

1 **Why aqueous alteration in asteroids was isochemical:**

2 **High porosity \neq high permeability**

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19 **ABSTRACT**

20 Carbonaceous chondrite meteorites are the most chemically primitive rocks in the solar system, but the most
21 compositionally pristine have experienced pervasive aqueous alteration, apparently within asteroid parent
22 bodies. Unfractionated soluble elements suggest minimal flow of liquid water, consistent with data from
23 oxygen isotopes, and meteorite petrography. However, numerical studies persistently predict large-scale water
24 transport in model asteroids. These models have tended to use permeabilities in the range 10^{-13} to 10^{-11} m². We
25 show that the permeability of plausible chondritic starting materials lies in the range 10^{-19} to 10^{-17} m² (0.1 – 10
26 μ D): around six orders-of-magnitude lower than previously assumed. This is largely a result of the extreme

1 fine grain size of primitive chondritic materials. Applying these permeability estimates in numerical models,
2 we predict very limited liquid water flow (distances of 100's μm at most), even in a high porosity, water-
3 saturated asteroid, with a high thermal gradient, over millions of years. Isochemical alteration, with minimal
4 fluid flow, is not a special circumstance. It is inevitable, once we consider the fundamental material properties
5 of these rocks. To achieve large-scale flow would require average matrix grain sizes in primitive materials of
6 10's – 100's μm – orders of magnitude larger than observed. Finally, in addition to reconciling numerical
7 modelling with meteorite data, our work explains several other features of these enigmatic rocks, most
8 particularly, why the most chemically primitive meteorites are also the most altered.

9

10 KEYWORDS

11 meteorite; carbonaceous chondrite; asteroidal alteration; isochemical alteration; permeability

12

13 1. Introduction

14 The first studies of carbonaceous chondrites (CCs) (e.g. Daubrée, 1867) recognised that
15 their mineralogy was highly altered, with many samples containing clay minerals,
16 magnetite, and carbonates etc. Initially, the abundant magnetite found in some CCs was
17 thought to be a nebula condensate (Larimer and Anders, 1967), or, together with clay
18 minerals, the product of aqueous alteration that occurred in the solar nebula by the reaction
19 of water vapour and anhydrous minerals (Grossman and Larimer, 1974). However, a range
20 of evidence now suggests that aqueous alteration occurred within CC parent asteroids
21 (Brearley, 2003).

22 1.1. Chemistry

23 Curiously, extensive alteration within asteroids did not modify CC chemistry. CI-type
24 carbonaceous chondrites have compositions that are within 10% of the solar photosphere
25 for most elements (Palme and Jones, 2003). Their similarity to photosphere abundances,

1 and the fact that CIs can be analysed in the lab, mean that they are frequently used to refine
2 photosphere abundances. Defining an ‘average’ solar system composition is also a property
3 which has made them the geochemical standard against which all other terrestrial rock
4 analyses are normalised. Yet CIs are also the most aqueously altered CCs. Why the most
5 chemically primitive rocks should have experienced the most alteration has always been a
6 paradox. Early analyses of other CC groups showed uniform volatility-controlled
7 depletions compared to CI (Kallemeyn and Wasson, 1981). These rocks also experienced
8 varying degrees of aqueous alteration. To preserve solar abundances in elements which are
9 easily mobilised in fluids it was postulated that aqueous processing in asteroids was
10 isochemical (McSween, 1979), occurring in a closed system (Kerridge et al., 1979). It is
11 noteworthy that recent estimates of solar chemical composition not only confirm the
12 excellent agreement between photospheric and CI chondrite abundances (Grevesse &
13 Asplund 2007), they also show that there is no evidence for fractionation of soluble
14 elements. Ignoring light elements, and elements where photospheric abundances are poorly
15 constrained, the mean difference between photospheric and CI abundances is 0.014 ± 0.06
16 dex (Grevesse & Asplund 2007). Selecting the most soluble elements from this group (25%
17 of the total), the mean difference between photospheric and meteoritic abundances is 0.016
18 ± 0.06 dex ie. there is no significant difference between photosphere / meteorite
19 compositions in the overall dataset, and photosphere / meteorite compositions in the soluble
20 element subset. Finally, isochemical / closed-system alteration requires the flow of liquid
21 water to be essentially zero (Brearley, 2003). More recent chemical analyses of small (10-
22 100 mg) bulk chondrite aliquots (Wulf et al., 1995) show remarkable reproducibility
23 between samples, and no evidence for element mobility. Similarly, analyses of the trace

1 element composition of fine-grained materials in CCs (Bland et al., 2005) indicate minimal
2 aqueous mobility.

3 *1.2. Petrography*

4 Geochemical analysis is broadly in agreement with chondrite petrography, where
5 evidence for metasomatism is generally restricted to distances of less than a few 100 μm .
6 Aqueous mobility over $\sim 100 \mu\text{m}$ appears to have occurred in some unequilibrated ordinary
7 chondrites, with ‘bleaching’ of chondrules and zonation in moderately volatile elements
8 and Ca (e.g. Grossman et al., 2000, 2002; Grossman and Alexander, 2004). Similarly,
9 evidence for iron-alkali aqueous metasomatism is found around chondrules and CAIs in CV
10 chondrites (Krot et al., 1995). In the CR2 chondrites, zones of alteration are observed
11 around chondrules, where aqueous elements have exchanged with surrounding rim or
12 matrix material. In the CR2s these zones are 50-100 μm thick (Burger and Brearley, 2004,
13 2005). A number of studies have investigated metasomatism in the CM chondrites.
14 Chizmadia and Brearley (2004) and Brearley and Chizmadia (2005) have studied the
15 behaviour of Fe, S, Ni, Ca, Na, K and P during aqueous alteration of CM2 carbonaceous
16 chondrites. Chondrule mesostasis, in even the least altered of these meteorites, is
17 completely replaced by phyllosilicates. But although chondrule glass is replaced, and a
18 proportion of the aqueous elements within it dissolved in the altering fluid, element
19 mobility is restricted to a layer $\sim 10 \mu\text{m}$ in thickness occurring at the chondrule-rim
20 boundary (in the case of less altered CM2s), or more homogenous distribution within the
21 rim (Brearley and Chizmadia 2005), up to 25 μm from the chondrule (in the case of more
22 altered CM2s). In the CI chondrites, in an extensive study, Morlok et al. (2006) found that
23 the variable level of alteration observed in these meteorites was consistent with a very low
24 mobility of materials in solution during aqueous alteration, probably on a scale of only 100

1 μm : closed system alteration was favoured (Morlok et al., 2006). From this review it is
2 apparent that even in rocks that have experienced substantial aqueous alteration, such as the
3 CM2 chondrites, elemental mobility was largely restricted to comparatively narrow zones
4 around chondrules. In the majority of cases, evidence for metasomatism over length scales
5 $\gg 100$'s μm is lacking.

6 *1.3. Alteration timescales*

7 Mn-Cr carbonate data can be interpreted as suggesting extended timescales for continual
8 aqueous alteration within C1 and C2 parent bodies (e.g. Hoppe et al., 2007; de Leuw et al.,
9 2009; Petitat et al., 2009; Tyra et al., 2009). However, thermodynamically (given fine-
10 grained reactants) this argument is difficult to support. Rather, as noted by Hoppe et al.
11 (2007), episodic alteration possibly resulting from impact heating and remobilisation of
12 fluids is also consistent with the existing Mn-Cr dataset. A plausible scenario would
13 involve the bulk of aqueous alteration occurring soon after asteroid accretion (when heat
14 generated from decay of short-lived radionuclides was available for melting water), and
15 being relatively brief, as alteration reactions are exothermic, and anhydrous reactants are
16 fine-grained (Wood and Walther, 1983); Olsen et al. 1988; Zolotov and Mironenko, 2008).
17 Heating and re-mobilisation of fluids following periodic impact, with minor additional
18 alteration, would be superimposed on this initial major phase.

