Fe-Ni metal. Meteorites were commonly found to be enriched in Sr and Ba, both highly mobile trace elements. In Antarctica, the action of circulating waters in meteorites stranded on the ice has also produced elemental enrichments. Delisle *et al.* (1989) measured an enrichment of U in some meteorites from Frontier Mountain, Antarctica, on the order of $300\times$ the normal value, a feature that was also noted by Welten and Nishiizumi (2000). The suggestion is that U salts have precipitated from solution in the meteorite after being leached from nearby granite.

2.7. Iodine Contamination

Although they have been stored in one of the most sterile terrestrial environments, I overabundances appear to be common in both Antarctic meteorites and rocks (Dreibus and Wänke, 1983; Heumann *et al.*, 1987; Ebihara *et al.*, 1989; Langenauer and Krähenbühl, 1992). Iodine is also seen to vary with depth in sample: Ebihara *et al.* (1989) found 11.2 ppm I in the outermost portion of an L6 chondrite, decreasing to 0.5 ppb at the center, concluding that the I overabundance was a result of interaction of atmospheric I compounds originating from the ocean with the surfaces of Antarctic rocks and meteorites. They postulated that

the majority of the sea salt found in meteorites is emitted as small particles by sea spray deposited on the Antarctic ice close to the ice edge. Longer-distance transport may involve oxidation of iodide at the surface of the ocean to elemental I that exists as a gas before being precipitated in snow. From analyses of atmospheric halogens, *Heumann et al.* (1987) concluded that the gaseous compound methyl iodide (CH_3I) is probably responsible for the enrichment of I in Antarctic meteorites. This organoiodine compound is formed by microorganisms in the surface layers of the ocean. Methyl iodide has a relatively long atmospheric lifetime compared with elemental I or other iodides, allowing the compound to be transported long distances in the atmosphere. After deposition it would be converted photochemically into other I species.

2.8. Noble Gases

In a study of the isotopic composition and concentration of noble gases in a suite of ordinary chondrites from the Sahara desert and Antarctica, *Scherer et al.* (1992) found that the concentration of atmospheric noble gases (specifically the heavy noble gases Kr and Xe) appears to vary with weathering. Krypton-84 is higher in chondrites from hot deserts than in modern falls or Antarctic finds. In later work *Scherer et al.* (1995) also showed that hot desert meteorites have excess ^{132}Xe . Xenon-132 was also found to be greater in Antarctic samples than in modern falls. The implication is that atmospheric Kr and Xe are incorporated into weathering products, presumably after being dissolved in ground water, the most highly weathered samples (hot desert meteorites) showing the largest enrichment (*Scherer et al.*, 1992).

2.9. Water-Soluble Minerals in Falls

It has recently become apparent that equilibrated ordinary chondrites may contain primary halides, recording aqueous alteration on the ordinary chondrite parent body (e.g., *Zolensky et al.*, 1998). These minerals are highly water soluble, and have only been observed in meteorites recovered soon after their fall. This raises the possibility that the mineral suite we observe in meteorites may be rapidly modified by the terrestrial environment. This is discussed in more detail in the section on carbonaceous chondrites.

2.10. Relationship Between Weathering and Terrestrial Age

Chondrites in hot deserts rarely have terrestrial ages >50 ka, but in exceptional cases they can survive for longer periods — *Welten et al.* (2004) recorded an H5 chondrite with a terrestrial age of 150 ± 40 ka. In Antarctica finds, terrestrial ages can be much greater. *Scherer et al.* (1997) and *Welten et al.* (1997) have discovered meteorites with terrestrial ages of 2 Ma and 2.35 Ma respectively.

