

Measuring the level of interstellar inheritance in the solar protoplanetary disk

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Abstract—The timing and extent to which the initial interstellar material was thermally processed provide fundamental constraints for models of the formation and early evolution of the solar protoplanetary disk. We argue that the nonsolar (solar $\Delta^{17}\text{O} \approx -29\text{‰}$) and near-terrestrial ($\Delta^{17}\text{O} \approx 0\text{‰}$) O-isotopic compositions of the Earth and most extra-terrestrial materials (Moon, Mars, asteroids, and comet dust) were established very early by heating of regions of the disk that were modestly enriched (dust/gas ≥ 5 –10 times solar) in primordial silicates ($\Delta^{17}\text{O} \approx -29\text{‰}$) and water-dominated ice ($\Delta^{17}\text{O} \approx 24\text{‰}$) relative to the gas. Such modest enrichments could be achieved by grain growth and settling of dust to the midplane in regions where the levels of turbulence were modest. The episodic heating of the disk associated with FU Orionis outbursts were the likely causes of this early thermal processing of dust. We also estimate that at the time of accretion the CI chondrite and interplanetary dust particle parent bodies were composed of ~ 5 –10% of pristine interstellar material. The matrices of all chondrites included roughly similar interstellar fractions. Whether this interstellar material avoided the thermal processing experienced by most dust during FU Orionis outbursts or was accreted by the disk after the outbursts ceased to be important remains to be established.

INTRODUCTION

How and when the original interstellar material inherited from the protosolar molecular cloud was processed in the early solar system, and what fraction survived to be incorporated into planetesimals and planets remain poorly constrained. Yet these are critical constraints on our understanding of how the solar system formed and, by extension, for understanding low mass star formation in general. It is thought that early in the formation of the solar system, most material was accreted close to the growing Sun and experienced high temperatures that destroyed all but, perhaps, the most refractory presolar oxide materials. To enable accretion onto the Sun, angular momentum would have been transferred from the accreted material through the early protoplanetary disk, causing a rapid outward expansion. As a result, the disk at this early stage can be dominated by material that had been reprocessed at high temperatures (e.g., Bockelée-Morvan et al. 2002; Dullemond et al.

2006; Ciesla 2011). However, there would have been continued accretion from the surrounding molecular cloud envelope onto the disk. In the cooler outer parts of the disk where the accretion shock would have been weak, all but the most volatile ices in this accreted material would have been essentially unprocessed. Turbulent mixing in the disk and cooling of the disk as accretion rates onto the Sun diminished would have enabled gradual homogenization of the processed and unprocessed material. Here, we attempt to test/quantify this qualitative picture of solar system formation.

Primitive asteroids and comets provide the best record of the early solar system available to us. We have samples of these objects in the form of chondritic meteorites, interplanetary dust particles (IDPs), and material returned by spacecraft. One group of primitive meteorites, the CI chondrites, have elemental compositions that are identical, within error, to the rock-forming component of the Sun (Asplund et al. 2009). Unfortunately, they experienced extensive aqueous alter-

ation in their parent body that has destroyed most of their primary materials (e.g., Brearley 2006). Nevertheless, they contain significant abundances of presolar circumstellar grains (e.g., Huss and Lewis 1995), demonstrating that at least some molecular cloud material survived solar system formation. These circumstellar grains, ranging in size from nanometers to tens of microns, are relatively refractory (Zinner 2014), although the noble gases that they contain suggest that they could not have been heated even briefly above 300–700° C (Huss and Lewis 1994; Lewis et al. 1994; Amari et al. 1995). CI chondrites also contain ~3.5 wt% C in organic matter, the bulk of which is a solvent insoluble organic matter (IOM) that appears to be macromolecular (e.g., Hayatsu et al. 1977; Cody and Alexander 2005; Remusat et al. 2005) and is present in grains that range from approximately tens of nanometers to several microns (e.g., Floss and Stadermann 2009; Remusat et al. 2010; Le Guillou and Brearley 2014; Le Guillou et al. 2014). Large D and ¹⁵N enrichments suggest that it formed in cold environments in the interstellar medium (ISM) and/or the outer solar system (Robert and Epstein 1982; Yang and Epstein 1983; Alexander et al. 2007b; Gourier et al. 2008).

The CI-like material seems to have been a major component of all chondrite matrices since they have roughly CI-like abundances of circumstellar grains, organics, and highly volatile elements (Huss and Lewis 1995; Alexander et al. 2001; Alexander 2005b; Zanda et al. 2009, 2011, 2012; Davidson et al. 2014). The exceptions are the circumstellar silicates, which are absent in CI chondrites and highly variable in other chondrites because they are so susceptible to destruction by parent body processes (Floss and Stadermann 2012; Leitner et al. 2012, 2016; Nittler et al. 2013). Not all chondrites have experienced the extensive alteration that the CI chondrites did, and the matrices in these chondrites suggest that the original CI material was predominantly a mixture of fine-grained crystalline and amorphous silicates (Alexander et al. 1989b; Brearley 1993; Greshake 1997; Chizmadia and Brearley 2008; Abreu and Brearley 2010; Hewins et al. 2014; Le Guillou and Brearley 2014; Leroux et al. 2015).

A plausible picture of what the original CI material may have looked like probably comes from anhydrous chondritic porous (CP-) IDPs. These IDPs, which may come from comets and/or primitive asteroids, have experienced little or no parent body processing and are primarily composed of fine-grained crystalline silicates, mostly olivine and pyroxene, and amorphous silicates (so-called GEMS or glass with embedded metal and sulfide) (Bradley 2003). There are many similarities between the mineralogies and compositions of the crystalline components in primitive chondrite matrices and IDPs, as well as comet Wild 2 (Alexander et al.