19 *1.4. Oxygen isotope models*

20 Popular oxygen isotope hydration models postulate simple exchange between a static
21 fluid and anhydrous phases in a closed system (Clayton and Mayeda, 1984), where
22 water:rock (w/r) ratios were relatively high (Clayton and Mayeda, 1984, 1999; Leshin et
23 al., 1997). However, there are alternatives to closed-system oxygen models. Briefly, the
24 suggestion is that CC meteorites vary in their mineralogies and oxygen isotope ratios as a

1 result of having come from different regions of a hydrological flow system (Young et al.
2 1999; Young, 2001a). The Young et al. open-system oxygen model defines a dramatically
3 different conceptual framework for understanding CC oxygen isotopes. However, based on
4 existing data, the jury is still out – open-system and closed-system models are debated, with
5 both types of models apparently capable of reproducing CC oxygen isotopes.

6 Although the Clayton and Mayeda (1984, 1999) and Leshin et al. (1997) models take
7 static exchange as a starting point, it has recently been suggested that the w/r ratios derived
8 from these models (and literature porosity data for CCs) make large-scale fluid flow
9 inevitable (Grimm, 2007). This is based on the assertion (Grimm, 2007) that measured
10 (post-reaction) porosities in the most altered CCs (CI and CM-type carbonaceous
11 chondrites) average only 11-12%. Given estimates for w/r ratio (and equivalent pre-reaction
12 porosity) from oxygen isotopes (Clayton and Mayeda, 1984, 1999; Leshin et al., 1997) this
13 would indicate a shortfall in porosity for CCs ie. >1 pore volume of water would be needed
14 to satisfy oxygen isotope constraints, thus requiring flow. There are a number of problems
15 with this argument, the most important being that there is no shortfall in porosity: CI and
16 CM porosities do not cluster over a narrow 11-12% range. CIs have measured porosities of
17 35% (Consolmagno and Britt, 1998), CMs have porosities up to 28% (Britt and
18 Consolmagno, 2003), and Tagish Lake (an intermediate CI/CM) has a calculated porosity
19 of 41% (Bland et al., 2004). In addition, there is ample evidence for remobilisation of
20 sulphates in the terrestrial environment acting to reduce porosity (Gounelle and Zolensky,
21 2001). Measured porosities therefore represent minimum pre-terrestrial values. Put simply,
22 there is no shortfall in porosity, so water pore volumes >1 are not required.

23 *1.5. Numerical modelling*

1 In contrast to geochemical, petrographic, and oxygen isotope studies of meteorites, all
2 numerical models of asteroidal aqueous and thermal alteration have predicted large-scale
3 flow of liquid water (Grimm and McSween, 1989; Cohen and Coker, 2000; Coker and
4 Cohen, 2001; Young et al., 1999, 2003; Young 2001a, 2001b, 2004; McSween et al., 2002;
5 Travis and Schubert, 2005; Grimm, 2007). In many cases, fluid transport occurs over 10's
6 km (Grimm and McSween 1989; Young et al. 1999; Coker and Cohen 2001; Travis and
7 Schubert 2005). Travis and Schubert (2005) found that the timing of the onset of
8 convection may vary, but that convective flow was observed in every scenario that was
9 considered, with convection patterns occurring as plumes or broad ridges of upwelling.
10 These authors concluded that convection should occur for a range of porosities, parent body
11 sizes, and radiogenic element concentrations, and that it should last for several Ma. Young
12 et al. (1999) proposed a model for the evolution of carbonaceous chondrite oxygen isotopes
13 based on down-temperature fluid flow within a 50km diameter body. In this situation, flow
14 occurs as a single pass, 'exhalation' flow, driven by internal gas pressure, rather than
15 convective circulation. Young et al. (2003) confine convective flow to bodies >80km
16 diameter, but note that 'exhalation' flow should occur in smaller objects. McSween et al.
17 (2002) observe this type of flow in bodies as small as 18km in diameter. But whether flow
18 is observed as a single pass 'exhalation' (Young et al., 1999), or in convecting cells
19 (Grimm and McSween, 1989; Cohen and Coker, 2000; Coker and Cohen, 2001; Young et
20 al., 1999, 2003; McSween et al., 2002; Travis and Schubert, 2005), the movement of liquid
21 water through rock should fractionate aqueous species. The modelling results appear to
22 directly contradict the meteorite data – the one indicates that alteration was open system,
23 with large-scale flow; the other, that alteration was isochemical, with minimal flow.

1 It is the goal of this study to resolve these paradoxes: why the most chemically primitive
2 rocks are the most aqueously altered, and why numerical modelling studies indicate large-
3 scale flow, which in general is not predicted by meteorite geochemistry, petrography, and
4 isotopic data.

5

6 **2. Estimating permeability of asteroid parent bodies**

7 For fluid flow to occur, a rock must be permeable. Permeability is a fundamental control
8 on fluid flow in porous media: it is the property of the media that determines the flow rate
9 which results from an applied pressure gradient. A rock can exhibit permeability at the
10 grain scale, or in fractures, or both. In C1 and C2 chondrites, pervasive, homogeneous
11 alteration at the grain scale clearly indicates that grain scale permeability was important.
12 Initial permeability estimates of 10^{-13} – 10^{-11} m² for chondritic asteroids (Grimm and
13 McSween, 1989) were based on what were felt to be appropriate terrestrial analogue
14 materials and measurements of lunar soil (Mercer et al., 1975; Lambe and Whitman, 1969;
15 Costes and Mitchell, 1970). This range of permeabilities was adopted in all subsequent
16 numerical modelling studies (Grimm and McSween, 1989; Cohen and Coker, 2000; Coker
17 and Cohen, 2001; Young et al., 1999, 2003; McSween et al., 2002; Travis and Schubert,
18 2005), but are these values appropriate for CC precursors?

19 Few measurements of meteorite permeability are available. Sugiura et al. (1984)
20 measured gas permeability for three chondrites, and Corrigan et al. (1997) extended the
21 permeability dataset to include more chondrites. However, it is not clear how subsequent
22 aqueous and thermal alteration, and impact shock, may have modified chondrite
23 permeability away from the primordial value that we are seeking. Therefore, in order to

1 constrain permeability (and therefore flow), what we require is a constraint on permeability
2 in the least altered, most primitive extraterrestrial materials currently available.

3 It is well known that permeability scales with pore-size, which in turn is related to grain
4 size, sorting, grain shape, grain packing, and the degree of cementation (Dullien, 1992). So
5 to extend our knowledge of meteorite permeability, we have investigated both grain- and
6 pore-size distributions in the most texturally and mineralogically primitive chondrite, and
7 derived a permeability range based on this analysis. The aim is to constrain the relevant
8 physical properties of the anhydrous precursor material, at the point when alteration begins.
9 A similar approach is taken in studies of flow in porous media in the terrestrial
10 environment. From our perspective, this will essentially define a grain-scale permeability
11 for chondrites. Following that, we will discuss the possibility of fracture permeability.

12 Chondrites are composed of two principal components: chondrules (spherical igneous
13 inclusions with diameters ranging from 100's μm to mm's), and a fine-grained
14 compositionally primitive matrix. The permeability of the matrix is the key control on fluid
15 flow, because this is the most abundant component in aqueously altered CCs (on average,
16 matrix in CMs accounts for >70% of the rock (McSween, 1979)), and water would initially
17 have been present in matrix pore spaces, or as ice rinds on matrix grains.

18 Despite the complexity of the relationship between permeability, pore size, grain size and
19 other textural properties, we can place a quantitative constraint on permeability using the
20 simple Carman–Kozeny equation, in which it is assumed that the porous medium can be
21 modelled as a series of equivalent conduits with a constant cross-sectional area of complex
22 shape. Permeability is then given by:

23

1
$$k = \frac{\varepsilon r_H^2}{h_{ck}} \quad (1)$$

2

3 where ε is the porosity, r_H is the hydraulic radius of the equivalent conduits and h_{ck} is a
4 constant which depends upon the pore geometry, and which has been found experimentally
5 to lie in the range $4.5 < h_{ck} < 5.5$ for unconsolidated porous media (Dullien, 1992). The
6 Carman–Kozeny equation successfully describes the permeability of many porous media,
7 particularly when there is a narrow range of grain-size and the grains are close to spherical
8 (Figure 3). For some consolidated porous media, the value of h_{ck} may be considerably
9 larger, with values up to 150 required to fit measured data (Dullien 1992).

