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## Why aqueous alteration in asteroids was isochemical: High porosity $\neq$ high permeability

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#### ABSTRACT

Carbonaceous chondrite meteorites are the most compositionally primitive rocks in the solar system, but the most chemically pristine (CI1 and CM2 chondrites) have experienced pervasive aqueous alteration, apparently within asteroid parent bodies. Unfractionated soluble elements suggest very limited flow of liquid water, indicting a closed-system at scales large than 100's µm, consistent with data from oxygen isotopes, and meteorite petrography. However, numerical studies persistently predict large-scale (10's km) water transport in model asteroids, either in convecting cells, or via 'exhalation' flow — an open-system at scales up to 10's km. These models have tended to use permeabilities in the range  $10^{-13}$  to  $10^{-11}$  m<sup>2</sup>. We show that the permeability of plausible chondritic starting materials lies in the range  $10^{-19}$  to  $10^{-17}$  m $^2$  (0.1–10  $\mu$ D): around six orders-of-magnitude lower than previously assumed. This low permeability is largely a result of the extreme fine grain-size of primitive chondritic materials. Applying these permeability estimates in numerical models, we predict very limited liquid water flow (distances of 100's µm at most), even in a high porosity, water-saturated asteroid, with a high thermal gradient, over millions of years. Isochemical alteration, with flow over minimal lengthscales, is not a special circumstance. It is inevitable, once we consider the fundamental material properties of these rocks. To achieve large-scale flow it would require average matrix grain sizes in primitive materials of 10's-100's µm - orders of magnitude larger than observed. Finally, in addition to reconciling numerical modelling with meteorite data, our work explains several other features of these enigmatic rocks, most particularly, why the most chemically primitive meteorites are also the most altered.

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#### 1. Introduction

The first studies of carbonaceous chondrites (CCs) (e.g. Daubrée, 1867) recognised that their mineralogy was highly altered, with many samples containing clays, magnetite, carbonates and other secondary minerals. Initially, the abundant magnetite found in some CCs was thought to be a nebula condensate (Larimer and Anders, 1967) or, together with clay minerals, the product of aqueous alteration that

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occurred in the solar nebula by the reaction of water vapour and anhydrous minerals (Grossman and Larimer, 1974). However, a range of evidence now suggests that aqueous alteration occurred within CC parent asteroids (Brearley, 2003). Moreover, geochemical and petrographic studies of meteorites suggest that aqueous alteration was isochemical. We review the evidence for this in the next section. In contrast, all numerical models of asteroidal aqueous and thermal alteration have predicted large-scale flow of liquid water (Grimm and McSween, 1989; Cohen and Coker, 2000; Coker and Cohen, 2001; Young et al., 1999, 2003; Young, 2001a,b; 2004; McSween et al., 2002; Travis and Schubert, 2005; Grimm, 2007). In many cases, liquid transport occurs over 10's km, either as a single-pass 'exhalation' from the interior of the body driven by excess gas pressure (Young et al., 1999; McSween et al., 2002), as episodic flow (Cohen and Coker,

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2000; Coker and Cohen, 2001), or in convection cells driven by buoyancy (Grimm and McSween, 1989; Young et al., 1999, 2003; McSween et al., 2002; Travis and Schubert, 2005). But whatever form it takes, the movement of liquid water through the rock matrix will fractionate aqueous species, which appears to directly contradict the available evidence from meteorites. Model predictions indicate that alteration was open-system with large-scale flow; measured data suggest that alteration was isochemical with very limited flow.

It is the goal of this study to resolve these paradoxes: why the most chemically primitive rocks are the most aqueously altered, and why numerical modelling studies predict large-scale flow which is not consistent with data from meteorite geochemistry and petrography studies. We begin by reviewing the evidence for isochemical aqueous alteration, the temperature dependence of alteration, and considering relevant oxygen isotope studies, before discussing the permeability values used in transport models of CC parent asteroids. We then quantify the pore- and grain-size distribution in a primitive materials, and use these data to estimate permeability in CC parent asteroids. Applying our revised estimates of permeability in numerical models, we find that their predictions can be reconciled with the observed data. Despite their high porosity, the estimated permeability of CC meteorites is very low, which restricts the transport of liquid water to very short lengthscales.

#### 1.1. Chemistry

Extensive alteration within asteroids has not modified CC chemistry. CI-type carbonaceous chondrites have compositions that are within 10% of the solar photosphere for most elements (Palme and Jones, 2003). Indeed, CIs are frequently used to refine estimates of photosphere element abundance, because they are available for laboratory study. Moreover, they are the geochemical standard against which all other terrestrial rock analyses are normalised, because it is recognised that they define an 'average' solar system composition. Yet CIs are also the most aqueously altered CCs. Why the most chemically primitive rocks should have experienced the most alteration has always been a paradox. Analyses of other CC groups showed monotonic, volatility-controlled depletions compared to CI (Kallemeyn and Wasson, 1981), indicating minimal geochemical fractionation. Yet these rocks also experienced varying degrees of aqueous alteration. To preserve solar abundances in elements which are easily mobilised in fluids, it was postulated that aqueous processing in asteroids was isochemical (McSween, 1979), occurring in a closed system (Kerridge et al., 1979) with element mobility restricted to short lengthscales (a few 100 µm at most).