A number of radionuclides, most notably ^{26}Al , ^{36}Cl , ^{81}Kr , and ^{14}C , have been used to estimate the terrestrial age of meteorites. Exposure ages of meteorites in space range from

a few million years to 50 m.y. (*Crabb and Schultz*, 1981). During this time, a meteorite is bombarded with cosmic rays, and through the interaction of these with meteoritic minerals accumulates a population of cosmogenic stable nuclides and cosmogenic radionuclides with variable half-lives. Following its fall to Earth, the meteorite is shielded from cosmic-ray bombardment, so a terrestrial age can be calculated using the known level of saturation of radionuclides such as ^{14}C within the sample and the observed concentration of cosmogenic radionuclides (*Jull*, 2006). The half-life of ^{14}C is 5730 yr, giving a maximum measurable terrestrial age of about 40 ka (*Jull et al.*, 1989). This isotope is commonly used to measure terrestrial ages of hot-desert meteorites (*Jull et al.*, 1991). Krypton-81 and ^{36}Cl have longer half-lives (210,000 and 301,000 yr respectively) and are more often used to measure the terrestrial age (or residence time) of Antarctic meteorites (*Freundel et al.*, 1986). The calculation of terrestrial ages from the ^{14}C composition of meteorites involves the heating of samples to >250°–500°C to remove terrestrial contamination. The bulk of the error on these measurements comes from the possibility of so-called “self-shielding,” i.e., from a variation in ^{14}C with depth in the original stone. The maximum terrestrial age measurable by ^{14}C dating is around 42 ka years due to *in situ* production of ^{14}C at Earth’s surface from cosmic rays that reach the ground.

In a ^{14}C study of terrestrial ages of meteorites from the central and southwestern United States, *Jull et al.* (1993a) estimated a weathering “half-life” for meteorites (or decay time) of between 10,000 and 15,000 yr, 4–5× longer than previous estimates (*Boeckl*, 1972) (the earlier estimate was based on a small sample population and substantially underestimated the average terrestrial age of meteorite finds from this region). Only a weak correlation between terrestrial age and weathering was found (*Jull et al.*, 1993a), although there appears to be a better correlation in the population of meteorites from Roosevelt County, New Mexico (*Jull et al.*, 1991; *Wlotzka et al.*, 1995). In a similar study of 13 meteorites from Western Libya, *Jull et al.* (1990) found a peak in the age distribution at 4–8 ka years and only two samples with older ages. They suggest the reason for this may be related to climate changes in North Africa around this time (*Butzer et al.*, 1972), which saw increased precipitation (>200 mm/year) between 10,000 and 7700 years ago (*Pachur*, 1980). In contrast, the meteorites from Texas and New Mexico show a deficit of “young” samples, so some selection process also appears to be at work in this region (*Jull et al.*, 1993b).

More recently, *Bland et al.* (1996a,b, 1998a,b,c) have used Mössbauer spectroscopy to show that oxidation increases over time, with a rapid initial period followed by more gradual subsequent alteration, and that weathering rate varies substantially between different regions. Correlations between the degree of weathering that a sample exhibits, and its terrestrial age, are observed for meteorites from all hot desert accumulations studied (Fig. 2). In addition [and contrary to earlier studies by *Burns et al.* (1995)], although the data show more scatter than is typical of hot desert

