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Synthesis and Characterisation of Analogues for Interplanetary Dust and Meteoric Smoke Particles

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Abstract

Analogues have been developed and characterised for both interplanetary dust and meteoric smoke particles. These include amorphous materials with elemental compositions similar to the olivine mineral solid solution series, a variety of iron oxides, undifferentiated meteorites (chondrites) and minerals which can be considered good terrestrial proxies to some phases present in meteorites. The products have been subjected to a suite of analytical techniques to demonstrate their suitability as analogues for the target materials.

Keywords: Interplanetary Dust; Meteoric Smoke; Micrometeorites; Sol-gel synthesis

1. Introduction

Many processes in atmospheric environments are controlled by the availability of reactive or catalytic surfaces which originate from outside that atmosphere [Nachbar, et al., 2016; Petrie, 2004; Plane, 2012; Turco, et al., 1981]. However, this extraterrestrial material is only partially characterised and understood in terms of these various atmospheric processes. Meanwhile, collecting the relatively large quantities required for surface science experiments (often grams to kilograms) from the atmosphere presents an extremely large engineering challenge [Hedin, et al., 2014]. In order to investigate and understand such atmospheric processes, it is therefore vital to employ suitable analogue materials which will mimic the behaviour of the environmental material.

The majority of dust which enters planetary atmospheres in our solar system originates from either Jupiter Family Comets (JFCs), the Asteroid Belt (AB), Halley Type Comets (HTCs) or Oort Cloud Comets (OCCs) [Nesvorný, et al., 2011; Nesvorný, et al., 2010]. To determine the appropriate material to mimic the atmospheric behaviour of Interplanetary Dust Particles (IDPs), the nature of such dust before encountering an atmosphere and the processes it will undergo during atmospheric entry must be considered.
1.1. Composition, morphology and size distribution of Interplanetary Dust

The primary source of information about the composition and morphology of IDPs are the large collections of micrometeorites retrieved in the stratosphere [Rietmeijer, 2000], the ice caps [Maurette, et al., 1987], Taylor, et al., 1998], deserts [Kohout, et al., 2014] and deep sea sediments [Blanchard, et al., 1980]. While these collections include particles exhibiting different degrees of thermal alteration (ranging from unmelted particles to cosmic spherules) and weathering, it has been possible to establish their general properties before aerobraking [Jessberger, et al., 2001], Kohout, et al., 2014]. IDPs size and composition have also been investigated by particle detectors on board orbital spacecraft such as the Long Duration Exposure Facility (LDEF) [Love and Brownlee, 1993] and the MIR Space Station [Hörz, et al., 2000], where the major difficulty has been characterising the effect of the high speed impacts (v > 11 km s\(^{-1}\)) on the exposed materials (e.g. aluminium, aerogel). Several past and ongoing space missions such as Stardust [Hörz, et al., 2006], Deep Impact [Lisse, et al., 2006] and Rosetta [Schulz, et al., 2015] have gathered information close to cometary sources of dust. Stardust is the only mission to date which captured dust particles ejected from a comet (81P Wild 2) and, after deceleration and disruption in the aerogel (SiO\(_2\)) collector [Trigo-Rodríguez and Llorca, 2007], returned them to Earth. Study of the aerogel keystones has allowed a thorough laboratory chemical and mineralogical characterisation of the particles [Frank, et al., 2014], Stephan, et al., 2008]. Rosetta currently analyses \textit{in situ} the composition, morphology and optical properties of particles released by comet 67P Churyumov-Gerasimenko [Hilchenbach, et al., 2016]. A caveat to these close-to-source studies is that a particular comet is only a sample among a population of millions. Comparison of Antarctic micrometeorites and Stardust samples shows clear similarities, but also differences [Dobrică, et al., 2009]. Astronomical observations and modelling have provided key information about the spatial distribution, the sources and the size distribution of the interplanetary dust forming the so-called Zodiacal Cloud [Ade, et al., 2014], Fixsen and Dwek, 2002].

The meteoroid particles which contribute most to the meteoric input to the Earth’s atmosphere are 10-50 µm in radius (0.01 to 1 µg assuming 3 g cm\(^{-3}\) density) [Carrillo-Sánchez, et al., 2015], Nesvorný, et al., 2011]. The analysis of the Antarctic collections has shown that IDPs are consistent with the Carbonaceous Ivuna (CI) chondritic composition, while mineralogically IDPs are closer to the Carbonaceous Mighei (CM) chondritic composition [Jessberger, et al., 2001], Taylor, et al., 2012]. Analysis of the more than 5000 micrometeorite (MM) particles of the 2000 South Pole Water Well collection, described by Taylor, et al. (2012) indicates that ~75% of IDPs are fine grained aggregates of CI-CM elemental composition, 10% coarse grain anhydrous minerals and less than 10% ordinary chondrites (OC). Less than 1% of the MMs have achondritic compositions, CAI components, or recognizable chondrules. Besides the low abundance of OC IDPs, they also show important compositional and morphologically differences with respect to carbonaceous...
chondritic (CC) meteorites. IDPs are extremely fine-grained aggregates, such that despite of the mineralogical heterogeneity of the sub-micron sized grains, the bulk chemistry matches CI and CM composition as a whole. By contrast, the matrix of carbonaceous meteorites is composed of micron-sized coarse grained aggregates, single mineral grains, chondrules, etc., and therefore a section of a meteorite of the same size of an IDP frequently does not match the bulk elemental composition [Jessberger, et al., 2001]. The porosity of IDPs is in line with that of CI and CM meteorites, with bulk densities around 2 g cm\(^{-3}\) [Jessberger, et al., 2001]. OC meteorites have bulk densities higher than 3 g cm\(^{-3}\) because they have experienced mild thermal metamorphism that reduces the pores [Consolmagno, et al., 2008].