10 The hydraulic radius r_H can be written as:

11

12
$$r_H = \frac{\varepsilon}{S(1-\varepsilon)} \quad (2)$$

13

14 where S is the specific surface area of the porous medium. Defining d to be the diameter of
15 the sphere which has the same specific surface area yields:

16

17
$$k = \frac{\varepsilon^3 d^2}{36h_{ck}(1-\varepsilon)^2} \quad (3)$$

18

19 which is a more familiar form of equation (1). The diameter d is often associated with
20 grain-size, although this is correct only for packed beds of spheres.

1 We estimate permeability based on the hydraulic radius determined from measured pore-
2 size distributions and equation (1). The relationship between porosity and permeability is
3 illustrated graphically, for a range of grain sizes in Figure 2 (in this case an intermediate h_{ck}
4 value of 50 was chosen).

5 In summary, although the individual accretionary setting for an asteroid varied, the
6 fundamental physical properties of matrix (grain size, sorting, grain shape etc.) may have
7 remained relatively constant. Defining those properties, and determining their significance
8 for permeability and fluid flow, are our initial goals. We use a combination of nuclear
9 magnetic resonance cryoporometry and transmission electron microscopy to constrain pore-
10 and grain-size distribution, and from that, permeability for a chondritic precursor using the
11 Carmen-Kozeny equation. Finally, using these derived permeabilities, we investigate
12 implications for fluid flow using a variety of asteroidal alteration models.

13

14 **3. Samples and Methodology**

15 In terms of its mineralogy and petrography, Acfer 094 is arguably the most primitive
16 carbonaceous chondrite in existence (Greshake, 1997; Nuth et al., 2005; Bland et al., 2007).
17 It provides a window on what chondritic materials may have looked like prior to aqueous
18 and thermal alteration. We have studied the matrix of this meteorite using transmission
19 electron microscopy (TEM) and nuclear magnetic resonance (NMR) cryoporometry
20 (Mitchell et al., 2008). TEM analysis was performed on two focussed ion beam (FIB) lift-
21 outs of portions of Acfer 094 matrix.

22 *3.1. Cryoporometry*

23 Measurement of pore-size distributions by NMR cryoporometry (for details see Mitchell
24 et al., 2008) were performed at the University of Kent. A probe liquid was imbibed into the

1 sample and NMR transverse (T_2) relaxation was used to monitor the phase changes as a
2 function of temperature, by studying changes in the dynamics of the liquid. This
3 information was then interpreted by application of the Gibbs-Thomson equation (constant-
4 pressure analogue of the Kelvin equation), whereby melting point is depressed inversely
5 with pore diameter.

6 *3.2. Microscopy*

7 FIB sample preparation and TEM observations were performed at both Imperial College
8 London and the University of Glasgow. At Glasgow, an 'in situ' FIB method was used: the
9 foil was cut from the thin section using a FEI Nova 200 Dualbeam FIB instrument. It was
10 then welded on to a Cu support and diffraction-contrast images were acquired using a FEI
11 T20 TEM operated at 20 kV. At Imperial College London, an 'ex-situ' preparation method
12 was used. The TEM section was prepared from the solid in the vacuum. It was transferred
13 outside the vacuum onto a TEM Cu grid covered with a thin carbon support film. Analysis
14 was performed using a JEOL JEM-2010 microscope operated at an accelerating voltage of
15 200 kV. An ISIS EDS system by Oxford Instruments was employed to analyse the
16 chemical composition of the samples and electron diffraction patterns used to identify
17 amorphous and crystalline phases. Importantly for this work, there is no differential
18 thinning in FIB preparation, so we can be confident that any porosity is primary, and not
19 enlarged by ion bombardment (as commonly occurs with 'normal' Ar ion milling for TEM
20 sample preparation).

21 *3.3. Numerical modelling*

22 We use a number of asteroidal alteration models (e.g. Grimm, 2007; Young et al., 2003;
23 Cohen and Coker 2000). Updated input parameters for Cohen and Coker (2000) (in
24 addition to those discussed in the text) include more recent evaluations for the vapour

1 pressure over ice, the thermal conductivity of ice, the thermal conductivity, viscosity, and
2 surface tension of liquid water, the heat capacity of minerals, the density of minerals and
3 bulk rock, and the thermal expansion coefficient and bulk modulus of minerals. We also
4 include dehydration (not relevant to models presented here), a porosity dependent
5 permeability (as discussed here), and the instantaneous venting of water vapour when the
6 vapour pressure exceeds the lithostatic pressure.

7

8 **4. Results**

9 *4.1. Grain- and pore-size distribution*

10 It is apparent from our own TEM analysis, and previous work (Nuth et al., 2005), that
11 there are abundant grains in the 50-100 nm size range (Figure 3) – over the total area of our
12 FIB sections ($\sim 110 \mu\text{m}^2$) we observe only a single grain of $\sim 1 \mu\text{m}$ in size. Defining a lower
13 limit on grain-size is difficult, since the smallest grains have a diameter much less than the
14 thickness of a FIB section, but a size range of 20-200 nm would appear to be appropriate
15 for the bulk of Acfer 094 matrix grains. This is consistent with the grain-size distribution in
16 IDPs (Rietmeijer, 1993). Even CO chondrites, which have experienced varying degrees of
17 thermal metamorphism, can exhibit similar root-mean-square (rms) matrix grain-sizes (e.g.
18 Kainsaz has an average rms grain-size of 249 nm (Brearley, 1996)).

19 Cryoporometry results are consistent with the TEM data. Figure 4 shows a pore-volume
20 distribution for Acfer 094 derived using NMR cryoporometry. The majority of the porosity
21 in Acfer 094 is at extremely low pore diameters. From 200 nm down to the lowest pore
22 diameter currently measurable by this technique in these meteorites (~ 20 nm), the pore
23 volume distribution averages $3 \pm 1 \mu\text{l}.\text{nm}^{-1}.\text{g}^{-1}$, with no sign of a drop-off at low pore
24 diameters. Estimating the mean pore-size is difficult, as the smallest pores in Acfer 094 are

1 below the resolution of our measurements. However, a geometric mean pore-size in the
2 range 10–100 nm is reasonable based on the available data.

3 *4.2. Defining permeability for a chondritic precursor*

4 As noted previously, permeability is based on the hydraulic radius, which is derived from
5 measured pore- and grain-size and equation (1). The hydraulic radius is related to the
6 geometric mean pore-size by $r_H = d_P / \xi$, where ξ depends upon the pore shape (Paterson,
7 1983; Brace, 1977). For circular pores $\xi = 4$, while for slot-shaped pores of high aspect
8 ratio $\xi = 2$. Using a geometric mean pore-size of order 10–100 nm, and assuming a value of
9 $\xi = 4$, yields estimates for hydraulic radius of order 2.5–25 nm (consistent with grain sizes
10 in the range 20–200 nm), and a corresponding permeability of order 10^{-19} – 10^{-17} m² (0.1–10
11 μ D) for a porosity of 40% (eqn 1; Figure 1; Figure 2). A porosity of 40% is consistent with
12 w/r ratios derived from oxygen isotope studies (Clayton and Mayeda, 1984, 1999; Leshin et
13 al., 1997). Lower porosity estimates yield lower values of estimated permeability, but the
14 principle control on permeability is the hydraulic radius, and hence the mean pore- and
15 grain-size (eqn. 1).

16 Also shown in Figure 3 are permeability data for siltstones and claystones, which are
17 terrestrial analogues with comparable pore-sizes to meteorite samples. These lithologies
18 exhibit permeabilities of order 10^{-21} – 10^{-19} m² (1–100 nD; see Figure 1), and act as barriers
19 to flow over millions of years. A permeability of 10^{-19} – 10^{-17} m² for a chondritic precursor is
20 six orders-of-magnitude lower than that used in earlier numerical modelling studies. As
21 noted previously, the grain-sizes observed in Acfer 094 matrix are consistent with literature
22 data, both for this meteorite, and for other primitive extraterrestrial materials (e.g.
23 chondrites, IDPs, Stardust sample-return etc). Since grain-size is a fundamental control on

1 permeability, the observation of consistently small grain-sizes (10's – 100's nm) in the
2 most primordial materials supports the contention that permeability in chondritic precursors
3 was very low.