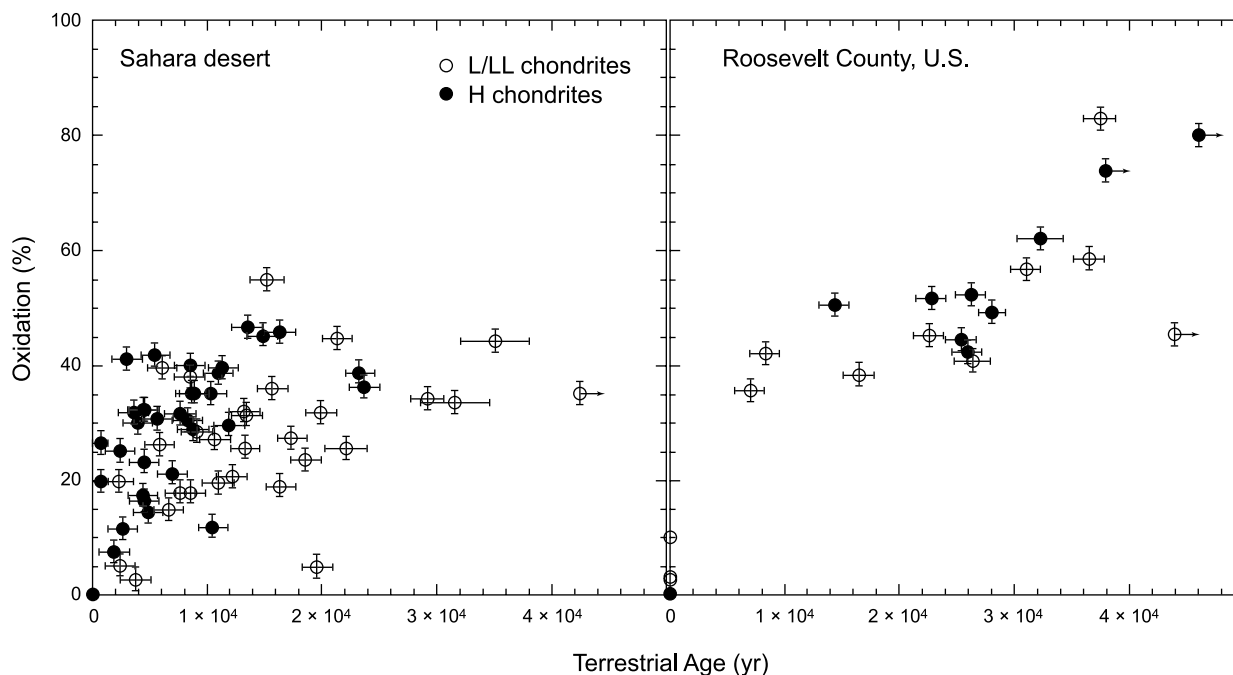


Fig. 2. Scatter of total ferric oxidation (as measured by ^{57}Fe Mössbauer spectroscopy) against terrestrial age for H and L (LL) ordinary chondrites from the Sahara Desert and Roosevelt County. Note the relatively rapid initial weathering phase.

populations, there is a correlation between weathering and terrestrial age for meteorites from Allan Hills, allowing an estimate of the rate of weathering over time for this accumulation (Bland *et al.*, 1998c). Overall, weathering rates in meteorites may be approximated by an appropriate power law (Fig. 3). It is apparent from this work that H chondrites weather significantly faster than L and LL chondrites.

2.11. Passivation of Weathering by Porosity Reduction

Ordinary chondrite falls are quite porous [11–15% (Wasson, 1974)]. Oxidation of metal leads to substantial volume expansion (Buddhue, 1957), which may reduce porosity, thus reducing the ability of water to penetrate the sample. Since porosity and permeability are among the most important factors influencing weathering rates in silicate rocks (Noack *et al.*, 1993), any variation in porosity (and permeability) in meteorites would affect the rate at which samples are weathered. Bland *et al.* (1998a) compared porosity and oxidation data (as measured by Mössbauer spectroscopy) for 23 ordinary chondrites, weathered in a variety of climatic regimes, to average data for falls. The results indicate an initial rapid decrease in porosity with weathering until after ~20% of the original Fe has been converted to Fe^{3+} , and porosity then stabilizes.

Given that we observe an initial rapid weathering phase, followed by a much-reduced weathering rate, some passivating mechanism is required to explain the survival of meteorites with low levels of weathering over extended time-scales. Bland *et al.* (1998a) suggest that the most significant

process involved is reduction in porosity, restricting water flow throughout the sample. Britt and Consolmagno (2003) reached a similar conclusion following a review of meteorite porosity data. The act of precipitating a diffusion-resistant film around Fe-Ni and troilite grains may also inhibit further oxidation, once some critical thickness is reached.