1.2. Ablation on Atmospheric Entry

Upon encountering an atmosphere IDPs heat frictionally, often to the point of melting and evaporating, forming a meteor. Ablation occurs both through sputtering by atmospheric and, if the particle melts, rapid evaporation of the constituent elements. This process is known as meteoric ablation. The majority of the incoming particles are too small to generate visually detectable meteoroids (particles need to be larger than 1 mm radius), but ionization of the ablated atoms by hyperthermal collisions with atmospheric molecules forms electrons which enable detection by radar, either through specular reflection from the resulting electron trail or by incoherent scatter from the ball of electrons around the meteoroid – termed the head echo (e.g. [Janches, et al., 2009]). The ablation rate of a particular meteoroid is a function of its mass, entry velocity and incident angle [Vondrak, et al., 2008]. Furthermore, the different melting points and volatilities of the various components of meteoroids result in a process known as differential ablation, whereby the most volatile components ablate first [Janches, et al., 2009, Trigo-Rodríguez and Llorca, 2007, Vondrak, et al., 2008]. An unablated particle will then sediment through the atmosphere under gravity and, if small enough (<1 μm for Earth) be transported by the general atmospheric circulation and eventually reach the surface, where it is termed a micrometeorite (MM). Partially ablated material has been identified on the Earth’s surface in the form of scoriaceous MMs and cosmic spherules, (which form when a particle melts but does not completely ablate) [Blanchard, et al., 1980, Kohout, et al., 2014, Taylor, et al., 1998]. The vaporized elements give rise to the layers of metallic atoms and ions in the mesosphere and lower thermosphere [Plane, et al., 2015].

1.3. Meteoric Smoke Formation and properties

The atmospheric chemistry of meteor-ablated metals depends on the atmosphere in question [Plane, et al., 2015, Whalley and Plane, 2010]. Gas-phase reactions produce stable reservoirs which can then polymerise into nanoparticles known as meteoric smoke particles (MSPs) [Hunten, et al., 1980, Plane, 2003]. In general the most stable reservoir for a metal, and therefore the composition of MSP, will depend on the reactivity of that metal and its derivatives with O\(_3\) and O\(_2\) to give oxides, H\(_2\)O to give hydroxides, Si(OH)\(_4\) and SiO\(_2\) to give
silicates and CO$_2$ to give carbonate and bicarbonates, as well as the stability of these compounds with respect to O and H atom reactions which reduce them back to metal atoms (Plane, et al., 2015). Based on laboratory experiments MSPs should then grow by rapid agglomeration as they are transported through an atmosphere (Saunders and Plane, 2006). In the case of the Earth’s atmosphere, MSPs are modelled to reach a radius of around 100 nm in the stratosphere (Bardeen, et al., 2008).

Here MSPs and IDPs are collectively referred to as meteoric material. Particles collected by aircraft in the stratosphere are often termed IDPs (Bigg, 2012), whilst those collected at the surface are labelled MMs (Taylor, et al., 2012). In order to determine the most appropriate analogue material, the key difference is between ablated and unablated material. In this paper, IDPs should be taken to refer to unablated material, MSPs to ablated matter, and MMs specifically to unablated material which sediments to the surface.

MSPs have been detected in a variety of ways. In the Earth’s atmosphere MSPs occur in a weak plasma and so a small fraction are charged by electron attachment, which enables them to be detected using rocket-borne Faraday cup detectors (Plane, et al., 2014, Rapp, et al., 2012, Robertson, et al., 2014). These detectors can be coupled to photo-ionisation flash lamps so that neutral particles can be detected simultaneously (Rapp, et al., 2012). However, attempts to capture and return MSPs for detailed analysis have had very limited success so far (Hedin, et al., 2014). Particles consisting of compact and chain aggregates have been retrieved from the upper stratosphere and the mesosphere (Bigg, 2012, Farlow, et al., 1970, Witt, et al., 1964). The chain aggregates are reminiscent of fractal-like laboratory-generated analogues (Saunders and Plane, 2006), but direct identification and characterisation of MSPs particles has been largely inconclusive. MSPs have also been observed by measuring their optical extinction using the SOFIE spectrometer on the AIM satellite (Russell, et al., 2009), but the composition of the particles are not well constrained (Hervig, et al., 2009). Observations of polar mesospheric clouds by SOFIE indicate the presence of MSPs in the cloud ice particles that could be consistent with wüstite (FeO), or magnesiowüstite (Mg$_x$Fe$_{1-x}$O, x = 0.1 – 0.6). Such investigation is limited by the knowledge of refractive indexes, with comparison only possible to bulk, crystalline materials. The likely amorphous nature of MSPs (Saunders and Plane, 2011) and further their fractal-like morphology significantly complicates understanding of their extinction properties (Sorensen, 2001).

### 1.4. Impacts of Meteoric Materials

The potential impacts of meteoric materials – both unablated material and MSPs - can be grouped into three categories. First is the removal of acids such as H$_2$SO$_4$ and HNO$_3$ (Frankland, et al., 2015, Saunders, et al., 2012), which undergo acid-base reactions with the metallic species in the particles. Second is the particles providing surfaces for heterogeneous chemistry, including the conversion of HO$_2$ into H$_2$O$_2$ in the Earth’s stratosphere (James, et al., 2016 - Unpublished results), the cyclo-trimerization of C$_2$H$_2$ into
C₆H₆ in Titan’s troposphere \cite{Frankland, et al., 2016 - in press}, and the oxidation of CO in Venus’s lower atmosphere \cite{Frankland, et al., 2016 - Unpublished results}. Third is the particles acting as heterogeneous nuclei, either via direct deposition of a supersaturated vapour such as H₂O or CO₂ onto the meteoric particles \cite{Nachbar, et al., 2016} \cite{Plane, 2011}, or via immersion of the particles in a liquid droplet, such as a H₂SO₄ droplet in the Earth’s stratosphere \cite{Hoyle, et al., 2013}.

Although the radiative forcing of MSPs is almost certainly negligible under present day conditions, MSPs may have played a larger climactic role during the Earth’s early history \cite{Saunders, et al., 2007}. In any case, MSP extinction needs to be taken into account when interpreting optical extinction measurements in the upper stratosphere \cite{Neely, et al., 2011}.

Investigating this wide range of processes involving extraterrestrial material requires access to appropriate analogues in the laboratory. In this paper the synthesis and characterisation of a number of analogues for IDPs and MSPs is reported and compared to those in the literature. The main aim of this study is therefore to lay the groundwork for future studies which can quantify the various impacts of meteoric material in planetary atmospheres. The paper is intended for atmospheric scientists, to introduce concepts which largely come from the field of meteoritics, and to aid the choice of materials in future studies of atmospheric processes.

2. Preparation of Analogues

Analogues have been prepared both by processing mineral and meteorite samples, and by chemical synthesis from appropriate precursors. The genesis of the various samples is described in Table 1 along with their proposed application, specific surface area and mass density as appropriate. Note that throughout this text any crystalline materials will be referred to by their names, whilst chemical compositions will be used for materials without significant crystal structure.