4 *4.3. Modelling fluid flow*

5 Using a gravity flow model (Grimm, 2007) as a starting point, and a permeability range
6 of 10^{-19} – 10^{-17} m², we can derive approximate lengthscales for liquid water transport as a
7 function of distance from the centre of a canonical asteroid (Figure 5). Given alteration
8 timescales of order 1Ma, and standard material properties (Young et al., 2003), this model
9 predicts buoyancy-driven water transport of 10's to 100's μ m – consistent with
10 petrographic studies (Krot et al., 1995; Grossman et al., 2002), and indicating essentially
11 isochemical alteration (Figure 5). Moreover, we can take a similar approach to that of
12 Young et al. (2003) to investigate the extent to which convective flow could occur, given
13 these new permeability values. Young et al. (2003) employed the standard 10^{-13} – 10^{-11} m²
14 permeability range, and showed that convection would have occurred in bodies >80km in
15 diameter (Figure 1 from Young et al. (2003)). However, if we take the permeability range
16 of 10^{-19} – 10^{-17} m² derived from our chondrite data, then even for bodies with 40% porosity,
17 and canonical ²⁶Al/²⁷Al, it is apparent that much larger objects are required before large-
18 scale convective water flow could be established (Figure 6): in this case, asteroid diameters
19 above ~440km.

20 In addition, we have taken a detailed numerical model of asteroidal alteration (Cohen and
21 Coker, 2000), and modified the input parameters, based (in part) on the above analysis. The
22 scenario shown in Figure 7 is for a 10 km diameter asteroid accreting at 3 AU, 1.25 Myrs
23 after CAI formation. The figure illustrates peak temperature as a function of radius. In this
24 model the asteroid composition is 20% forsterite, 15% enstatite, 40% water ice, initial voids

1 of less than 1%, and the remainder inert rock. Previously, using higher permeabilities, flow
2 occurred over several km's, and temperatures in most scenarios substantially exceeded
3 those expected for CC alteration (Cohen and Coker, 2000). Using these updated input
4 parameters, the same model (Cohen and Coker, 2000) shows essentially zero fluid flow
5 (less than a single cell size – consistent with the predictions of the gravity (Grimm, 2007)
6 and convection (Young et al., 2003) models when updated input parameters are used (e.g.
7 Figure 5)). In addition, we observe moderated peak temperatures throughout the centre of
8 the asteroid: while still high, temperatures are closer to those expected from oxygen isotope
9 studies of CC alteration (Clayton and Mayeda, 1984, 1999; Leshin et al., 1997). We are
10 currently exploring the additional effect of varying parameters such as composition, size,
11 timing of accretion, percentage void space, and surface boundary temperature, on peak
12 internal aqueous alteration temperatures.

13

14 **5. Discussion**

15 *5.1 Model permeability estimates compared to literature data*

16 Thermal, aqueous, and impact processing of meteorites can all act to modify porosity
17 (and potentially permeability), as can atmospheric entry and storage in the terrestrial
18 environment. With those caveats in mind, how do our permeability estimates for precursor
19 materials compare with measurements from carbonaceous chondrites? Sugiura et al. (1984)
20 measured gas permeabilities for three chondrites. One of these was a sample of Allende,
21 which was found to have a permeability of c. 10^{-16} m². Another recorded a permeability of
22 c. 10^{-15} m² (1 mD; 1 D = 9.87×10^{-13} m²); the permeability of a third was too small to
23 detect ($<10^{-21}$ m² or 1 nD). Unfortunately no description of the pore-space morphology of
24 the samples was provided. Corrigan et al. (1997) significantly expanded the permeability

1 dataset for chondrites, also measuring gas permeabilities. The average permeability for all
2 the meteorites analysed was $8.3 \times 10^{-17} \text{ m}^2$. CM chondrites – included in this average –
3 proved to be an analytical challenge due to the presence of cracks caused by desiccation
4 while on Earth (Corrigan et al., 1997 and pers. comm.), occasionally resulting in relatively
5 high permeabilities. For the most mineralogically primitive meteorites in their suite
6 (samples of Vigarano, Efremovka, Leoville – all reduced CV3s), Corrigan et al. (1997)
7 found an average permeability of $2.5 \times 10^{-18} \text{ m}^2$. Both the Sugiura et al. (1984) and
8 Corrigan et al. (1997) measurements likely represent an upper limit for liquid water
9 permeability, as the experiments were in the low pressure, molecular gas flow domain. It is
10 therefore encouraging that these data (particularly the permeabilities from the least
11 aqueously and thermally altered, most mineralogically primitive samples) appear to be in
12 excellent agreement with our estimated permeabilities of 10^{-19} – 10^{-17} m^2 for a chondritic
13 precursor material.

14 *5.2. Constraining fracture permeability*

15 We have constrained permeability based on the material properties (grain size, porosity,
16 pore-size distribution etc) of primitive meteorites. This is essentially a grain-scale
17 permeability, taking the rock as a homogeneous whole. As C1 and C2 chondrites are
18 distinguished by pervasive, homogeneous alteration of matrix at the grain-scale, the
19 implication is that grain-scale permeability was important. But what constraints can we
20 impose on the relative importance of fracture permeability?

21 In studying meteorites we are hampered by the obvious fact that we have hand samples –
22 we cannot go to the outcrop. Therefore, any discussion of the possible contribution of
23 fracture, or microfracture permeability is unlikely to lead to a definitive conclusion.
24 However, it is possible to impose some constraints. The nearest meteoriticists can get to an

1 outcrop-scale is in large CC falls. In the case of the CMs, the largest fall is Murchison
2 (>100kg). There are large numbers of hand specimens of this meteorite, but at the hand
3 specimen scale, abundant alteration-filled fractures are not observed (Figure 8a). At finer
4 scales, veins filled by secondary minerals have occasionally been described. However,
5 these features appear to be exceedingly rare. Figure 8b shows an energy dispersive element
6 map for a section of Murchison – alteration phases dominate matrix, but they are not
7 associated with veins. More detailed studies have come to a similar conclusion: veins
8 appear to be rare or absent in CM chondrites (Benedix et al. 2003; Tyra et al. 2009). The
9 same is true for CI chondrites. Carbonate and sulphate veins in Orgueil were once
10 considered to be evidence for aqueous flow. However, it is now clear that these features are
11 a product of terrestrial alteration (Gounelle and Zolensky, 2001).

12 The work of Corrigan et al. (1997) is also relevant here. The samples measured for
13 permeability were all relatively large hand samples, ranging in size from ~3 cm upto ~15
14 cm in some cases (Corrigan pers. comm.). Despite this, low permeabilities – consistent with
15 the grains-scale permeability derived above – were the norm. To summarise the
16 observational data, there does not appear to be evidence for pervasive interconnected
17 fractures at scales ranging from 10's- μm to 10's-cm in CCs, either from studies of
18 chondrite petrography, or permeability measurements.

19 Finally, the pervasive, homogeneous nature of alteration in C1 and C2 chondrites is
20 actually quite remarkable, and places its own constraint on the importance of fractures as
21 focussed conduits for flow. For the CM2s this homogeneity has recently been quantified
22 through X-ray diffraction measurements of modal mineralogy (Howard et al., 2009). Even
23 including meteorites that are considered highly altered (e.g. Nogoya and Cold Bokkeveld),
24 modal total phyllosilicate only varied by a few % amongst CM2 falls (the total range was

1 73–79%). If flow had occurred through focussed conduits, it would not result in
2 homogeneous alteration of matrix. Rather, alteration would be concentrated around
3 fractures. In addition, significant interchange of fluid between the fracture network and
4 enclosed blocks would be severely limited by the low permeability of matrix – once again,
5 grain-scale permeability imposes a fundamental constraint. As we have shown, grain-scale
6 permeability in CCs is low, so transport lengthscales are short (100’s μm at most, even
7 given alteration timescales of 1Ma). To produce uniform wholesale alteration of the bulk
8 rock, the density of microfractures would need to be of this order. The only remaining
9 possibility for significant fracture permeability would be a much larger scale fracture
10 network, effectively decoupled from individual meteorite-sized blocks. This is not excluded
11 by our data, however, we note that (by definition) it is not testable through meteorite
12 sample analysis. In addition, it does not appear to be required (in terms of stabilising parent
13 body temperatures) based on numerical modelling (Figure 7).