1989b; Klöck et al. 1989; Gounelle et al. 2008; Ishii et al. 2008; Zolensky et al. 2008). There is also abundant evidence for amorphous material in the matrices of primitive chondrites that some have suggested may be related to GEMS (Alexander et al. 1989a; Brearley 1993; Greshake 1997; Le Guillou and Brearley 2014; Leroux et al. 2015). These similarities suggest that the same fine-grained materials were dispersed throughout the chondrite and comet-formation regions, that is, much of the solar nebula.

The silicate dust in the diffuse ISM is >98% amorphous (Kemper et al. 2004, 2005). Hence, the fine-grained crystalline material in chondrite matrices, CP-IDPs, and comets must be the products of thermal processing in the solar system, which is consistent with their near-terrestrial rather than solar O-isotopic compositions (Aléon et al. 2009; Keller and Messenger 2011; Nakashima et al. 2012b; Starkey and Franchi 2013). However, some fraction of the amorphous silicates and carbonaceous material could be interstellar. It has also been argued that the nonsolar D/H ratios of chondritic water indicate that some interstellar water also survived solar system formation (Cleeves et al. 2014, 2016).

Here, we use O isotopes to constrain how and when the fine-grained crystalline and amorphous material that appears to be common to chondrite matrices, IDPs, and comets formed. We then use the H isotopes of water accreted by the CI and CM chondrites, the abundance of IOM in CI chondrites, and the abundances of roughly CI composition GEMS in IDPs to estimate what fraction of the original interstellar material survived in the chondrite and IDP parent body formation regions.

THE O ISOTOPES OF FINE-GRAINED CRYSTALLINE SILICATES: EARLY PROCESSING OF ICE-RICH DUST

Some or all of the crystalline material in IDPs appears to have condensed from a hot gas (Bradley et al. 1983). The many chemical and mineralogical similarities between chondrite matrices, comet Wild 2 dust, and CP-IDPs suggests that the fine-grained crystalline materials in matrices and Wild 2 dust are also largely condensates, although there are contributions from chondrules and refractory inclusions even in the Wild 2 samples (e.g., Zolensky et al. 2006; Nakamura et al. 2008). The presence of crystalline silicates in other comets (e.g., Wooden 2008; Hanner and Zolensky 2010) indicates that they accreted similarly thermally processed materials as well. Indeed, the results of the Deep Impact experiment into comet Tempel 1 (Lisse et al. 2006) suggest that crystalline silicate abundances in many comets may have been significantly underestimated.

Given the widespread distribution of this fine-grained material, the thermal processing must have occurred early, certainly before the formation of the chondrite and IDP parent bodies, as well as comets. The chondrites began forming ~ 2 Ma after the formation of CV CAIs (Grimm and McSween 1993; Harrison and Grimm 2010; Henke et al. 2013; Sugiura and Fujiya 2014), but the formation times of comets and the parent bodies of IDPs are unknown. The IIAB, IIIAB, and IVA magmatic iron meteorites, whose parent bodies may have formed ~ 0.1 – 0.5 Ma after CAIs (Kruijer et al. 2014), have O-isotopic compositions (Figs. 1 and 2) that are near-terrestrial rather than solar (Moroz et al. 1988; Clayton and Mayeda 1996; Wang et al. 2004; McCoy et al. 2011). If, as is argued below, this nonsolar O-isotopic signature was established in dust by the same thermal processing that produced the fine-grained crystalline silicates, the thermal processing must have occurred very early. This would be consistent with astronomical observations that the abundances of crystalline silicates at the surfaces of T Tauri disks, at least out to ~ 10 AU, are established at ≤ 1 Ma and do not then vary significantly with radial distance or with disk age (Watson et al. 2009; Olofsson et al. 2010; Oliveira et al. 2011). The crystalline fractions at T Tauri disk surfaces (typically 10–20%, but can be up to 80%) are generally lower than in chondrite matrix and IDPs, but it is unlikely that the dust grains observed at disk surfaces faithfully reflect the nature of the dust at their midplanes where most of the dust mass resides (Krijt and Ciesla 2016).

The atomic $\text{Mg}/(\text{Mg} + \text{Fe})$ ratios ($\text{Mg}\#$ s), of the majority of the olivine and pyroxene grains in primitive chondrite matrices, IDPs and Wild 2 particles range from 1 to ~ 0.5 (Zolensky et al. 2008; Frank et al. 2014). Magnesium numbers much lower than unity require more oxidizing conditions than would have existed in a canonical solar nebula (Ebel and Grossman 2000; Ebel et al. 2012; Grossman et al. 2012). The fact that there is such a large range in $\text{Mg}\#$ s indicates a very broad range of $f\text{O}_2$ s during the formation of the crystalline silicates. The simplest means for creating oxidizing conditions in the nebula is to increase the dust/gas ratio, particularly if the water-ice/silicate ratio in the dust is high and organic/silicate ratio is low. Varying the dust/gas ratio, as well as the water ice and organic contents in the dust, provides considerable scope for varying the oxidation state upon heating.

Ice-bearing dust also has the potential for at least partially resolving a long-standing mystery, namely that all but the most refractory extraterrestrial materials have mass-independently fractionated