In an early study of meteorite weathering, Buddhue (1957) calculated that (given a specific gravity of pure Fe of 7.85, and meteorite rust of 4.24, and assuming spherical metal grains) oxidation of 10 wt% metal would produce a total volume increase of ~12%. The average metal content of an H-chondrite fall is 16–18 wt% (Keil, 1962; Jarosewich, 1990; McSween *et al.*, 1991), so oxidation of half the metal in a fresh H chondrite would be sufficient to reduce porosity to zero. As such, the minimum oxidation (measured by Mössbauer spectroscopy) required to passivate weathering may be as low as 8–10%. Lee and Bland (2004) modeled the volume changes following alteration of primary minerals for a range of weathering products, and showed that the primary porosity of most meteorites is sufficient to accommodate weathering products. Dilation of primary pores and brecciation, which has been observed in parts of some meteorites, will only occur if the meteorite is especially metal-rich, or has a low primary porosity.

3. WEATHERING OF CARBONACEOUS CHONDRITES

In terms of the alteration of anhydrous silicates, metal, and sulfide, there will clearly be a number of similarities between ordinary and carbonaceous chondrite weathering.

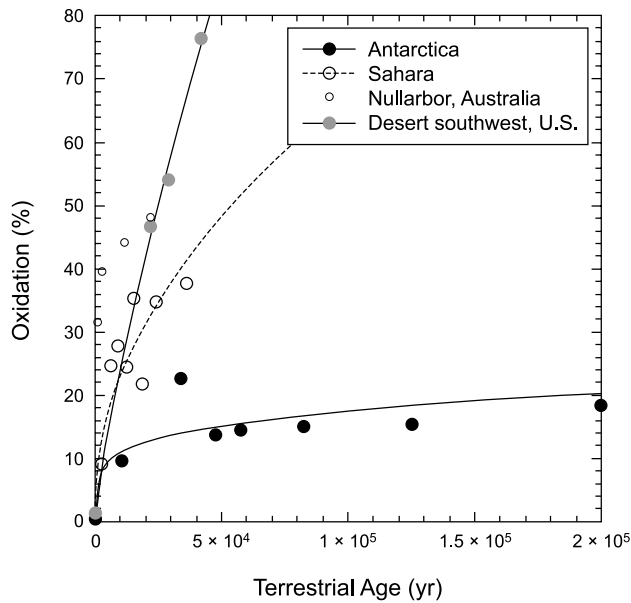


Fig. 3. Terrestrial age vs. oxidation compared between sites. Data are binned by terrestrial age (for each site) so that a similar number of data points fall in each bin. This comparison shows that there is significant variation in weathering rate between sites, presumably as a result of broad differences in regional climate over the lifetime of the accumulation.

However, carbonaceous chondrites differ in a number of ways that are significant in considering how they respond to the terrestrial environment. These include a larger proportion of fine grained matrix (and a correspondingly larger surface area of reactant), enrichment in volatile elements, and the presence of abundant organic compounds. Although meteorite finds will clearly be affected to varying degrees by weathering, some of these factors make carbonaceous chondrite falls susceptible to alteration. It is noteworthy, for instance, that although we have tens of thousands of recovered finds, the only CI1 chondrites in our collections are falls: CIs are clearly not robust enough to last long in the terrestrial environment.

3.1. Alteration of Carbonaceous Chondrite Finds

3.1.1. Elemental mobilization. In an INAA study of CR chondrites, including falls and finds, *Kallemeyn et al.* (1994) observed a large variability in Br abundance in the Antarctic finds — as in the case of I contamination discussed previously, Br can be transported from sea spray (*Heumann et al.*, 1987). Concentrations of Na are also low and highly variable, which *Kallemeyn et al.* (1994) suggests may be related to loss of Na by leaching of sulfates during weathering. Saharan samples also appear to be anomalously high in K (*Kallemeyn et al.*, 1994). In a trace-element study of carbonaceous chondrite matrix, *Bland et al.* (2005) also observed superchondritic abundances of Sr in two Saharan

finds, similar to observations of weathered Omani ordinary chondrites (*Al-Kathiri et al.*, 2005).