14 *5.3. Implications for fluid flow*

15 The permeability estimates that we derive for a chondritic precursor are in agreement
16 with permeability measurements from mineralogically primitive chondrites (Corrigan et al.
17 1997). Given that we do not see compelling evidence for a pervasive interconnected
18 fracture network at length scales that would be effective at producing homogeneous
19 alteration of CCs, our conclusion is that grain-scale permeability is dominant, and that
20 permeability values in the range 10^{-19} – 10^{-17} m^2 for a porosity of 40% are appropriate for
21 chondritic asteroids at the onset of aqueous alteration. Applying these permeability
22 estimates in numerical models, we find buoyancy-driven water transport over distances of
23 100’s μm at most, and similar short distances for element mobility via diffusion. We note
24 that these predicted flow / transport distances are in excellent agreement with distances for

1 metasomatism from petrographic observations of chondrites. Fundamentally, isochemical
2 alteration, with minimal fluid flow, is not a special circumstance. It is unavoidable, once we
3 consider the material properties of these rocks. The extreme fine grain size of chondritic
4 matrix results in very low permeability, which results in minimal flow, even where w/r
5 ratios were high. Clearly this imposes constraints on a number of existing models.

6 *5.4. Why are the most chemically primitive rocks also the most altered?*

7 Fine-grained condensates would have hosted volatile elements, including water.
8 Assuming that water was initially present as ice rinds on matrix grains, then water-saturated
9 porosity in the bulk rock would scale with matrix abundance. The most primitive rocks are
10 100% fine-grained matrix. They will be the most altered, but permeabilities are such that
11 fluid flow would be minimal, even when calculated w/r ratios are high (Clayton and
12 Mayeda, 1984, 1999; Leshin et al., 1997). The greater the proportion of fine-grained
13 material in a chondrite, the closer it is to a primitive ‘solar’ composition, but also the more
14 water was available for alteration.

15

16 **6. Conclusions**

17 We have studied the pore- and grain-size distribution in a carbonaceous chondrite that has
18 experienced minimal thermal and aqueous alteration. Both datasets are consistent,
19 indicating a geometric mean pore-size of 5–50 nm, a hydraulic radius of order 2-20 nm
20 (consistent with grain sizes in the range 20-200 nm), and a permeability in the range 10^{-19} –
21 10^{-17} m² (for a porosity of 40%). We note these grain sizes are also consistent with previous
22 studies of other primitive extraterrestrial materials (primitive chondrites, IDPs, and Stardust
23 sample-return materials), and that our estimated permeability range is consistent with
24 measurements from primitive chondrites (Corrigan et al., 1997). Assuming these new

1 permeability values are representative of a ‘canonical’ chondritic precursor material, our
2 modelling shows that, even at the upper permeability bound (10^{-17} m^2 , for matrix grain sizes
3 of order 100’s nm), convective flow would not occur unless the asteroid diameter exceeded
4 ~440km. In addition, we show that lengthscales for liquid water transport are in the range
5 10’s - 100’s μm (even with alteration timescales of order 1Ma), indicating essentially
6 isochemical alteration, and consistent with meteorite petrography, geochemistry, and a
7 closed-system oxygen isotope model (e.g. Clayton and Mayeda, 1984, 1999; Leshin et al.,
8 1997).

9 In summary, our analysis resolves the paradox of how primitive chemistry can co-exist
10 with altered mineralogy; how we can have oxygen isotopic evidence for high *w/r* ratios
11 despite minimal evidence for flow; why previous numerical modelling studies have
12 produced results that appear to be at odds with meteorite chemistry and petrography; and
13 why the most primitive rocks are also the most altered.

14

15 **Acknowledgements**

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17 Facilities Council (STFC) under grant number PPA/G/S/2003/00071.

18

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- 3

1 **Figure captions**

2 **Fig. 1.** Permeability / porosity as a function of hydraulic radius. Data are shown for a variety of terrestrial
3 porous media (Doyen, 1988; Bourbie and Zinszner, 1985; Paterson, 1983; Mortensen et al., 1998;
4 Hildenbrand et al., 2002), and predicted using the Carman-Kozeny equation (1). The solid line denotes the
5 predicted permeability for a value of $h_{ck} = 5$, appropriate for unconsolidated and some consolidated porous
6 media. The dashed line denotes the predicted permeability for a value of $h_{ck} = 100$, appropriate for some
7 consolidated sandstones and siltstones. The shaded region denotes the estimated hydraulic radius for
8 meteorite samples.

9
10 **Fig. 2.** Permeability vs. porosity for a range of grain sizes, and a value of $h_{ck} = 50$ (intermediate between some
11 unconsolidated media and consolidated siltstone (Figure 1)). This figure illustrates graphically the relationship
12 between permeability, porosity, pore geometry, and grain size defined in eqn. (3).

13
14 **Fig. 3.** A selection of transmission electron microscopy (TEM) images showing the matrix of Acfer 094.
15 TEM analysis followed focussed ion beam lift-out of two portions of fine-grained matrix material from the
16 carbonaceous chondrite Acfer 094. FIB sections covered a total area of $\sim 110 \mu\text{m}^2$. A variety of matrix grains
17 can be distinguished (in these images, coherent features which have a relatively constant grey-scale), set
18 against a background of amorphous and finer-grained material. The great majority of grains are smaller than
19 200 nm in diameter.

20
21 **Fig. 4.** Nuclear magnetic resonance cryoporometry data. Pore volume distribution vs pore diameter for the
22 carbonaceous chondrite Acfer 094. The data are consistent with the TEM results, and indicate that the bulk of
23 the porosity in Acfer 094 is at very low pore diameters.

24
25 **Fig. 5.** Modelling asteroidal alteration. Transport lengthscale as a function of distance from a model asteroid
26 centre. Results for permeabilities (k) of 10^{-17} m^2 and 10^{-19} m^2 are shown, using a gravity flow model (Grimm,
27 2007), standard material properties (Young et al., 2003), a porosity of 40%, and assuming an alteration
28 timescale of 1Ma. The shaded region denotes the approximate lengthscale for metasomatism observed in
29 altered carbonaceous chondrites, derived from a range of petrographic studies.

30

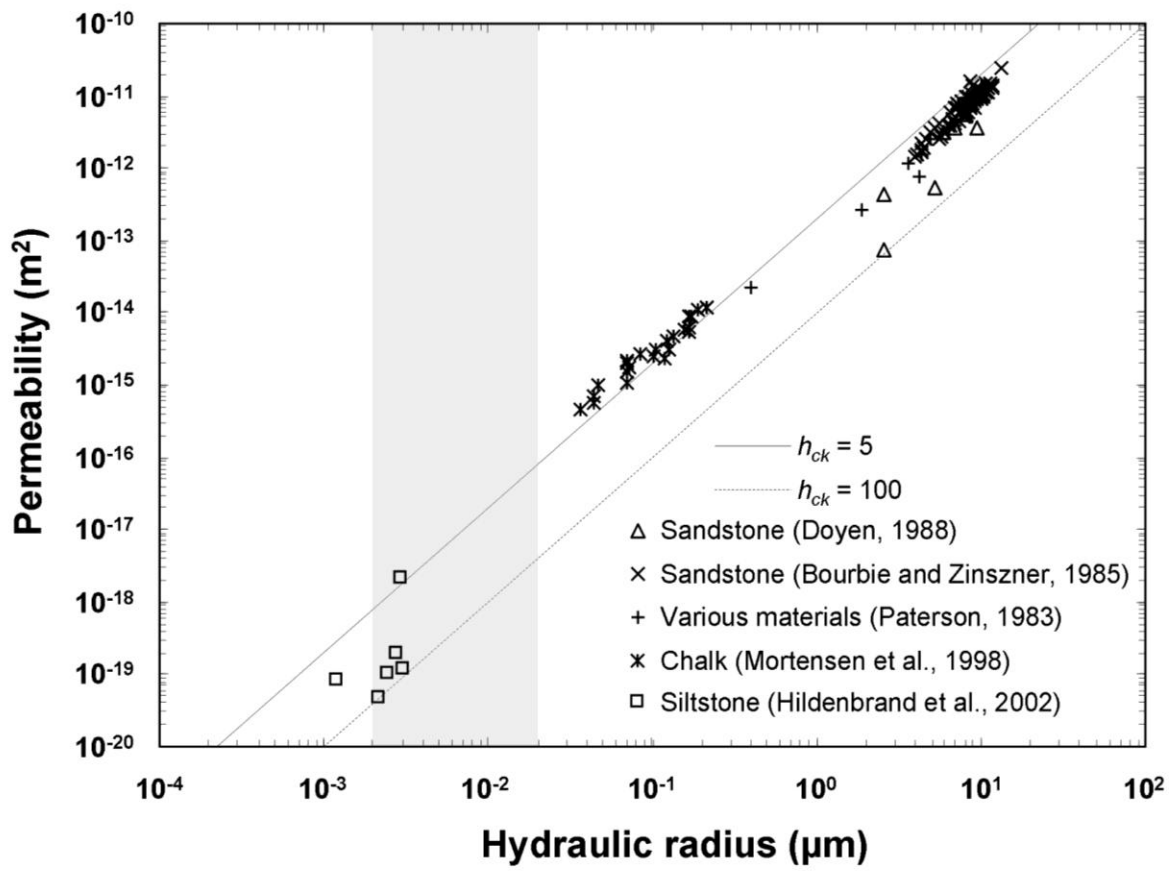
1 **Fig. 6.** Modelling asteroidal alteration. The range of critical radii for the onset of convective flow, for a
2 spherical asteroid with 40% given heat production from radionuclide decay (the ‘canonical’ initial Solar
3 System $^{26}\text{Al}/^{27}\text{Al}$ value is taken to be $\sim 5 \times 10^{-5}$). Contours are for permeabilities of 10^{-19} and 10^{-17} m^2 , derived
4 from our analyses of the primitive chondrite Acfer 094. It is apparent that, even at the upper permeability
5 bound of 10^{-17} m^2 , objects with diameters in excess of 440km are required before large-scale convective water
6 flow could be established.