2.1. Meteoritic Material

Samples of the Allende (CV3) \cite{Clarke, et al., 1971}, Murchison (CM2) \cite{Fuchs, et al., 1973}, North West Africa 5515 (NWA, CK4) \cite{Weisberg, et al., 2009} and Chergach (H5) \cite{Weisberg, et al., 2008} meteorites were ground by hand using a pestle and mortar and then sieved (Endecottes test sieves, pore sizes of 38, 106, 150, 250 and 355 µm) to obtain size fractions relevant for use as IDP analogues (tens to hundreds of µm radius \cite{Bardeen, et al., 2008} \cite{Bigg, 2012} \cite{Carrillo-Sánchez, et al., 2015}). These meteorites represent several carbonaceous groups and one ordinary chondrite. Allende and Murchison have notably been used in a number of previous studies investigating IDP behaviour whilst NWA and Chergach have been included to represent outlier groups in the likely IDP flux (see Section 3.6.) \cite{Burchell, et al., 2006} \cite{Court and Sephton, 2011} \cite{Toppani, et al., 2001}.
2.2 Terrestrial Minerals

Terrestrial minerals were also used here as analogues for fine and coarse-grained anhydrous IDP material. These were obtained by grinding peridot olivine, labradorite, anorthite and albite followed by size separation. These minerals were chosen as example members of solution series. Other members of such series or other minerals could also be used, either alone or in mixtures, to obtain a wider range of compositions. Similar terrestrial minerals have been used in the past to investigate IDP processing. These include simulated changes in pyrrhotites during aerobraking e.g. Greshake, et al., 1998 and olivine and pyroxene as a thermometer for the temperatures reached by meteors Sandford and Bradley, 1989.

2.3 Synthetic Samples

Sol-gel synthetic routes to produce both crystalline mineral samples and amorphous materials of suitable composition were also used. Directly synthesised compounds include amorphous materials with compositions covering the olivine solid solution series \((\text{Mg}_{x}\text{Fe}_{2-x}\text{SiO}_4\text{ where }0 \leq x \leq 2)\) and goethite. These can be annealed to produce secondary products including hematite and mixtures of crystalline phases such as enstatite and hematite.

\(\text{Mg}_{x}\text{Fe}_{2-x}\text{SiO}_4\) compounds were synthesised by stirring at room temperature for 7 days a mixture of stoichiometric amounts (relative to 0.1 mol l\(^{-1}\) product) of \(\text{MgCl}_2\) (Aldrich), \(\text{Fe(SO}_4\text{)}_2\text{(NH}_4\text{)}_2\) (Sigma-Aldrich) and \(\text{Na}_4\text{SiO}_4\) (Alfa Aesar) [Frankland, et al., 2015]. Solutions immediately turn blue when Fe is present but otherwise are colourless. All solutions are viscous but do not pass the inversion test. After purification, annealing of this amorphous product at 1273 K for 24 hours was also used to generate a mixture crystalline enstatite and hematite.

Here we report a novel method to produce pure goethite and, by processing this, hematite. Goethite was obtained by stirring a solution of 0.1 mol l\(^{-1}\) each of \(\text{Fe(SO}_4\text{)}_2\text{(NH}_4\text{)}_2\) and \(\text{NaOH}\) for 3 days with a flow of compressed air bubbling through the reacting solution. The purified goethite product of this reaction was dehydrated by annealing at 573 K for 24 hours to produce hematite. All products of room temperature reactions were collected and dried in petri dishes. Residual byproducts were removed using repeated dialysis (at least 15 hours total) in a Soxhlet apparatus with the particles held in water permeable tubing (Snakeskin 7000 MWCO).

Purified products, along with samples of meteoritic and terrestrial materials were then subjected to a suite of characterisation techniques in order to assess their appropriateness as analogues for IDPs and MSPs. Mass yields after purification are >90 %, although the possibility that \(\text{H}_2\text{O}\) is adsorbed to the large surface area of the synthetic particles adds an upward bias to such measurements. In any case, approximately 20 g of \(\text{Mg}_{x}\text{Fe}_{2-x}\text{SiO}_4\) and 3-5 g goethite can be produced in a typical 2 l batch synthesis, providing sufficient material for characterisation and further studies.
3. Characterisation of Analogues

A thorough characterisation of each of the products described above is required to inform their use as analogues for IDPs and/or MSPs. The properties which are required for a given analogue depend on the application to which it is put. This is compounded by the differing size scales of IDPs (up to several tens to hundred μm radius) and MSPs (single molecules which polymerise and agglomerate to several hundred nm radius), which restricts somewhat the techniques available for the characterisation of each analogue.

Where ablation is examined, it is important to understand the elemental composition and crystal structure, particularly as this has implications for melting and evaporation. In studies of catalysis, however, the surface properties of a sample are of paramount importance. Such properties have been investigated for samples of the products discussed in Section 2.

3.1. Elemental composition

The elemental composition of a material has clear relevance to studies of virtually all properties of meteoric material, and is arguably the most widely applicable characterisation of both environmental and analogue samples. Taylor, et al. (2012) examined around 5000 MMs from the South Polar Water Well and showed that IDPs have likely compositions similar to CI or CM meteorites. Whilst there is considerable uncertainty in the mineral composition of MSPs e.g. Hervig, et al., 2012, the elemental ratios of the metals (primarily Fe, Mg) and SiO₂ available for their formation are relatively well known Plane, et al., 2015.

Elemental compositions were measured by Energy Dispersive X-Ray Spectroscopy in combination with Scanning Electron Microscopy (SEM-EDX, Joel JSM 6610LV coupled to an Oxford Instruments INCA X max80 EDS). The carbon signal is omitted due to interference from carbon tape used as an SEM substrate, while the oxygen signal is affected by surface adsorbed water. This technique is limited by statistical uncertainty, since values are measured for individual particles or agglomerates rather than large, representative samples, and by reduced sensitivity to lighter elements. Average values with a 95% standard error (typically from 4-8 measurements) are therefore listed. Inductively Coupled Plasma – Atomic Emission Spectroscopy (ICP-AES, ICAP 6500 ThermoElectron) has been used for two meteoritic samples. Solutions were prepared from approximately 0.025 g of each sample fluxed with 0.05 g of Li-metaborate and dissolved in 30 ml HNO₃ 1 molal and 1 drop of HF. Four standard reference materials provided by the US Geological Survey were used for external calibration; internal calibration of the equipment was carried out before the measurements and rhodium was used as internal standard. Three determinations of the elemental composition were carried out and averaged for each meteorite, showing standard errors lower than 5% for most elements.