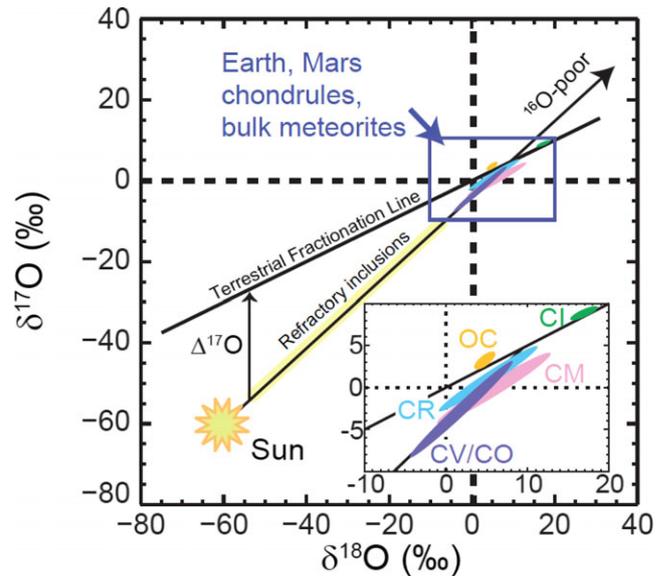


Fig. 1. A schematic three-O isotope plot. In such a plot, most chemical and physical processes cause O isotopes to vary in a mass-dependent way along a line of slope 0.52. This is why almost all terrestrial materials fall on the terrestrial fractionation line. However, except for the Moon, most extraterrestrial materials are mass-independently fractionated relative to the Earth ($\Delta^{17}\text{O} \neq 0$). This is true for chondritic meteorites (inset), but is most obvious for refractory inclusions. Refractory inclusions (and carbonaceous chondrite chondrules) lie on a line of roughly slope 1 that connects the Sun's inferred composition (McKeegan et al. 2011) with those of the chondrites.

O-isotopic compositions (usually measured as $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$, see also Fig. 1) that more closely resemble the compositions of the Earth and Mars than that of the Sun (Fig. 2a). This is certainly true for IDPs and comet Wild 2 samples (see Fig. 2a), as well as chondrite matrices (Grossman et al. 1987; Brearley et al. 1989; Clayton and Mayeda 1999). UV self-shielding, either in the presolar molecular cloud (Yurimoto and Kuramoto 2004; Lee et al. 2008) or the early protoplanetary disk (Clayton 2002; Lyons and Young 2005), is the currently favored process for ultimately producing the range in $\Delta^{17}\text{O}$ values among extraterrestrial materials. UV self-shielding is capable of creating ^{16}O -rich CO and ^{16}O -poor H_2O with compositions that lie at or beyond the extreme ranges seen in extraterrestrial materials (Kobayashi et al. 2003; Sakamoto et al. 2007). Hence, fractionation of the CO from the H_2O is able to explain the full range of O isotopes in solar system materials. The simplest way to achieve this fractionation is to condense the H_2O onto dust grains, leaving the CO in the gas phase, and then fractionate the gas from the dust. However, this

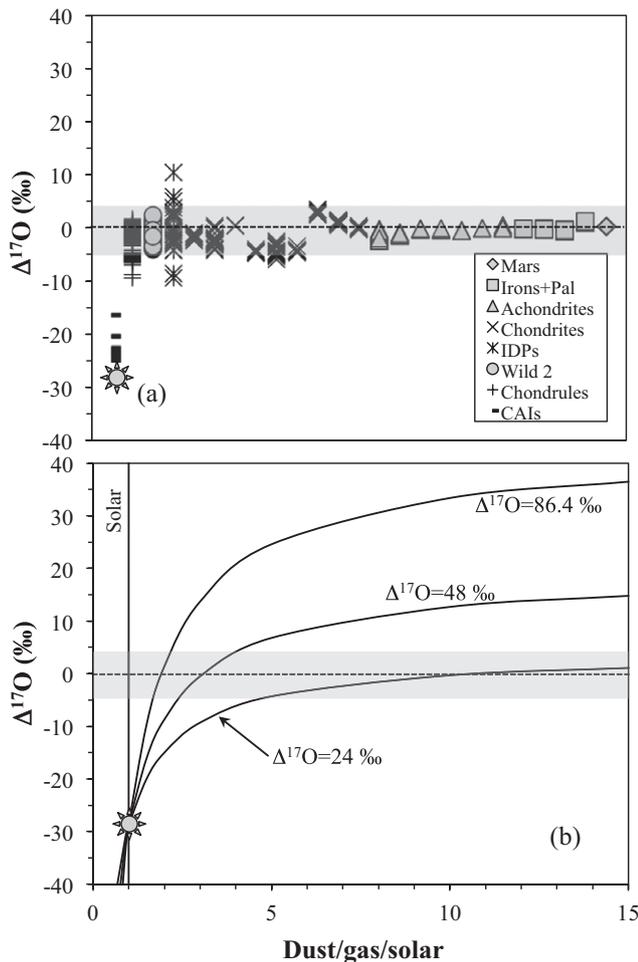


Fig. 2. a) The range of mass-independent O-isotopic fractionations in bulk meteorites, IDPs, comet Wild 2 ferromagnesian samples, chondrules, and refractory inclusions. The mass-independently fractionated O-isotopic composition of the Earth–Moon system is 0‰, and that of the Sun is roughly -29 ‰. b) The predicted mass-independent O-isotopic fractionation in second-generation dust produced by heating of regions of the primordial solar nebula with variable degrees of dust (silicate + IOM + ice) enrichment. Three O-isotopic compositions for interstellar ice were considered. The value of $\Delta^{17}\text{O} = 24$ ‰ is able to produce isotopic compositions that fall within the shaded region over the widest range (>4) of dust/gas ratios. See text and Table 1 for further details. Sources of data: Sun (McKeegan et al. 2011), Mars (Clayton and Mayeda 1996), irons and pallasites (Clayton and Mayeda 1996; Greenwood et al. 2006, 2015), achondrites (Clayton and Mayeda 1996; McCoy et al. 1997; Newton et al. 2000; Greenwood et al. 2005, 2012), chondrites (Clayton et al. 1991; Schulze et al. 1994; Clayton and Mayeda 1999; Newton et al. 2000; McCanta et al. 2008), IDPs (Aléon et al. 2009; Keller and Messenger 2011; Nakashima et al. 2012b; Starkey and Franchi 2013; Starkey et al. 2014), Wild 2 (McKeegan et al. 2006; Nakamura et al. 2008; Nakashima et al. 2012a; Oglione et al. 2015), chondrules (Connolly and Huss 2010; Kita et al. 2010; Weisberg et al. 2011; Ushikubo et al. 2012; Tenner et al. 2013, 2015), and CAIs (Krot et al. 2014).

requires ambient temperatures of <150 – 170 K, and subsequently high temperatures to modify the silicate dust composition.