7
8 **Fig. 7.** Modelling asteroidal alteration. Temperature as a function of distance from a model asteroid centre.
9 Results are shown for permeabilities based on eqn. 3, using a 1000-cell Cohen and Coker (2000) type model,
10 and including the effects of gas venting. The heat source is radionuclide decay, and the model uses a Cassen-
11 based time-dependent surface temperature boundary condition (Cassen, 1994). Additional details can be
12 found in Cohen and Coker (2000).

13
14 **Fig. 8.** Murchison CM2 chondrite, in hand specimen and thin section. (a) A hand specimen, $\sim 10\text{cm}$ across
15 (courtesy Linda Welzenbach, Smithsonian Institution). (b) Energy dispersive element map of a thin section of
16 Murchison. These images illustrate the homogeneous nature of alteration in CM chondrites. Neither here, nor
17 in the literature do we find evidence for a pervasive fracture network.

18

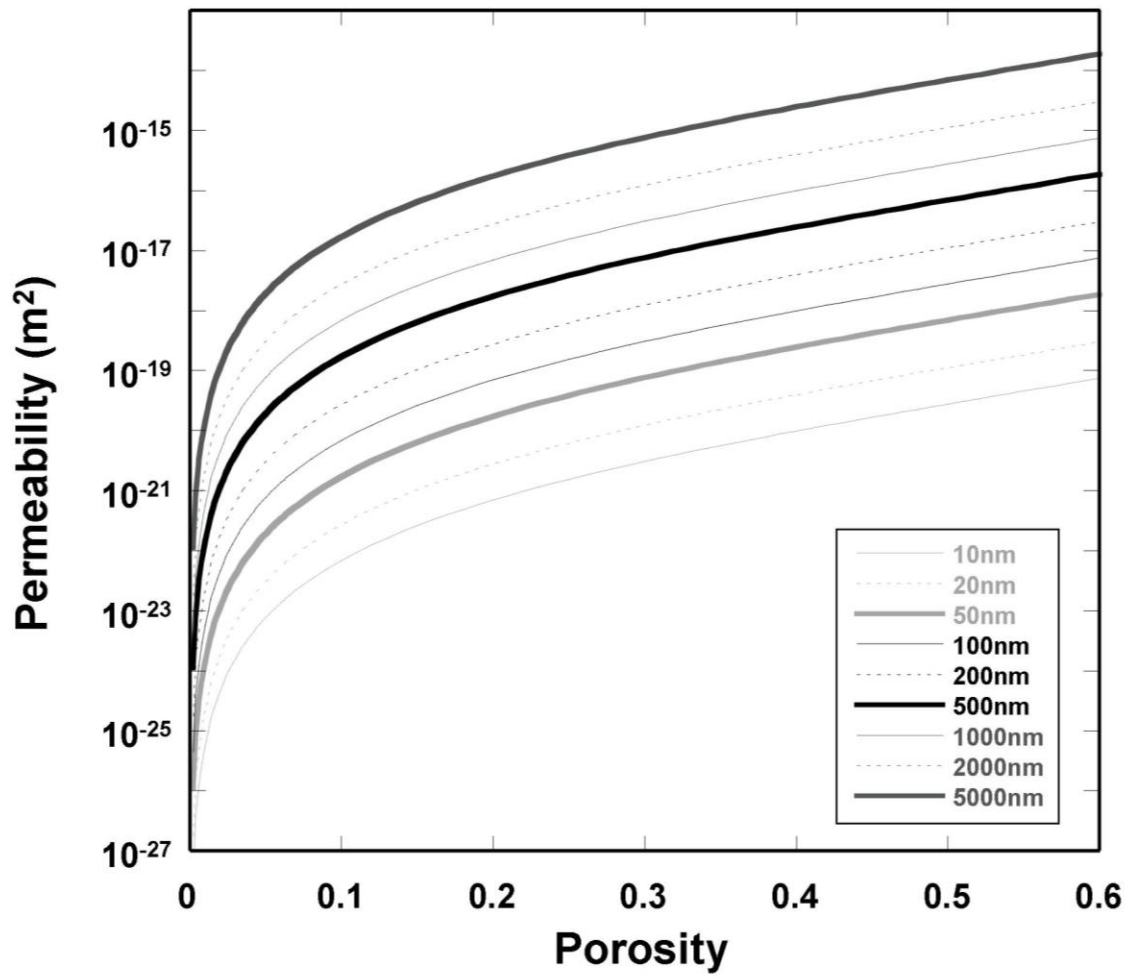
1 Figure 1



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3

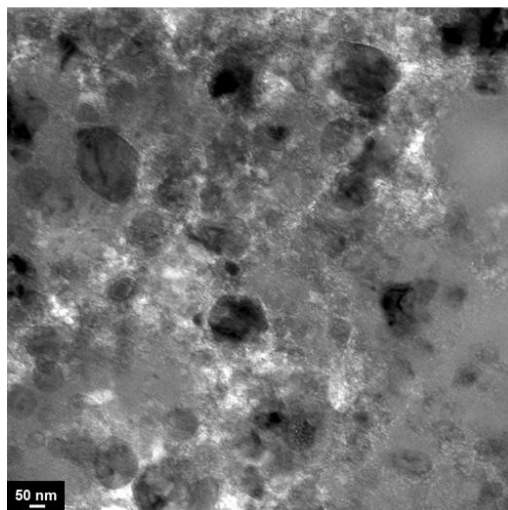
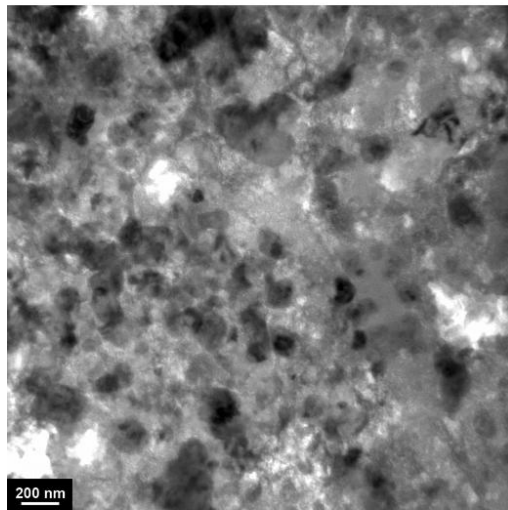
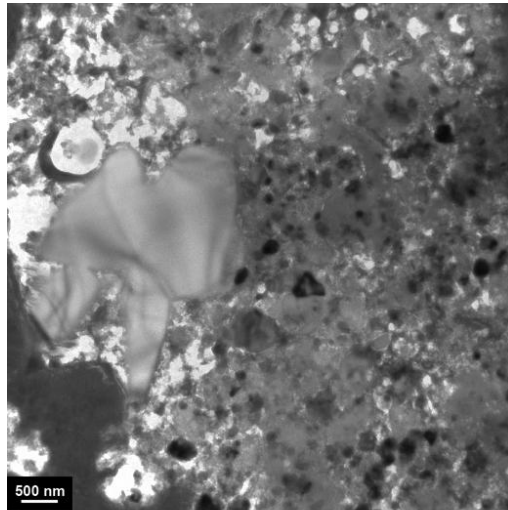
1 Figure 2