3.1.2. Weathering of meteoritic organic matter and contamination by terrestrial carbon. Within carbonaceous chondrites the effects of weathering are likely to be most severe for the more labile materials, such as organic matter. The majority of organic matter in meteorites comprises aromatic rings with C-bearing side chains and O-containing functional groups. This material is, at present, our only available record of natural prebiotic chemical evolution and appears to contain some exceedingly ancient materials that are only partially transformed from presolar matter (*Sephton*, 2002, 2004). Hence, it is important to understand just how pristine and uncorrupted the organic matter is in carbonaceous finds.

It is apparent that the organic material in carbonaceous chondrites recovered from hot deserts has been substantially modified. Meteorites from Reg el A fer (Algeria) show a depletion in bulk C, faster than any gain in C from evaporitic carbonate (*Ash and Pillinger*, 1992b), and it is likely that the macromolecular C these meteorites contained has in most cases been destroyed by extreme weathering conditions. *Newton et al.* (1995) also has found a relationship between the isotopic composition of C in CO chondrites and the recovery site: Fresh falls have the most primitive isotopic composition; meteorites from Allan Hills, Antarctica, trend toward more terrestrial isotopic values; while the most contaminated samples are those from Reg el A fer, Algeria, and the Yamato Mountains, Antarctica. This is interesting, as Antarctic samples are frequently assumed to have experienced only limited alteration.

The effect of Antarctic weathering on meteoritic organic matter has recently been investigated using a combination of M ssbauer spectroscopy and thermal-extraction/pyrolysis-gas chromatography-mass spectrometry (Py-GC-MS) (*Sephton et al.*, 2004). Preterrestrial aqueous processing and terrestrial weathering of meteorite samples is usually associated with the oxidation of Fe-bearing minerals that can be quantified using M ssbauer spectroscopy, allowing samples to be placed on a scale of relative alteration. Any corresponding alteration of the overall organic constitution of small meteorite samples can be determined by using Py-GC-MS. In an attempt to discriminate between non-Antarctic and Antarctic processes, *Sephton et al.* (2004) studied a series of organic-rich CM falls and CM Antarctic finds.

Interestingly, the inorganic and organic constituents of CM chondrites seem to exhibit responses to both parent-body aqueous alteration and terrestrial weathering. Increased oxidation levels for Fe-bearing minerals within the non-Antarctic chondrites appear to be a response to greater amounts of parent-body aqueous alteration. Moreover, when the M ssbauer responses are used as a guide it is evident that parent-body processing removes O-containing organic entities from organic material and C-bearing side chains from its constituent units.

M ssbauer spectroscopy indicates that the level of alteration (oxidation) displayed by Fe-bearing minerals in the Antarctic samples is generally higher than the non-Antar-

tic sample set. Organic responses to Antarctic weathering proceed in a similar fashion to parent-body aqueous alteration (removal of side chains and loss of O-containing functional groups) but are more extreme, leading to an enhancement of the organic responses observed in the falls. The record in Antarctic chondrites is clearly not pristine. Instead, these meteorites have been altered in a subtle way that mimics genuine preterrestrial alteration. As was noted in the discussion of cronstedtite recently observed in ordinary chondrites weathered in Antarctica (*Lee and Bland, 2004*), there appear to be a number of similarities between the chemical microenvironments occurring within chondrites undergoing weathering on the Antarctic ice at the present day, and conditions during aqueous alteration on carbonaceous chondrite parent bodies in the early solar system.

3.2. Alteration of Carbonaceous Chondrite Falls

What makes carbonaceous chondrites unusual among the various meteorite groups is that there is significant evidence for terrestrial alteration of falls. This takes a number of forms, including addition of hygroscopic water, formation of sulfate and carbonate veins, mobilization of aqueous trace elements, and possible reordering of phyllosilicates.

3.2.1. Elemental mobilization. *Friedrich et al. (2002)* compared the composition of pristine Tagish Lake to that from samples that had come into contact with icewater. Elements depleted in the altered sample were Sr and Bi, both highly mobile trace elements. Uranium, In, and Cd were enriched in the altered sample, most likely deposited from contaminated meltwater.