Many factors will go into determining the O isotopes of condensates in any single thermal event. The full range of parameters that would need to be considered is too large to explore here, but some simple estimates will illustrate that the proposed process can reasonably explain the O isotopes of the fine-grained dust. One important unknown parameter is the O-isotopic composition of the ISM silicates. In many ways, the simplest explanation for the near-terrestrial ($\Delta^{17}\text{O} \approx 0$ ‰) O-isotopic compositions of most solar system materials analyzed to date would be that the interstellar dust composition was also close to terrestrial (Krot et al. 2010). However, the lifetimes of grains in the ISM are generally thought to be short compared to the residence time of material in the ISM (e.g., Jones et al. 1996; Hirashita 2010), although the uncertainties in such estimates are large (Jones and Nuth 2011). While no satisfactory mechanism for producing silicates at low temperatures in the ISM has been identified, most of the ISM silicate dust probably formed there. In which case, it seems likely that it would have an isotopic composition that is similar to the bulk O-isotopic composition of the ISM. Assuming that the Sun’s composition reflects that of the bulk ISM material from which the solar system formed, the ISM silicates in the early solar system would have had bulk O isotopes of $\delta^{17}\text{O} \approx \delta^{18}\text{O} \approx -60$ ‰, $\Delta^{17}\text{O} \approx -29$ ‰ (McKeegan et al. 2011).

If we assume a solar O-isotopic composition for the ISM silicates, an ISM water O-isotopic composition that is equal to the most ^{16}O -poor composition recorded in meteorites (“cosmic symplectite” in the Acfer 094 carbonaceous chondrite with $\delta^{18}\text{O} \approx 180$ ‰, $\Delta^{17}\text{O} \approx 86$ ‰; Sakamoto et al. 2007) and complete fractionation of dust (ice + silicates) from gas, then $\sim 25\%$ of the O in the dust must be from the water ice to produce a terrestrial O-isotopic composition. This percentage implies incomplete water ice condensation as there are roughly equal masses of “rock” (silicates, sulfides, and metal) and H_2O in a solar composition system (e.g., Lodders 2003). This required O fraction from H_2O is independent of the assumed fraction of Fe in FeO in the ISM silicates, which remains quite uncertain (e.g., Voshchinnikov and Henning 2010; Valencic and Smith 2013). To explain the full range of bulk IDP O-isotopic compositions ($\Delta^{17}\text{O} \approx -9$ to $+6$ ‰) (Aléon et al. 2009; Keller and Messenger 2011; Nakashima et al. 2012b; Starkey and Franchi 2013; Starkey et al. 2014), the total range for the fraction of the O that must be supplied by water is 17–30%. However, it should be borne in mind

Table 1. The atomic abundances of O in silicate and organic dust, H₂O ice, and CO in the gas in a solar composition system (normalized to H = 10¹² atoms), along with their initial mass-independent O isotope fractionations, used in the generation of the three curves in Fig. 2.

	O (at.)	$\Delta^{17}\text{O}$ (‰)	$\Delta^{17}\text{O}$ (‰)	$\Delta^{17}\text{O}$ (‰)
Dust (sil. + org.)	146800000	-28.8	-28.8	-28.8
H ₂ O ice	242600000	86.4	48.0	24.0
Dust + ice	389500000	43.0	19.0	4.1
CO	147600000	-218.2	-155.1	-115.6

See text for the details of how the compositions of these components were determined.

that some of the variation in $\Delta^{17}\text{O}$ could instead be due to variable amounts of fine-grained refractory material with $\Delta^{17}\text{O}$ values like those of refractory inclusions, as well as interstellar grains with solar-like compositions. The same may also apply to bulk meteorites.

These estimates for the amount of O in the dust that must be supplied by water ice are almost certainly minimum estimates. This is because complete fractionation of dust from gas is unrealistic and, therefore, there will be a contribution from ¹⁶O-rich CO. This will require a higher contribution from water to counter the influence of the CO. A higher contribution from water would also be more consistent with the likely water ice abundances of cold interstellar dust. To first order, the abundance of H₂O in ISM material should depend on the fraction of the total C in carbonaceous dust, the C/O of the carbonaceous dust, and the fraction of the total C in gases that, unlike the dominant CO, contain no O (e.g., CH₄). Ultimately, the O-isotopic compositions of condensates will depend on many poorly constrained parameters. By assigning reasonable values to most of these parameters, we can explore the influence of the two most pertinent parameters to this study, which are the $\Delta^{17}\text{O}$ value of the interstellar H₂O and the dust/gas ratio at the time of the processing of the silicates. To this end, we assume (1) that the solar elemental composition is that of the bulk Sun estimated by Asplund et al. (2009) but using their CI element/Si ratios for elements from Na to Ni as they are more accurate than the solar photosphere ratios; (2) that the bulk solar and ISM silicate dust $\Delta^{17}\text{O}$ values are -28.8‰ (McKeegan et al. 2011); (3) that ~50% of the C is in dust, as in the diffuse ISM (DISM) (Compiègne et al. 2011); (4) that the atomic O/C ratio is 0.15 for the carbonaceous dust, like that for IOM in chondrites (Alexander et al. 2007b) and comet Halley refractory carbonaceous dust (Kissel and Krueger 1987); (5) that all water is condensed with the silicates; (6) that all C in

the gas is CO, and that CO is the major O reservoir in the gas; (7) that all S is in the dust as FeS; (8) that half of the remaining Fe is FeO giving an ISM-like Mg/(Mg + FeO) = 0.83 (Poteet et al. 2015) in the dust before and after heating; and (9) that all Fe not in FeS and FeO is in metal along with all the Ni and P. With these assumptions, the abundances of O in the three main reservoirs (silicate dust, H₂O, and CO) can be calculated, and they are given in Table 1 normalized to 10¹² atoms of H. Also shown in Table 1 are the $\Delta^{17}\text{O}$ values for the major O reservoirs for three assumed H₂O $\Delta^{17}\text{O}$ values and the corresponding $\Delta^{17}\text{O}$ for the CO needed to maintain a bulk solar composition of $\Delta^{17}\text{O} = -28.8\text{‰}$.