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3

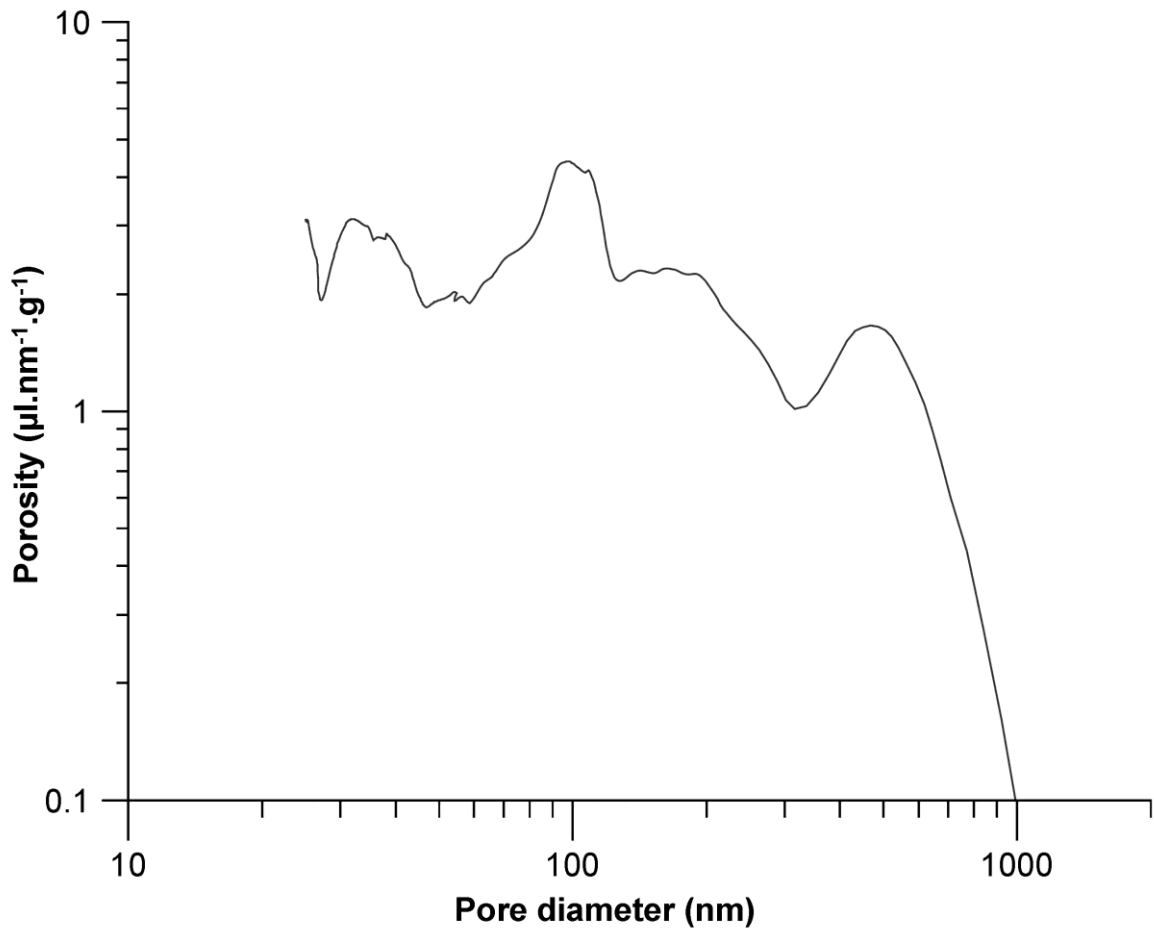
1 **Figure 3**



2

3

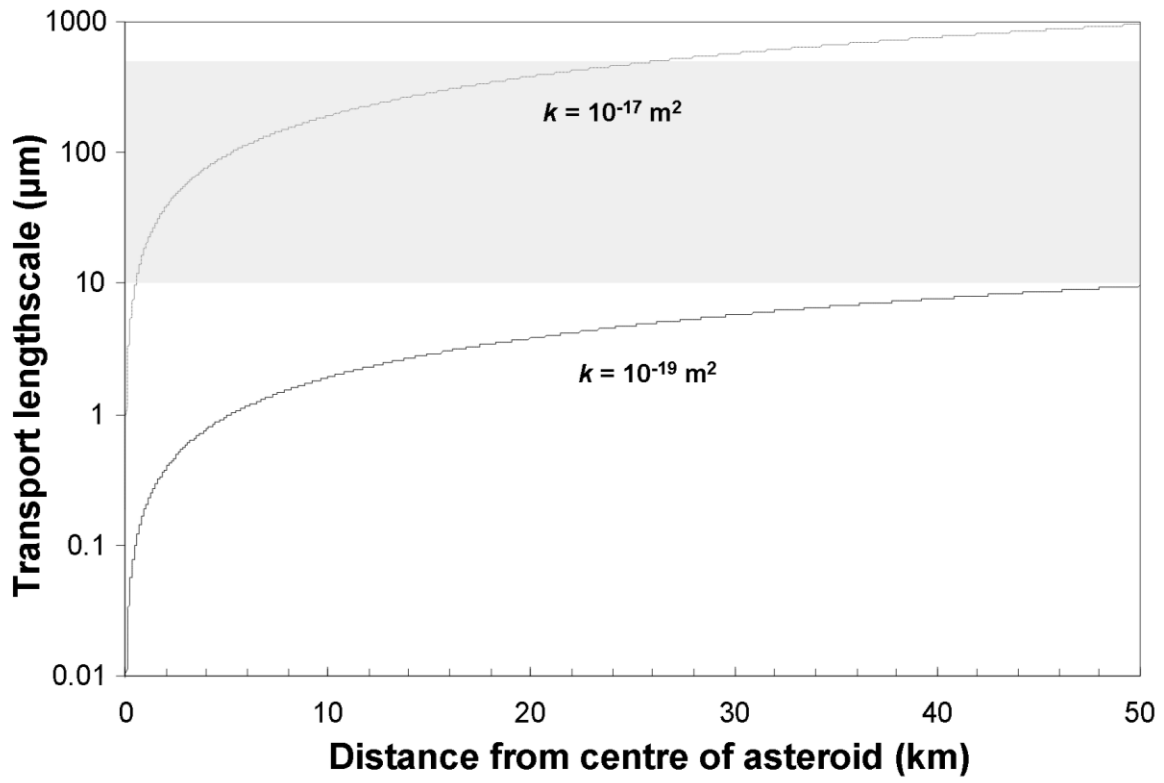
1 **Figure 4**



2

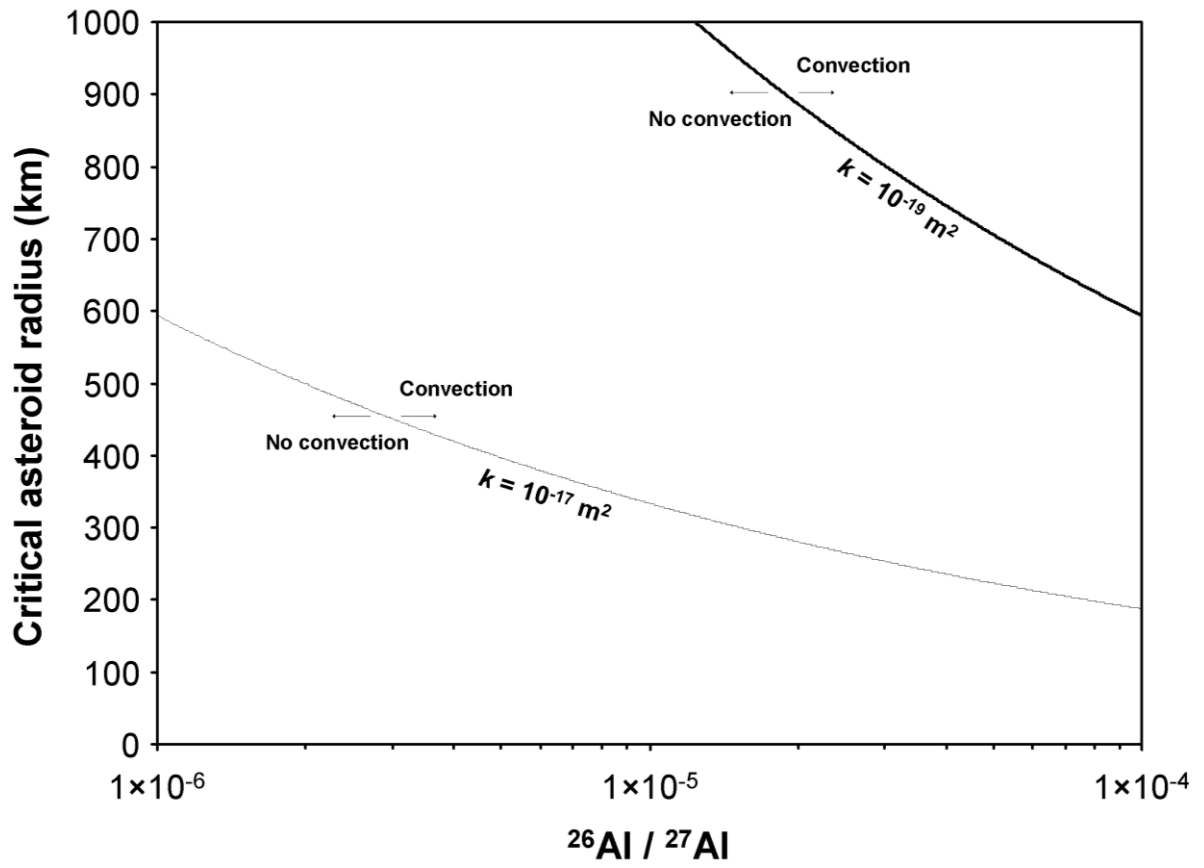