3.1.1. Meteoric Material

Several size fractions (particles of <19 μm and >177.5 μm radii) of the Chergach and NWA (particles of 19-53 μm and 125-177.5 μm radii) meteorites were analysed by SEM-EDX. The
composition of the Allende and Murchison meteorite samples were measured by ICP-AES. Elemental compositions of all four meteorites are compared to CI, CM composition and to their respective groups [Hutchison, 2004] in Figure 1. Note that the differences between standard compositions are generally smaller than the standard errors in the compositions. This is due to the coarse grained nature of meteorites in general. After grinding, grains of individual minerals distort the measured composition away from the mean of a larger sample.

The Chergach meteorite, shown in Figure 1 a), Si, Cr, Mn, Mg, Ca, K and P all agree within error with the standard compositions. Na and Al are enhanced (although Al is in agreement with the CM compositions) suggesting that the sample contained albite. Fe, Ni and S are depleted, suggesting that minor phases such as Fe-Ni metallic alloys and metal sulfides or sulfates are underrepresented in the sample. The Chergach meteorite has a rather Mg rich olivine content, with much of the Fe being contained in a metallic or alloy form, consistent with an enhancement of major phases over minor in these measurements [Weisberg, et al., 2008]. Differences in the two size fractions are generally smaller than the uncertainty. However major constituent elements are enhanced in the smaller size fractions, whilst minor elements are depleted. This suggest that some softer phases are enhanced in the smaller size fractions, whilst phases which grind less easily are enhanced in larger size fractions. There is also a potential effect of particle size; since a flat surface is required for quantitative measurements, single particles are used and measurements of the smaller size fractions therefore survey less material. This could lead to an underrepresentation of rare component phases and elements. In compositional terms, therefore, Chergach (and potentially other OCs) are a reasonable approximation for IDPs. This is significant since OCs are generally more available than CCs.

The Murchison and Allende meteorites, shown in Figure 1b), have been included here as examples of carbonaceous chondrites which are commonly used as analogues for IDPs [Burchell, et al., 2006, Court and Sephton, 2011, Toppani, et al., 2001]. Allende has suffered some degree of thermal metamorphism so has compositions within the CI-CM range for all elements except Mg, Na and K. The Allende meteorite is known to have an Fe rich olivine matrix, accounting for the Mg depletion. The low abundance of relatively volatile elements such as Na and K is typical of CV meteorites but is known to be extreme in the case of Allende [Clarke, et al., 1971]. However, these elemental constituents and their relatively volatile phases (melting points <1500 K) are present in all samples observed. The Murchison sample agrees well with the CI or CM elemental composition, a fact which, combined with its relatively large available mass (>100 kg) has led to its wide use as an IDP analogue.

The 125-177.5 µm radius size fraction of the NWA meteorite sample, shown in Figure 1 c), is significantly enriched in Ca and depleted in Na. In addition minor elements including Ti, Mn, K, P, Co and S are below the limit of detection. In the 19-53 µm radius size fraction, SEM-EDX analysis shows a highly heterogeneous composition, with each particle appearing to represent a single phase (elemental ratios consistent with Andesine, Olivine and Augite...
were observed). This suggests a large content of refractory (melting point >2000 K) and coarse grained components, inconsistent with the bulk of the IDP flux. This highlights the fact that not all CC meteorites are inherently suitable as IDP analogues. Such materials can be useful, however for examining extreme events, e.g. differential ablation of particles with large entry velocities.

Ground meteorites are therefore variably suitable analogues for IDPs in terms of elemental composition, however they do contain many relevant phases (see section 3.2.1) and since the ablation process is dominantly controlled by the melting point of the phase \cite{Vondrak et al., 2008}, should act as satisfactory analogues in particular cases.

### 3.1.2. Terrestrial Material

Peridot olivine, labradorite, anorthite and albite samples have also been subjected to SEM-EDX analysis. The olivine was found to be \( \text{Fo}_{90} \) forsterite (ratios of Mg:Fe = 10 ± 4 and (Fe+Mg):Si = 1.9 ± 0.6) with some content of Ni, Ca and Al, consistent with the volcanic basalt casing around the peridot in which the olivine formed. The labradorite was measured as having An\(_{55}\) composition (ratios of Na:Ca = 0.8 ± 0.2 and (Na+Ca):(Si+Al) = 0.244 ± 0.014). The anorthite sample was found to have An\(_{97}\) composition (ratios of Na:Ca = 0.04 ± 0.02 and (Na+Ca):(Si+Al) = 0.244 ± 0.014). The albite sample was found to have a composition consistent with such a sodium feldspar (ratios of Al:Si = 0.344 ± 0.013 and Na:Si = 0.33 ± 0.04). The reproducibility (standard errors typically <20 %) of these measurements and their agreement with expected ratios for each mineral support the use of the SEM-EDX characterisation technique so long as numerous spectra are measured for each sample.

### 3.1.3. Synthetic samples

SEM-EDX has been used extensively here to assess the composition of the synthetic products, particularly with regards to the removal of NaCl and Na\(_2\)SO\(_4\) salt byproducts. In general it was found that around 10 hours of processing in the Soxhlet apparatus was sufficient to remove these byproducts. This validates the synthesis described above.

Subsequent to the purification process, compositions were confirmed for goethite (Fe:O = 0.6 ± 0.4), hematite (Fe:O = 0.7 ± 0.3), Mg\(_2\)SiO\(_4\) (Mg:Si = 2.0 ± 0.5) and Fe\(_2\)SiO\(_4\) (Fe:Si = 2.4 ± 0.8). When a ratio of precursors designed to produce MgFeSiO\(_4\) was used, a ratio of Mg:Fe = 0.43 ± 0.12 and (Fe+Mg):Si = 1.8 ± 0.3 was measured. The overabundance of Fe can partially be explained by the presence of a ferrihydrite or goethite impurity (see section 3.2.2); however, since the ratio of Fe+Mg to Si is not less than 2 it is possible that Mg accommodation into the synthetic Mg\(_x\)Fe\(_{2-x}\)SiO\(_4\) is less effective than that of Fe. This measured composition did not significantly vary on annealing to enstatite / hematite.

Note that oxygen ratios are reported for the crystalline iron oxides, but not for amorphous silicates. This is due to contamination by surface adsorbed H\(_2\)O in the case of the amorphous silicates, despite storing the samples in a vacuum dessicator for several weeks prior to
composition measurements. This is thought to be due to the relatively large surface area of
the silicate materials with respect to the iron oxides (hundreds as opposed to several m² g⁻¹
respectively, see Table 1 and Section 3.3). For applications where surface reactivity is
important, this surface water could be removed by heating in a vacuum, though potential
phase changes in the dust by such annealing should be considered where this is carried out
(see section 3.2). In generally, target compositions could be successfully synthesised and
assayed using the techniques described here. Of the elements described here, Fe is the
most abundant in Earth’s atmosphere. Mg and Si have a slightly lower abundance with Na, K
e tc. present in smaller amounts [Plane, et al., 2015]. Elementally then, these materials
therefore represent suitable compositional analogues for MSPs.