The H₂O is assumed to be fully condensed and is, therefore, a component of the dust. Consequently, varying the dust/gas ratio simplifies to two-component mixing between silicates plus H₂O and CO. Figure 2b shows the calculated $\Delta^{17}\text{O}$ values of thermally processed dust as a function of dust/gas ratio. Near the solar dust/gas ratio the O-isotopic composition changes rapidly, but as the dust enrichment increases the $\Delta^{17}\text{O}$ of the dust and gas asymptotically approaches the bulk composition of the dust. This asymptotic behavior starts at solar-normalized dust/gas ratios of 5–10. These levels of enrichment can be achieved at the midplane beyond the snowline simply by grain growth and settling in regions with modest levels ($\alpha < 5 \times 10^{-4}$) of turbulence (Krijt et al. 2016). Yurimoto and Kuramoto (2004) calculated the isotopic composition of inner solar system gas enriched by migration of ice-bearing dust past the snowline and found asymptotic behavior began at H₂O enrichments of >20 times solar, reflecting their somewhat different assumptions. For a water $\Delta^{17}\text{O}$ value of 86.4‰ (i.e., that of cosmic symplectite: Sakamoto et al. 2007), the bulk $\Delta^{17}\text{O} \approx 0\text{‰}$ for a dust enrichment of slightly less than two. However, the bulk $\Delta^{17}\text{O}$ is a sensitive function of the dust/gas ratio at these low enrichments, and it requires a very restrictive range of enrichments to explain the limited range of bulk $\Delta^{17}\text{O}$ values seen in most extraterrestrial materials.

A more satisfying explanation for why most extraterrestrial materials have a relatively restricted range of $\Delta^{17}\text{O}$ values could simply be that the thermal processing of primordial dust mostly took place in regions with somewhat larger but still modest dust enrichments of ≥ 5 –10 when bulk O-isotopic compositions become less sensitive to the dust/gas ratio. With the many assumptions listed above, to obtain roughly terrestrial values of $\Delta^{17}\text{O}$ in the bulk dust at dust enrichments ≥ 5 –10 requires that a fully condensed, pure H₂O primordial ice have a $\Delta^{17}\text{O} \approx 24\text{‰}$, which is at the lower limit of the range estimated by Yurimoto and Kuramoto (2004). This leaves the high $\Delta^{17}\text{O}$ values (~86‰) of the cosmic

symplectites in Acfer 094 (Sakamoto et al. 2007) unexplained but they appear to be unique to that meteorite, although an isotopically similar material has been reported in one IDP (Starkey et al. 2014). On the other hand, materials with $\Delta^{17}\text{O} \approx 24\text{‰}$ are also rare in extraterrestrial materials, although since the reprocessing of interstellar material seems to have been very efficient perhaps this is not surprising. Reducing the fraction of C in and the Mg# of the interstellar dust will both modestly reduce the bulk $\Delta^{17}\text{O}$ of the processed dust, but not enough to allow for interstellar water $\Delta^{17}\text{O}$ values like those of the cosmic symplectite.

Interstellar and cometary ices are not pure H_2O (e.g., Mumma and Charnley 2011; Öberg et al. 2011a). They generally include some CO and/or CO_2 , although not all of these gases are trapped in the H_2O ices. The condensation of some CO and CO_2 into the H_2O ice will not only reduce the bulk $\Delta^{17}\text{O}$ of the ice but also drive up the $\Delta^{17}\text{O}$ of the bulk gas. For ices in low mass star-forming regions, Öberg et al. (2011a) recommend $\text{CO}/\text{H}_2\text{O} = 0.13$ and $\text{CO}_2/\text{H}_2\text{O} = 0.2$ for the mole ratios in the H_2O -dominated ices. If the CO_2 forms primarily via the reaction $\text{CO} + \text{OH} = \text{CO}_2 + \text{H}$ and the OH is related to the H_2O , then the $\Delta^{17}\text{O}$ of the CO_2 should be the average of the CO and H_2O $\Delta^{17}\text{O}$ values. Remarkably, with these ice and gas compositions, a $\Delta^{17}\text{O} = 86\text{‰}$ for the H_2O and a $\text{CO}_2/\text{CO} \approx 0.5$ for the bulk system (Öberg et al. 2011b), the $\Delta^{17}\text{O}$ behavior as a function of dust enrichment is similar to the $\Delta^{17}\text{O} = 24\text{‰}$ curve for pure H_2O ice in Fig. 2. To some degree this must be happenstance since there are significant uncertainties associated with the astronomical measurements and the assumptions made here. Nevertheless, this explanation is attractive in many ways, but it does mean that the ambient temperatures would have to have remained very low prior to heating to prevent degassing of the ice. The temperatures at which H_2O ices degas depends on their volatile content and grain size (Sandford and Allamandola 1990; Collings et al. 2004; Fayolle et al. 2011), but CO tends to largely degas above 40–50 K and CO_2 at the concentrations required here tends to degas at 70–100 K. Whether these temperatures are realistic for regions of the young disk that would have been affected by periodic intense heat (for more discussion see below) remains to be seen.

One other way to reduce the $\Delta^{17}\text{O}$ of the processed dust is to reduce the fraction of the water that is condensed onto it prior to heating. If slightly less than 30% of the H_2O ($\Delta^{17}\text{O} = 86.4\text{‰}$) in the system was condensed as pure H_2O ice, then the behavior with dust enrichment would be similar to the $\Delta^{17}\text{O} = 24\text{‰}$ curve in Fig. 2. However, to achieve this and to do so relatively consistently would seem to require special circumstances that make it an unattractive option.