3.2.2. Sulfate and carbonate veining. *Gounelle and Zolensky (2001)* showed that the ubiquitous sulfate veins observed in CI1 chondrites were of terrestrial origin. The first description of sulfate veins in Orgueil was made in 1961 by *Dufresne and Anders (1962)*, almost 100 years after the meteorite fell. A close reading of the earlier descriptions of CI1 chondrites finds no mention of sulfate veins, despite the fact that these earlier workers were limited to describing the macroscopic properties of a sample. Rather, the extremely friable, loose nature of the material, as well as its high porosity, were regularly noted. An additional property that interested early workers was the extreme avidity of CI1 material for water. *Pisani (1864)* dehydrated a sample of Orgueil, and observed that after a few hours it had regained almost all the hygroscopic water that it had lost (9.15 wt%). It appears that over the period since these meteorites fell, they have continued to acquire water — *Cloëz (1864)* reported 9.06 wt% water in Orgueil a few weeks after the fall, and nearly a century later, *Wiik (1956)* found 19.89 wt%. It is probable that this property of CI1 chondrites has led to the remobilization of any initial fine-grained sulfate, and the oxidation of sulfides, to produce vein sulfate (*Gounelle and Zolensky, 2001*). The final word on a terrestrial origin is provided by the O-isotopic composition of Orgueil sulfate, which is terrestrial (*Aireau et al.,*

2001), and the example of the 2.5-kg sample of Orgueil at Musée d'Histoire Naturelle de Montauban, which shows abundant white sulfate veins growing up through the black fusion crust (*Gounelle and Zolensky, 2001*).

A recent study of the CV3 chondrite Vigarano (*Abreu and Brearley, 2005*) observed a variety of calcite morphologies, including networks of veins, vesicle fillings in the fusion crust, and pseudomorphic replacement of augite. Most veins vary in thickness from $<1 \mu\text{m}$ to $25 \mu\text{m}$, and extend for several hundred micrometers. Some veins crosscut the fusion crust and are connected to a carbonate coating on the exterior of the meteorite. The conclusion is that the veins are terrestrial in origin (*Abreu and Brearley, 2005*).

3.2.3. Alteration of CM chondrite falls? It may be that similar processes are also acting to modify CM falls. CM chondrites contain minerals that formed by interaction between liquid water and anhydrous minerals (*McSween, 1979*). How this process progressed on the parent body is important for understanding how to unravel the effects of aqueous alteration in these meteorites. Studies undertaken to qualify (*McSween, 1979*) and quantify (*Browning et al., 1996*) the extent of alteration found in the CMs resulted in a mineralogic alteration index (MAI) (*Browning et al., 1996*). The MAI attempted to provide a numerical representation of the Fe^{3+}/Si ratio in matrix phyllosilicates in CM meteorite falls. It has been found to correlate with carbonate (*Benedix et al., 2003*) and sulfate (*Aireau et al., 2001*) O-isotopic composition. But as we have seen, these minerals may form in carbonaceous chondrite falls in the terrestrial environment. So a key question is the extent of terrestrial alteration within these meteorites.

Figure 4 is a plot of MAI vs. year of fall (*Benedix and Bland, 2004*) for all the CM chondrites for which an MAI was determined (*Browning et al., 1996*). The MAI has a