3.2. Textural analysis and compositional maps

The homogeneity of analogues is one of the key differences between ground bulk samples
and IDPs. This can be evident in the elemental or mineralogical composition of the analogue
material. Particle topography and homogeneity, both within each particle and across whole
samples, was assessed using SEM-EDX mapping (FEG-SEM – FEI Nova 450) with EDX
(AmteK) at 18 kV) for the Chergach and Allende meteorites. Particles forming the Allende
CV3 carbonaceous chondrite are heterogeneous even to the naked eye, with some white
and some rounded glassy particles evident. These correspond respectively with Ca- and
Al-rich inclusions and chondrules. Chergach particles, on the other hand, appear more
homogeneous within each size fraction because of the recrystallization shown in this
petrologic type (H5) of ordinary chondrites. These trends hold through to the microscopic
scale, as demonstrated by the following analysis.

Figure 2 shows a micrograph and co-located compositional maps for a particle of Chergach.
The Mg rich olivine matrix is clearly demonstrated, with smaller grains of a variety of phases
evident. These potentially include metallic Fe (top left, bottom centre) albite / feldspar
(regions with Na and Ca).

Figure 3 shows a wide angle micrograph of a large section of the Allende meteorite before
grinding. Large features (100s of µm) are visible including a barred olivine chondrule (A), a
porphyritic olivine chondrule (B), the fine grained carbonaceous matrix (C) and a metal
sulfide inclusion (D). The scale of this image makes it clear that, while the mean of a large
sample of particles may be a Cl or CM (or in this case CV) composition as are IDPs, a ground
sample of the meteorite will contain an overabundance of coarse and simple mineral grains,
which would be representative of only a small fraction of IDPs.

Figure 4 shows an aggregate particle of Allende, where a region of scoreacious material is in
contact with a fine grained region (boundary marked by a dashed white line). EDX mapping,
performed on the region shown by the white box on the micrograph, shows that the
scoreacious region is depleted in volatile elements such as S, likely indicating loss of volatile
phases such as troilite or more likely metal sulfate [Burgess, et al., 1991]. The fine grained
region also shows a more heterogeneous content, with some Ca containing domains
evident. This suggests that the right-hand region has undergone significantly more heating, sufficient for volatile phases to evaporate and leave only the anhydrous metal silicate content. The left-hand region, however, has undergone less thermal alteration and resembles more closely a fine-grained IDP.

Figure 5 shows two grains of Allende, one single mineral grain (left) and one fine aggregate (right). The single mineral particle appears to be a sodalite or nepheline (sodium silicate, albite is not present in significant quantities in the Allende meteorite [Clarke, et al., 1971]). The fine-grained aggregate demonstrates the Fe-rich olivine matrix of the Allende meteorite, with a heterogeneous content of Na, Al, Ca and some detectable Ni.

These examples have been selected to provide a flavour of the variability in the chondrites examined here. Such heterogeneous compositions are common in IDPs, however the fine-grained nature of the majority of IDPs means that large grains such as those observed here are rare. Other authors have reported on the particle-to-particle variability inherent in ground meteorites [Jessberger, et al., 2001]; however, the direct comparison presented here allows some clear conclusions to be drawn. The variability displayed here for the Allende meteorite demonstrates the need to characterise samples in detail, since this heterogeneity could have significant consequences when analysing the surface or bulk properties. For example, the rates of catalytic processes or early or late release of an ablating material could be dominated by a statistically uncommon particle or active site.

One further observation is that the elemental heterogeneity in Chergach is not significantly different to that in Allende. While the texture of the grains is significantly different, grains of similar elements are present with a similar heterogeneity. This suggests that Chergach and other OC meteorites may be similar to the comparatively rare CC meteorites as IDP analogues.

### 3.3. Crystal Structure

The crystal structure of the analogue used can be important for many applications. It is crucial, for example, when studying the differential ablation of IDPs since each element will be released dependent upon the melting points of its constituent phases and its vapour pressure over these phases. The crystal structure of the analogues was investigated by powder X-Ray Diffraction (XRD, Bruker D8 equipped with a germanium monochromator, using Cu K-α radiation). Measured patterns were compared to literature structures via a Rietveld refinement. This involves reproducing a pattern by comparison to one or more structures. Inputs such as the crystallographic space group, lattice parameters and atomic positions allow the prediction of patterns which can be compared to the experimental observations. Factors such as nanocrystallinity and sample displacement can also be simulated to investigate peak shifts and broadening effects. Here this was carried out using the TOPAS software [McCusker, et al., 1999].
3.3.1. Meteoric and Terrestrial Material

XRD analysis was only performed for the Chergach H5 OC due to the limited availability of other samples. Observed patterns for the albite and peridot olivine samples are compared to that of Chergach in Figure 6. Measured patterns (in black) can be reproduced by Rietveld refinements (in red). For peridot olivine a forsterite structure produces good agreement, for albite a structure of that mineral agrees well, and for Chergach structures of forsterite, albite and ferrosilite combine to give a good representation of the data. Whilst ferrosilite is not likely present in significant quantities, the enstatite end member of the same solid solution series has a similar structure. This suggests that in structural terms Chergach chondrite can be represented simply by a mixture of terrestrial minerals. Comparison to the SEM-EDX mapping, particularly the similarity of the heterogeneous phases present suggests that this can also be extended to other meteorites and IDPs. It is worth noting that the sample likely contains many other minor phases which are below the limits of detection here.

In terms of ablation experiments, individual minerals are useful to characterise phase changes as a function of heating [Sandford and Bradley, 1989]. Mixtures of minerals may be valuable to compare to meteoritic aggregates or even to IDPs if available, and hence evaluate the effect of grain aggregation and porosity on the release of the elemental constituents. Consideration should be given, however, to the variation of melting points across solid solution series e.g. [Bowen, 1913]. In the specific case of the Chergach meteorite (olivine composition Fo$_{81.6}$ [Weisberg, et al., 2008]) the melting point of some phases may be similar, however for a more general study such considerations should be taken into account and terrestrial mineral analogues chosen with care. Terrestrial minerals can also be of great value in calibrating instruments, providing simple systems with known elemental ratios which are easy to characterise and available in greater supply than meteorite samples.