While there are at least three potential mechanisms for producing the necessary dust and gas compositions, the enrichments in dust/gas ratios of ≥ 5 –10 that are required are very modest compared to some of the enrichments that have been estimated for chondrule formation (Ebel and Grossman 2000; Alexander 2004; Alexander et al. 2008; Kropf et al. 2009; Alexander and Ebel 2012; Hewins et al. 2012; Fedkin and Grossman 2013), although chondrules have a similar range of Mg#s and O isotopes (e.g., Jones et al. 2005) to the fine-grained silicates. However, the chondrule estimates are based on the need to have stable metal-silicate melts and to efficiently retain volatiles at peak formation temperatures. There is no evidence that metal-silicate melts were present when the fine-grained silicates formed. Unlike chondrules, CI chondrites and IDPs also have roughly solar bulk elemental compositions (other than H, C, N, and noble gases), so there was no loss of even highly volatile elements from the systems during thermal processing. This lack of volatile element fractionation can be simply explained since fine-grained condensates would remain coupled to the gas and their high surface area would enable even the most volatile trace elements to condense efficiently (Alexander 2005a). We would argue that the similar range of O isotopes between chondrules and the fine-grained silicates simply reflects that by the time chondrules formed most of the dust from which they formed had already been thermally processed and had taken on near-terrestrial O-isotopic compositions. It has been suggested that ^{16}O -rich molten CAIs (e.g., type Bs and compact type As) may have formed at dust/gas ratios of only 20–80 times solar if total pressures were as high as $\sim 10^{-3}$ bars (Alexander 2004). Higher dust/gas ratios would be needed at lower pressures. The ^{16}O -rich compositions of CAIs could be simply explained if the ambient temperatures where they formed were above the sublimation temperature of water ice in the nebula (150–170 K). Refractory inclusions in general are rare in chondrite matrices, IDPs, and Wild 2 dust. To avoid making molten CAIs in large numbers when processing interstellar dust into the fine-grained silicates that dominate IDPs, matrices, etc. would suggest that dust/gas ratios never much exceeded 5–10 and/or pressures were significantly below 10^{-3} bars.

If one assumes FeO-poor precursor dust, higher ice/silicate ratios are needed to produce lower Mg#s. Thus, one might expect an inverse correlation between Mg# and $\Delta^{17}\text{O}$ values in individual grains and bulk materials. However, this need not be the case if multiple heating and ice/silicate fractionation events reprocessed the dust, changing the initial silicate and ice O-isotopic compositions and destroying the organics.

This scenario for explaining the near-terrestrial O isotopes of all inner solar system bodies that have been sampled, as well as IDPs and comet Wild 2 dust, has the advantage of not requiring oxidation of the entire inner nebula through migration of ice from the outer solar system as envisaged by Yurimoto and Kuramoto (2004), Lyons and Young (2005), and Ciesla and Cuzzi (2006). Instead, the ^{16}O -poor compositions reflect local conditions in the dust-enriched regions of the early nebula. Heating of the complementary dust-depleted regions would have produced dust with subsolar $\Delta^{17}\text{O}$ values. This dust would also be highly reduced and more refractory since for the same total pressure the condensation temperatures of minerals decrease with decreasing dust concentration. Thermally processed dust from these dust-depleted regions should also be rarer. Searches of fine-grained corundum, spinel, and hibonite for rare grains with subsolar $\Delta^{17}\text{O}$ values (but close to the slope-one O three isotope line, see Fig. 1) might be the best approach to try to look for this material.

ORIGIN OF AMORPHOUS SILICATES— CIRCUMSTELLAR, INTERSTELLAR, AND SOLAR

The crystalline silicates in IDPs probably condensed, for the most part, from a hot gas. Since the silicate dust in the ISM is >98% amorphous (Kemper et al. 2004, 2005), this crystalline material is almost certainly predominantly solar system in origin, which is consistent with its near-terrestrial bulk O-isotopic compositions (Aléon et al. 2009; Keller and Messenger 2011; Nakashima et al. 2012b; Starkey and Franchi 2013). But what is the origin(s) of the amorphous GEMS, and more specifically, what fraction of them may be surviving interstellar grains?

There has been and continues to be a vigorous debate about whether GEMS are solar or interstellar in origin (Keller and Messenger 2011, 2013; Bradley 2013). In aggregate, the crystalline silicates in IDPs do not have a solar bulk elemental composition. Therefore, some fraction of the GEMS must be solar in origin and are the complement to the crystalline material. On the other hand, Keller and Messenger (2011) identified four circumstellar GEMS grains with highly anomalous O isotopes and estimated that such grains make up roughly 1–6% of GEMS. While there are many uncertainties (Jones and Nuth 2011), the lifetimes of grains in the ISM are generally thought to be short relative to the recharge rate from stellar winds (e.g., Jones et al. 1996; Hirashita 2010), implying that most grains in the ISM formed there (e.g., Dwek 1998; Zhukovska et al. 2008; Draine 2009; Pipino et al. 2011; Hirashita 2012; Asano et al. 2013). Hence, the abundance of amorphous silicates in the ISM that formed there should be significantly higher

Table 2. A compilation of the reported crystallinities of circumstellar silicate grains analyzed by TEM (see Appendix for sources).

	IDPs	Meteorites
Amorphous	10	20
Crystalline	6	11
Composite	0	2
Total	16	33
Amorphous (%)	62.5	60.6
Crystalline (%)	37.5	39.4

than the abundance of circumstellar ones that formed in stellar outflows. Unfortunately, the precisions of O-isotopic measurements of individual GEMS has thus far been insufficient to distinguish solar (as expected for ISM-formed dust) from planetary compositions, and we must rely on other constraints to estimate the fraction of noncircumstellar GEMS that are interstellar in origin.