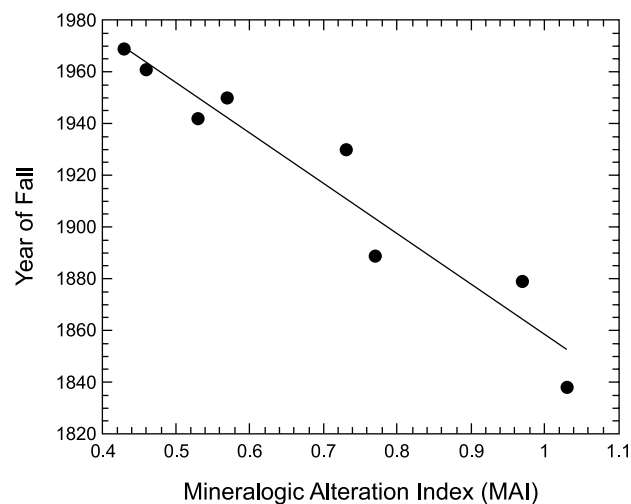


Fig. 4. Mineralogic alteration index (from *Browning et al., 1996*) vs. year of fall of CM chondrites. The line has a correlation coefficient of 0.93.

strong positive correlation with the year of fall ($R^2 = 0.93$); i.e., the more altered meteorites are older. There is no such systematic correlation between year of fall and matrix or bulk $\Delta^{17}\text{O}$, $\delta^{18}\text{O}$ (Clayton and Mayeda, 1999), water content, or δD (Eiler and Kitchen, 2004). Although not as robust, a correlation also exists between the year of fall and $\Delta^{17}\text{O}$ in sulfate and carbonate, two minerals that are susceptible to interaction with liquid water. The fact that MAI and carbonate and sulfate $\Delta^{17}\text{O}$ track with the year of fall indicates that interaction with the terrestrial atmosphere may have taken place, even though the meteorite was recovered soon after its fall (Benedix and Bland, 2004).

If the MAI is an indicator of terrestrial contamination, what does this measure? The procedure for determining an MAI involves an assumption of a generalized stoichiometry for phyllosilicate to derive Fe^{3+} content. But determining the abundance of Fe^{3+} is difficult using probe techniques, even in monomineralic, well-crystalline materials. CM matrix is fine-grained, and each probe analysis includes multiple phases. In addition, phyllosilicates are frequently inter-layered and of poor crystallinity. As such, the MAI may be recording a variety of factors in addition to Fe^{3+} abundance. One of these that may be significant for this discussion involves S-bearing phases. The MAI procedure involves the use of multiple screening devices that exclude analyses containing obvious unwanted phases (>50% of probe analyses are excluded in this way): S-rich analyses were excluded, but S is ubiquitous, and so S-bearing inclusions (assumed to be pyrrhotite) were also subtracted from each analysis. It is apparent that S is highly mobile in carbonaceous chondrite falls, and it appears at least possible that this mobilization is influencing the measured MAI.

Whether the MAI is recording terrestrial or preterrestrial alteration remains an open question. What is apparent is that carbonaceous chondrite falls are susceptible to terrestrial weathering: Determining where asteroidal alteration ends and terrestrial alteration begins is clearly a nontrivial task.

4. CONCLUSIONS

Terrestrial weathering can affect many of the properties of meteorites that make them valuable to researchers: chemistry, mineralogy, composition of organic matter, C- and O-isotopic composition, noble gases, etc. In analyzing meteorite finds it is important to be aware of any overprint on a sample's primordial composition imposed by the terrestrial environment. For a number of reasons, carbonaceous chondrites are more susceptible to terrestrial alteration than other meteorite types. It appears that in this group of meteorites, even falls may display the effects of significant terrestrial modification.

Although terrestrial weathering may obscure many of the properties that make a meteorite interesting to researchers, it can also provide an analog to help us understand asteroidal alteration. The weathering environment of Antarctica is similar in a number of ways to the alteration environment

in asteroids in the early solar system: The recognition that the same alteration products are produced (Lee and Bland, 2004), and that organic material responds in a similar way in both environments, serves to highlight this.

Finally, the fact that meteorites of similar composition are deposited over Earth's entire surface, and that their terrestrial exposure time may be determined quite precisely by measuring their cosmogenic radionuclide abundance, makes them an effective "standard sample" for studying how rocks are affected by the terrestrial environment. Meteorites are one of the few geological materials for which field weathering rates have been determined, allowing the influence of climate on rock weathering rate to be deduced.

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