3.3.2. Synthetic samples

XRD patterns for the synthetic samples discussed in this study are shown in Figure 7. Reitveld refinement (see Section 3.3, above) has a variable, though generally good, ability to reproduce the observed patterns and in some cases providing useful information regarding the synthetic product. Mg$_x$Fe$_{2-x}$SiO$_4$ materials are mainly amorphous by XRD. In the case of Fe$_2$SiO$_4$ the broad crystalline peaks observed can be fitted with a goethite phase. For MgFeSiO$_4$ the similarly broad peaks are compared to a 6 line ferrihydrite phase. Ferrihydrite is a hydrated iron oxide similar to goethite. The broadness of these peaks, particularly that around 35°, is an indication of an amorphous phase or of crystals on the nm scale. For MgFeSiO$_4$ the pattern has been fitted assuming crystallites of 2.5 nm radius and for Fe$_2$SiO$_4$ the crystallites are assumed to have radii of 5.2 nm. This nanocrystallinity is discussed further in the context of
the particle morphologies in section 3.4. Implications of this crystallinity in terms of using these materials as analogues for MSPs are discussed in section 3.6.

For an annealed sample of MgFeSiO$_4$ strong crystalline peaks are observed, which can be fitted with a hematite phase. Repeated SEM-EDX analysis shows that the composition does not change, suggesting that Mg and Si are present as an amorphous or nanocrystalline MgSiO$_3$ phase.

The XRD pattern of synthetic goethite is well fitted by assuming a structure of that mineral. Upon annealing a sample of this material at 573 K for 24 hours the XRD pattern could be modelled reasonably well by a hematite structure, consistent with the dehydration of the goethite (Gualtieri and Venturelli, 1999). The fit in this case is imperfect, notably some peaks show significant broadening and the relative peak intensities are not well represented. Since peak broadening due to crystalline size is angle dependent this is likely an indication of relatively small crystallites (perhaps 50-100 nm in radius) but we were not able to find a crystallites size which gave a good fit. Peak intensity variations are likely due to some preferred orientation. Implications of this crystallinity in terms of using these materials as analogues for MSPs are also discussed in section 3.5.

### 3.4. Surface Area and Morphology

Surface properties are of paramount importance for the heterogeneous reactivity of an atmospheric solid and therefore an analogue intended to investigate such phenomena. The upper limit to the available reactive surface area was measured for the materials discussed here by the Brunauer, Emmet and Teller (BET, Micrometrics ASAP 2020) method. The results are shown in Table 1. The anomalously high specific surface area for the Mg$_x$Fe$_{2-x}$SiO$_4$ materials is due to their unusual surface morphology.

This was investigated by Transmission Electron Microscopy (TEM, FEI Tecnai F20 200kV FEGTEM fitted with a Gatan Orius SC600 CCD camera), with a representative micrograph shown in Figure 8. The thin, folded sheet like nature of the material leads to a large surface potentially being available for uptake and reaction of gases.

The inset to Figure 8 shows that at atomic resolution nanocrystalline domains can be seen on the surface of the MgFeSiO$_4$. The scale of these domains is close to the 2.5 nm radius required to resolve the ferrihydrate peaks in the XRD analysis discussed above. This is compelling evidence that these crystalline domains produce the peaks seen in the XRD patterns and can therefore be considered an upper limit to their size. The implications of this nanocrystallinity in terms of using these materials as analogues for MSPs are discussed in section 3.5.

The surface area which is actually involved in reaction will depend on the reactivity in a given system, however this large upper limit allows measurement of processes which are relatively slow (Frankland, et al., 2015). The question of available surface area is also pertinent in the environment, since fractal or dendritic particles are often represented as
spheres for reasons of computation efficiency [Saunders, et al., 2007]. Atmospheric
processing by H₂O and acidic gases may also change the available surface area over time.
Any use of such materials for surface science should include a careful study of the available
surface area. In contrast, crystalline materials such as the goethite and hematite analogues
presented here generally have BET surface areas close to that calculated by assuming
spherical particles of reasonable size distribution (see Section 3.5). In that case the available
surface area in a given experiment can often be well approximated by considering the
experimental geometry (e.g. a coated flow tube), or assuming that layered samples of
particles are close packed spheres with associated pore spaces [Keyser, et al., 1991].

3.5. Size Distributions and Density

The size distribution of an analogue can be important for many reasons. For example, a
layered sample of uniform sized particles might be expected to close pack with open pores,
whilst a more heterogeneous distribution might have these pores between larger particles
filled by smaller particles. Evaporation of particles may be kinetically controlled by the
surface for smaller particles, or diffusion through the molten bulk of larger particles. Density
is also important in thermal studies since it impacts on the conductivity of the material, and
in surface studies since the density of active sites is likely related to the density of a
particular surface feature (e.g. a particular ion or structural defect).

For dense, approximately spheroidal particles such as the hematite, goethite, enstatite /
 hematite and ground meteorites and minerals described here measuring the particle size
distribution, which results from grinding, is important for designing experiments and
interpreting results. For particles on the µm scale, optical microscopy can be used to probe
such properties. Here we have used a backlit optical microscope equipped with an objective
with a magnification of 10× to image arrays of particles. Typically around 3 dozen images
containing 300-600 individual particles were produced using the following algorithm. First,
particles were differentiated from the image background by the pixel brightness; second, to
account for particles lying on top of each other in the images, the identified particles were
artificially eroded until they separate and then rebuilt until they touch; particles in contact
with the image edge were discarded; a minimum particle size was set to remove a
background of smaller particles from each image; the area of each particle was then
calculated (pixels were converted to µm² by reference to a standard 1 µm image). Control of
the minimum size and pixel brightness threshold allow the analysis to function even for
moderately transparent or reflective particles. Each particle area was converted to an
equivalent circular radius, and the resulting particle size distribution was then fitted to a log-
normal distribution and normalised to a probability density function for comparison.

The erosion and reconstruction process introduces several uncertainties into the analysis.
First, the volume of a particle which lies on top of another is not counted, giving a
downward bias in the estimated size, while imperfect separation of particles where the
distance of contact is similar to the particle dimensions leads to an upward bias. Treating
particle radius based on area is a major assumption of this method. For example, if the
particle contacts a substrate or gas medium in profile then this may be valid, provided that
the particles lay similarly \textit{in situ} to their position on the microscope slide. However if volume
properties (such as optical extinction) of the material is to be investigated then this method
would likely overestimate the volume distribution, since gravitational settling on the surface
of the microscope slide will tend to favour the particles laying horizontally. This would result
in the shortest dimension being in the vertical (which is neglected here).