The outflows of O-rich giant and supergiant stars are the major stellar contributors of silicate dust to the ISM in the galaxy. Kemper et al. (2004) found that 10–20% of silicates condensing in the outflows of these stars are crystalline. A comparable fraction, roughly one-third, of presolar circumstellar silicates in IDPs and primitive chondrites are crystalline (Table 2). This similarity suggests that interstellar processing preserves the crystalline/amorphous ratio in circumstellar silicates, for example, favoring complete grain destruction over amorphization, and provides further evidence that most amorphous interstellar silicates formed in the ISM. The upper limit of ~2% for the abundance of crystalline silicates in the ISM can thus be combined with the 10–20% crystallinity of stellar sources of silicate dust to obtain an upper limit of ≤ 8 –18% for the abundance of amorphous circumstellar silicates in the ISM. This means that there is at least 5–12 times as much silicate dust in the ISM that formed there compared to that which formed in stars. Hence, if 1–6% of GEMS are circumstellar (Keller and Messenger 2011), it follows that at least 5–74% of GEMS are interstellar in origin. Unfortunately, this range does not provide the more stringent constraints that we need for the purposes of this paper.

To obtain a better estimate of the fraction of preserved interstellar silicate grains in CP-IDPs, we use the reported chemical compositions of GEMS to first estimate what fraction is interstellar and then estimate the average GEMS content of IDPs. Keller and Messenger (2011) suggested that one way to discriminate between solar and interstellar GEMS was that interstellar GEMS on average should have bulk Mg, Al, Si, Ca, and Fe relative abundances that are close to CI. These authors showed that GEMS are internally

compositionally heterogeneous and that many GEMS have sizes that exceed the nominal thicknesses of the sections used in the transmission electron microscope (TEM) studies (~ 70 nm). Thus, it is likely that in many cases the composition of a GEMS section is not representative of the GEMS as a whole. This will add additional scatter to the GEMS data, but the interstellar GEMS should still be distributed around the CI composition. The GEMS compositions reported by Keller and Messenger (2011), Messenger et al. (2015), and Ishii et al. (2008) exhibit considerable diversity (Fig. 3), and it is likely that at least some of the scatter reflects unrepresentative sampling. The dashed lines in Fig. 3 show the $\pm 20\%$ range around the CI composition. If $\pm 20\%$ of CI is taken as a reasonable range (due to intrinsic variability and unrepresentative sampling) to be expected of interstellar GEMS compositions for Mg/Si, Al/Si, and Ca/Si, then only 2 GEMS out of 47 ($\sim 4\%$) in the Messenger et al. (2015) study and only 1 out of 42 ($\sim 2\%$) in the Ishii et al. (2008) study meet these criteria. Keller and Messenger (2011) did not list the compositions of individual GEMS. If the criteria are relaxed to $\pm 30\%$ (the points labeled CI-like in Fig. 3), the number of possible interstellar GEMS go up to roughly 21% and 19%, respectively, for the two studies. Fewer of these interstellar candidates also have CI-like Fe/Si ratios, but if the metal and sulfide in GEMS preferentially decorate the GEMS surfaces (Keller and Messenger 2011) random sectioning will lead to an underestimation of their abundances and, therefore, the (Fe, Ni, S)/Si ratios of GEMS. Thus, using the roughly CI-like compositional criteria for (Mg, Al, Ca)/Si and combining the two studies suggests that no more than 3–20% of GEMS can be interstellar, that is, at the lower end of the range estimated earlier.

Estimating the range of abundances of interstellar GEMS in bulk CP-IDPs requires the average GEMS content of CP-IDPs. Unfortunately, this quantity has not been reported, but it can be estimated based on the average composition of GEMS and the assumption that they complement the crystalline material to give a solar- or CI-like bulk CP-IDP composition. For instance, if where the GEMS condensed the dominant crystalline phase was forsterite, then the concentrations of the incompatible elements Al and Ca in the complementary GEMS can be used to estimate the mass fraction of olivine that had formed.

Figure 4 shows the average GEMS compositions from three studies (Ishii et al. 2008; Keller and Messenger 2011; Messenger et al. 2015) and the directions of trends that the GEMS compositions would follow if only a single type of mineral were fractionated from the CI bulk composition. We have attempted to explain the mean