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1 **Figure 5**



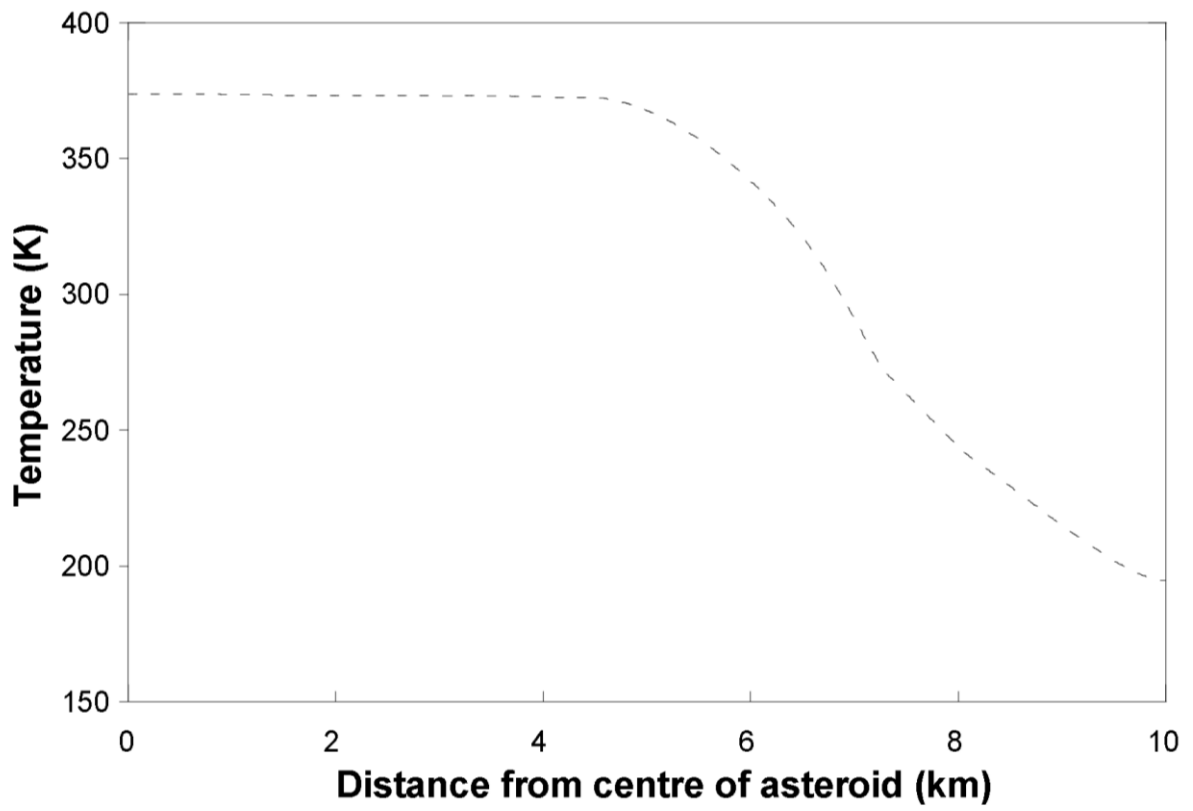
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1 Figure 6



2

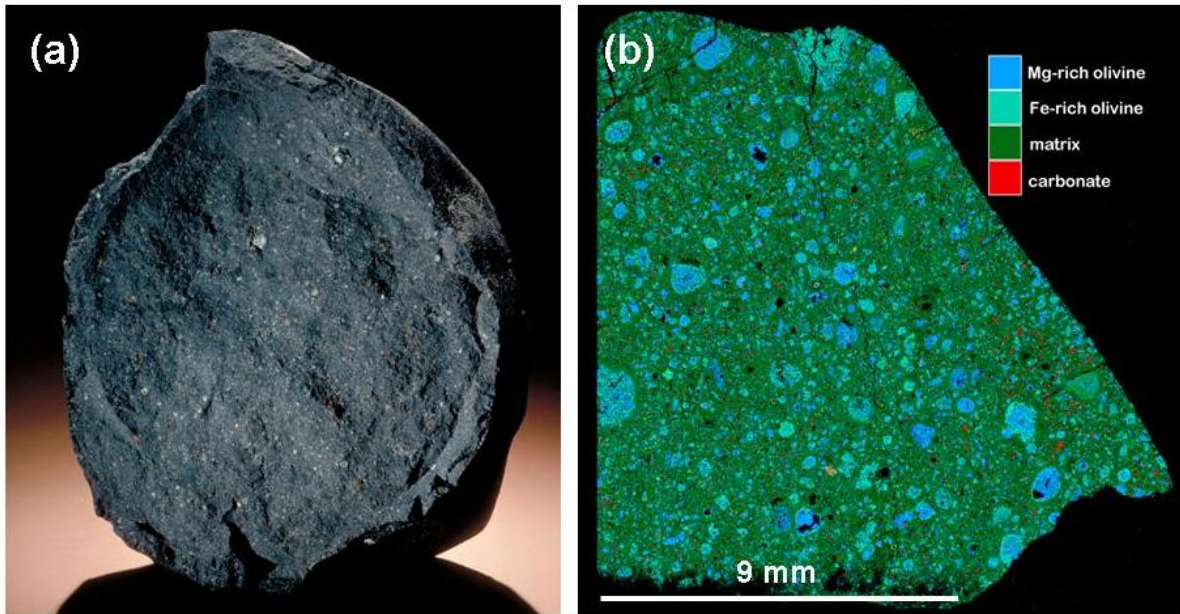
1 **Figure 7**



2

3

1 Figure 8



2