Example images for two of the meteorites and two of the terrestrial minerals discussed here
are shown along with the measured size distributions for the Chergach meteorite in Figure
9. Figure 9 (b) shows the result of analysing Figure 9 (a) using the image analysis software,
where overlapping particles have been separated and all those which were touching the
image edge have been discarded. The sizes of the remaining particles are used in
determining the size distributions. These observed distributions have mean sizes within the
stated pore size of the sieves used for separation. In general, materials which form
needle-like particles (e.g. albite, see Figure 7 d) give slightly larger sizes in this analysis since
they are better able to penetrate the pores in the sieves.

Size analysis techniques for smaller particles generally involve suspending particles in either
a gaseous (e.g. Scanning Mobility Particle Size (SMPS) analysis \cite{Liu2003}) or
liquid (e.g. Dynamic Light Scattering (DLS) \cite{Chu2008}) medium. Aggregation can influence
the results of these experiments such that in some cases only the limits of particle sizes are
measurable. Measurements by both SMPS and DLS have shown that the Mg$_x$Fe$_{2-x}$SiO$_4$
analogs discussed here have primary particle radii on the order of 200-500 nm. TEM
imaging shows the presence of some particles on smaller scales (down to 10s of nm).

Proper measurement and interpretation of particle sizes is extremely specific to the
application in question, however one general point to note is that while generating
analogs to IDPs on the appropriate scale is relatively easy, the relatively small size of
MSPs (molecular dimensions up to 100 nm radii \cite{Bardeen2008}) means that
manufacturing analogues for these particles in sufficient quantity for many experiments is
extremely challenging. Instead, the approach in the present study is to generate particle
analogs likely to have similar properties, and the measured properties (e.g. rates or
extinctions) extrapolated to atmospherically relevant particle size distributions.

Another volume dependent quantity which can be important for experimental applications
is the mass density of the analogue. Mass densities for some of the analogues discussed
here are given in Table 1. The synthetic samples have densities significantly lower than for
their equivalent bulk minerals, including those materials which are crystalline. This suggests
a significant porosity, in agreement with the morphology described above for the
Mg$_x$Fe$_{2-x}$SiO$_4$ materials. \cite{Jessberger2001} also noted a significant porosity in collected
micrometeorites and IDPs.
3.6. Discussion

The meteorite samples discussed here represent a range of types (CC and OC, differing metamorphic groups). Whilst CC materials such as CI and CM appear to dominate the IDP flux, design of experiments using meteorites as IDP analogues is generally restricted to those types for which large masses of sample are readily available. The elemental composition, heterogeneity and crystal structure data presented indicate, however, that in many cases OC meteorites or even single minerals represent suitable analogues to IDPs (even beyond the 10 % of IDPs which are OC material). Murchison is a good example of an aqueously altered chondrite (groups 2) and contains significant hydrated minerals such as phyllosilicates, as found in fine grained IDPs [Taylor, et al., 2012]. These phases will dehydrate and recrystallise when heated and so will most likely ablate similarly to the feldspathic minerals present in OCs. In addition, analysis of the NWA meteorite shows that there are large deviations within CC meteorites, and that many of these may be useful only for examining extreme or rare cases of IDPs. For example, studies are underway at the University of Leeds into differential ablation using a new Meteor Ablation Simulator (MASI) instrument [Gómez Martín, et al., 2016 - Unpublished results] [Kearsley, et al. (2009)]

produced cometary analogues for impact studies in aerogel and Al foil by aggregating mineral fragments using an acrylic adhesive. These analogues provide excellent insights into the collection of IDPs and cometary dust in aerogel. From the point of view of chemical ablation and surface chemistry studies, which take place on a relatively long time scale compared to impact and where the adhesive may cause undesired interferences, further characterisation experiments are required. The adhesive coating would also be likely to interfere with surface science experiments, where the mineral surface should be exposed to be observed directly.

Saunders and Plane (2011) have described a method of producing MSP analogues by photolysis of chemical precursors which then recondense in the gas phase. Amorphous Mg$_x$Fe$_{2-x}$SiO$_4$ materials and Fe oxides are produced with a primary particle radius of 5-10 nm. These small primary particles agglomerate to produce fractal-like particles, likely a good representation of environmental MSPs. These particles have been probed for their optical properties and ice nucleating ability [Saunders, et al., 2010] [Saunders and Plane, 2006]. We note that the amorphous nature found for those particles is similar to that described here. Where the primary particle radius is 5-10 nm, this sets the upper limit of the crystallite size similar to Mg$_x$Fe$_{2-x}$SiO$_4$ sheets with maximum thicknesses / domain radii of several nm (Figure 8).

One significant difference is that the photochemical methods are able to produce Fe oxides which are amorphous, whereas we only observe crystalline products. This may simply be due to the size of the crystallites formed. Whereas in the photochemistry experiments many particles nucleate, take up the available precursor and at some point grow faster by agglomeration than deposition to existing particles, our solution phase synthesis allows relatively fewer particles to grow to larger sizes at which their crystallinity is measurable.
Since MSP remain at small sizes, the photochemical method is likely more representative, so that care should be taken when using the materials synthesised by sol-gel processes as analogues for MSP.

Biermann, et al. (1996), who used micrometeorites as analogues for MSP nucleating crystallisation in polar stratospheric clouds, found that nucleation rates were not high enough to explain observed cloud. As the authors in that study observed, micrometeorites are generally compact, dense particles coated with magnetite crystals. MSPs which are present in stratospheric droplets, on the other hand, will have been processed and largely dissolved by the acidic content of those droplets [Saunders, et al., 2012, Murphy, et al. (2014) used single particle mass spectrometer measurements to show that silicon and aluminium in such droplets are generally solid while most other metals are in solution. A silica or alumina substrate would therefore seem more appropriate for investigating nucleation in polar stratospheric clouds.

Nachbar, et al. (2016) used photolysis of chemical precursors to produce very small (several nm radii) particles, which were held in an ion trap at supersaturated conditions and used to measure nucleation kinetics and particle growth. These particles are alike both in size and composition to MSPs and therefore are highly appropriate as MSP analogues. As with the photochemical technique of Saunders and Plane (2011), the difficulty in using this material to investigate a wider range of applications lies in producing bulk amounts (typically several grams are required for uptake experiments).