Recent estimates of solar chemical composition not only confirm the excellent agreement between photospheric and CI chondrite abundances (Grevesse et al., 2007), they also show that there is no evidence for fractionation of soluble elements. Ignoring light elements, and elements where photospheric abundances are poorly constrained, the mean difference between photospheric and CI abundances is  $0.014 \pm$ 0.06 dex (Grevesse et al., 2007). Selecting the most soluble elements from this group (25% of the total), the mean difference between photospheric and meteoritic abundances is  $0.016 \pm 0.06$  dex. There is no difference, within experimental error, between photosphere and meteorite compositions in the subset of soluble elements and the overall dataset. Fractionation of the soluble elements has not occurred, so aqueous alteration (by definition) must have been isochemical. It is the consensus view that isochemical alteration requires the flow of liquid water to be minimal (Brearley, 2003) i.e. a closed system over lengthscales > 100's µm. Finally, chemical analyses of small (10-100 mg) bulk chondrite aliquots support this conclusion, showing remarkable reproducibility between samples and no evidence for element mobility (Wulf et al., 1995). Similarly, analyses of the trace element composition of fine-grained materials in CCs indicate minimal aqueous mobility (Bland et al., 2005).

#### 1.2. Petrography

Regardless of the primitive meteorite group under consideration, chondrite petrography is broadly in agreement with the geochemical analysis, with evidence for metasomatism generally restricted to distances of less than a few 100 µm. In some unequilibrated ordinary chondrites, 'bleaching' of chondrules, and zonation in moderately volatile elements and Ca, indicate aqueous mobility over ~100 μm (e.g. Grossman et al., 2000, 2002; Grossman and Alexander, 2004). Similarly, evidence for iron-alkali aqueous metasomatism over comparable lengthscales is found around chondrules and calcium-aluminium-rich inclusions in CV chondrites (Krot et al., 1995). In the CR2 chondrites, 50- $100 \, \mu m$  thick zones of alteration are observed around chondrules, where aqueous elements have exchanged with surrounding rim or matrix material (Burger and Brearley, 2004, 2005). Chizmadia and Brearley (2004) and Brearley and Chizmadia (2005) studied the behaviour of Fe, S, Ni, Ca, Na, K and P during aqueous alteration of CM2 chondrites and showed that chondrule mesostasis, in even the least altered of these meteorites, is completely replaced by phyllosilicates. But although chondrule glass is replaced, and a proportion of the aqueous elements within it dissolved in the altering fluid, element mobility is restricted to a layer ~10 µm in thickness at the chondrule-rim boundary in the less altered CM2s, or a more homogenous distribution within the rim up to 25 µm from the chondrule in the more altered CM2s (Brearley and Chizmadia, 2005). In the CI chondrites, Morlok et al. (2006) found that the variable level of alteration observed in these meteorites was consistent with a very low mobility of materials in solution during aqueous alteration, indicating closed system alteration over lengthscales of ~100 μm. From this review, it is apparent that even in rocks that have experienced substantial aqueous alteration, such as the CM2 chondrites, element mobility was largely restricted to narrow zones around chondrules. Evidence for metasomatism over lengthscales > 100's µm is lacking.

#### 1.3. Element mobility and temperature

It has been suggested that large-scale flow may have taken place, but that soluble elements were not fractionated as alteration occurred at low temperatures. If water temperatures were low, what would one expect? Element solubility would certainly decrease, and less of the element would go into solution. But (unless there were rapid temperature changes over very short lengthscales), we would not expect element reprecipitation on small scales. Temperature cannot therefore account for the small-scale (10's to 100's µm mobility) that is observed in meteorite petrographic studies. Low temperature alteration could account for decreased evidence of total solubility, but a cold fluid moving through a large asteroid would mobilise a minor proportion of an element, and deposit that element at large distances. In contrast, the petrographic data from meteorites indicates that soluble elements were significantly fractionated by fluids, mobilised, and then reprecipitated, but over distances not greater than 100's µm.

In addition, temperatures of alteration do not appear to have been uniformly low. Initial estimates for alteration temperatures in CM2s were in the range 0–25 °C (Clayton and Mayeda, 1984, 1999; Zolensky et al., 1989; Rosenberg et al, 2001; Benedix et al., 2003). A recent study applying the 'clumped-isotope' carbonate thermometer to CM2s (Guo and Eiler, 2007) has refined this, with six out of seven carbonate formation temperatures falling in the range 20–35 °C (Guo and Eiler, 2007). CIs appear to have experienced alteration temperatures of 50 to > 150 °C (Clayton and Mayeda, 1984, 1999; Zolensky et al., 1989; Leshin et al., 1997), while the anomalous C1/C2 chondrite Tagish Lake may have experienced alteration temperatures intermediate between CM and CI (Baker et al., 2002). In meteorites, these temperatures have led to dissolution of primary phases and element mobility — but again, mobility is only observed over relatively small distances (e.g. in aqueously altered CVs (Bali), CMs, and CRs), we see evidence for

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