4. Conclusions

This study has demonstrated the preparation of a variety of analogues for IDPs and MSPs. The methods are fairly straightforward to implement and use materials which are widely available. No single analogue best represents IDPs or MSPs in general since this rather depends on the application being investigated. Instead some common issues are raised in order to help researchers choose materials for a wide range of applications.

It has been shown that reasonable analogues for IDPs can be produced by grinding and size segregating meteoritic material. The elemental composition of such analogues will vary slightly from the target material but phases which are present in IDPs will be well represented in ground meteorites. OCs are scarce in micrometeorite collections, but samples of OC meteorites are much easier to source than CCs. Although ground OCs produce denser, more compact analogues, elemental compositions and mineralogy are reasonably close to those of average IDPs. Terrestrial materials such as peridot olivine, labradorite, anorthite and albite, when similarly ground, are good analogues for individual phases within IDPs, particularly for applications such as instrument calibration where paucity of sample can be an issue. In the cases of both meteorites and terrestrial minerals it was shown that mechanical sieving can reliably produce target particle size fractions which are representative of IDPs.
Chemical syntheses were presented for compounds with elemental compositions similar to the olivine solid solution series. These are thought to be nanocrystalline folded sheets. Such a morphology is not likely representative of MSPs or IDPs, however it should be noted that the domain size of such crystallites is similar to the primary particle size of MSPs and therefore a similar nanocrystallinity may be possible. The large surface area presented by this morphology allows measurement of relatively unreactive processes which occur at the interface, whilst the elemental composition is similar to that expected for MSPs. These materials can be annealed to produce enstatite and hematite, which are also suitable analogues for potential components of MSPs.

A chemical synthesis was also presented for crystalline goethite, which could be dehydrated to produce hematite. Goethite is a potential component of MSP whilst hematite could be present in both IDPs and MSPs. The crystalline nature of these materials may not be representative of MSPs; nevertheless for some applications they may still be considered suitable analogues.

Finally, it is clearly of great importance that in laboratory experiments where analogues are used for IDP or MSP materials, careful attention is paid to characterising the analogue and ensuring that the limitations of its applicability are understood.

5. Acknowledgements

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21


Table 1. Genesis, proposed analogue use, BET surface area and bulk density of samples described in this study.

<table>
<thead>
<tr>
<th>Analogue</th>
<th>Genesis</th>
<th>Analogue for</th>
<th>BET surface area / m² g⁻¹</th>
<th>Mass density / g cm⁻³</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mg₂SiO₄</td>
<td>MgCl₂ + Na₄SiO₄</td>
<td>MSPs</td>
<td>102 ± 5</td>
<td></td>
</tr>
<tr>
<td>MgFeSiO₄</td>
<td>Fe(SO₄)₂(NH₄)₂ + MgCl₂ + Na₄SiO₄</td>
<td>MSPs</td>
<td>358 ± 16</td>
<td>2.647 ± 0.004</td>
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<td>Enstatite / Hematite</td>
<td>From MgFeSiO₄ as above, sintered at 1273 K for 24 hours</td>
<td>MSPs</td>
<td>3.07 ± 0.03</td>
<td></td>
</tr>
<tr>
<td>Fe₂SiO₄</td>
<td>Fe(SO₄)₂(NH₄)₂ + Na₄SiO₄</td>
<td>MSPs</td>
<td>244 ± 2</td>
<td>3.2638 ± 0.0007</td>
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<tr>
<td>Goethite</td>
<td>Fe(SO₄)₂(NH₄)₂ + NaOH + compressed air</td>
<td>MSPs</td>
<td>41.4 ± 0.4</td>
<td>3.964 ± 0.006</td>
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<tr>
<td>Hematite</td>
<td>From Goethite as above, dehydrated at 573 K for 24 hours</td>
<td>MSPs</td>
<td>34.4 ± 0.2</td>
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<td>Peridot Olivine*</td>
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<td>IDPs</td>
<td>n/a</td>
<td></td>
</tr>
<tr>
<td>Albite*</td>
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<td>IDPs</td>
<td>n/a</td>
<td></td>
</tr>
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<td>IDPs</td>
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<td>2.9†</td>
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<tr>
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<tr>
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<td>IDPs</td>
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<td>2.7†</td>
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</tbody>
</table>

* Terrestrial mineral samples were obtained from the University of Leeds, School of Earth and Environment research collections
†Values from the literature. NWA from Opeil Sj, et al. (2012) others as referenced in Section 2.1
Figure Captions

Figure 1. Elemental compositions, normalised to Si, comparing each of the meteorites used here to the composition of CI, CM and its own group [Hutchison, 2004]. a) Two size fractions (<19 µm and >177.5 µm) of the Chergach (H5) meteorite. b) The Allende (CV3) and Merchison (CM2) meteorites. c) The North West Africa 5515 (CK4) meteorite, 125-177.5 µm size fraction. Error bars show the 95% confidence interval of 4-8 measurements. Measurement techniques vary, see text for details.

Figure 2. SEM-EDX mapping for a particle of the Chergach meteorite. Si is omitted due to interference from the substrate. The bulk Fo\textsubscript{82} olivine is clearly shown in the Mg panel. Grains containing Fe & Ca (lower centre), Na & K (lower right and centre right), and each of those elements individually.

Figure 3. Backscatter electron image of a thin section of the Allende meteorite before grinding demonstrating the scale of individual features such as a barred olivine chondrule (A), a porphyritic olivine chondrule (B), the fine grained carbonaceous matrix (C) and a metal sulfide inclusion (D).

Figure 4. SEM-EDX mapping for a particle of the Allende meteorite, showing both a fine aggregate and scoreacious grains. The fine aggregate particle has a higher S content and is more heterogeneous than the scoreacious grain.

Figure 5. SEM-EDX mapping for two particles of the Allende meteorite, showing one single mineral particle and one fine aggregate grain. Si can be included here since a Cu substrate was used. The single mineral (left) particle shows a homogeneous composition consistent with nepheline or sodelite, whilst the fine grain aggregate appears to have an olivine matrix containing grains of diverse composition.

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Figure 8. Typical transmission electron microscope image of the MgFeSiO\textsubscript{4} described in this study showing ‘folded sheet like’ morphology. Insert shows an example of nanocrystalline domains on the sheet surface.

Figure 9. Example microscope images of the 75-125 µm radius fraction for (a) the Chergach meteorite, (c) the Allende meteorite (d) the peridot olivine and (e) the albite samples. Scales for other images are as for the Chergach sample. Panel (b) shows the particles found by the
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Figures

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