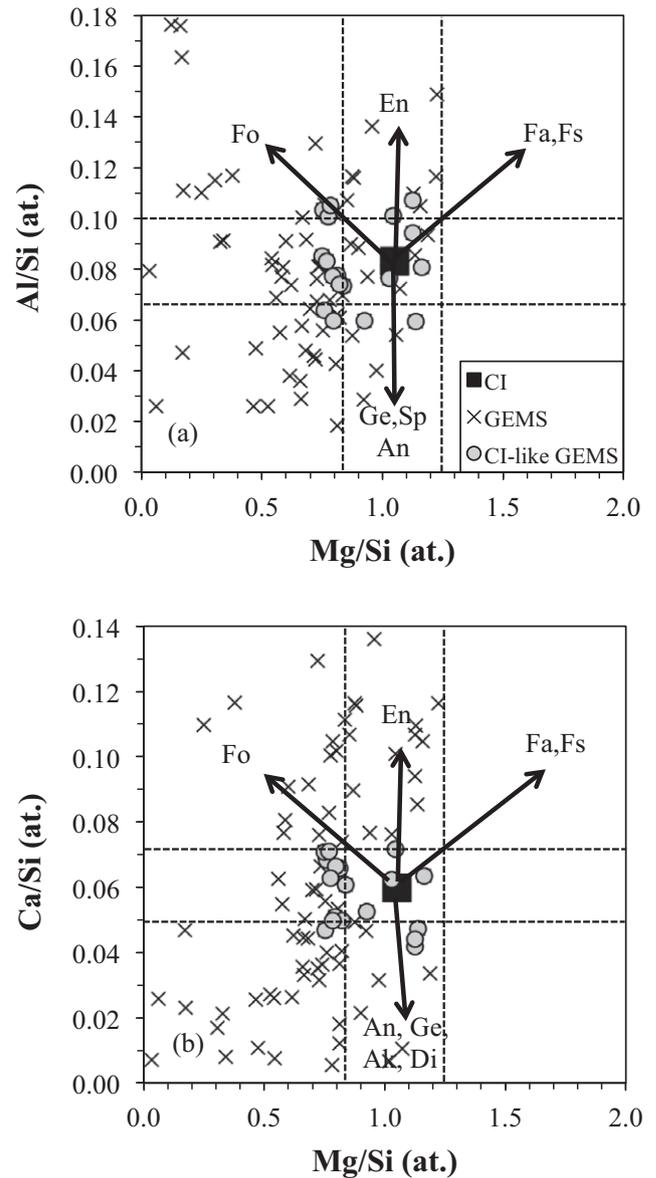


Fig. 3. The elemental compositions of individual GEMS (Ishii et al. 2008; Messenger et al. 2015). The dashed lines are for $\pm 20\%$ of the CI composition. The filled circles are the GEMS with the most CI-like ($< 30\%$ deviation) compositions in all three ratios (Mg/Si, Al/Si, and Ca/Si).

GEMS compositions as products of fractional condensation with three very simplistic models. All three of the GEMS studies discussed here have very similar average Mg/Si ratios. In Fig. 4a, one of the two measurements of bulk IDP compositions (Table 3) and the mean GEMS composition of Ishii et al. (2008) fall close to the line for forsterite fractionation. So in Model 1 the GEMS compositions were calculated (Table 3 and Fig. 4) by fractionating forsterite, FeS, and Fe,Ni metal from the CI composition to roughly reproduce the average Mg/Si, Fe/Si, and S/Si ratios for GEMS from the three GEMS

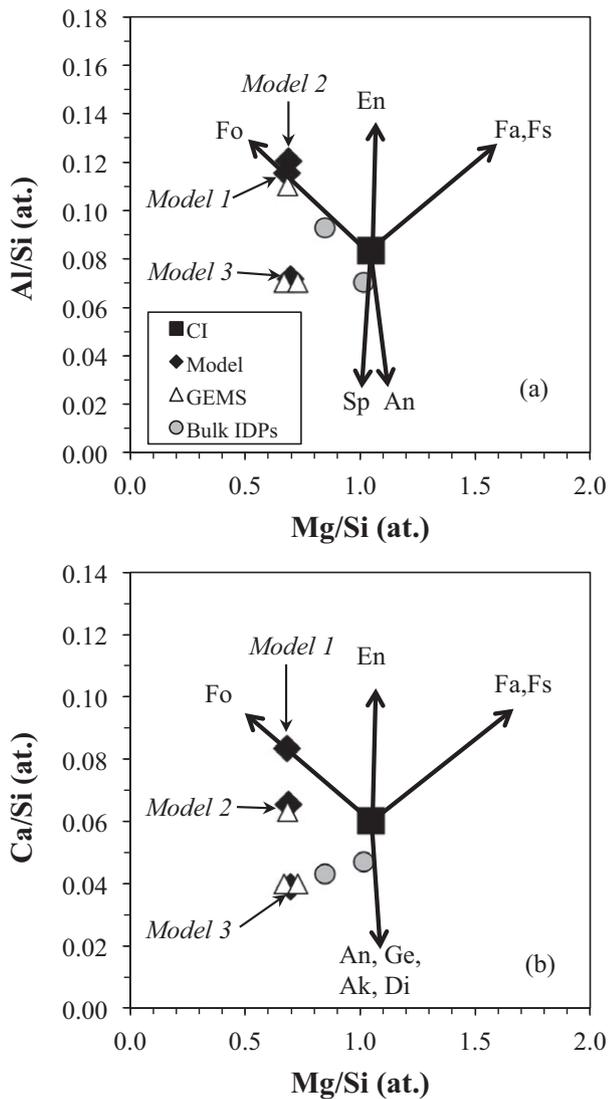


Fig. 4. The average compositions of GEMS from three studies (Ishii et al. 2008; Keller and Messenger 2011; Messenger et al. 2015), along with three model GEMS compositions. Also shown are two determinations of mean bulk IDP compositions (Schramm et al. 1989; Thomas et al. 1993). The arrows show the directions GEMS compositions would follow assuming that they formed from the material remaining after fractionation from an initially CI composition of forsterite (Fo), enstatite (En), fayalite (Fa), ferrosilite (Fs), anorthite (An), diopside (Di), spinel (Sp), gehlenite (Ge), and åkermanite (Ak). The three model compositions involve fractionation of forsterite (Model 1), forsterite and diopside (Model 2), and forsterite and gehlenite (Model 3). Models 2 and 3 reproduce the mean GEMS compositions quite well. However, note that in Fig. 3 the individual GEMS compositions exhibit considerable scatter, with little evidence for the trends that might be expected for simple one- or two-mineral fractionation.

studies. In this case, ~47% of the Mg, ~72% of the Si, and 40% of the Fe remain in the GEMS. However, the model does not reproduce the Ishii et al. (2008) mean Ca/

Si ratio (or the nonsolar mean Ca/Al ratio), which can be explained by the additional fractionation of a small amount of diopside (Model 2). Despite their similar Mg/Si ratios, there are marked differences in the average Al/Si and Ca/Si ratios in GEMS between those of Ishii et al. (2008) and the other two GEMS studies. The average Keller and Messenger (2011) and Messenger et al. (2015) compositions have roughly solar Ca/Al ratios but fractionated (Ca,Al)/Si ratios, and can be most simply explained by fractionation of a small amount of gehlenite (Model 3) with its roughly solar Ca/Al ratio from the Model 1 composition.

While Models 2 and 3 can reproduce the measured average GEMS compositions, this may largely be a coincidence as there is considerable scatter in the individual GEMS compositions from all three studies and there are no obvious compositional trends. The modeling is also a gross oversimplification. For instance, olivine grains in IDPs are not generally pure forsterite and low-Ca pyroxene is common. To reproduce the GEMS Mg/Si ratios will require the formation of more Fe-bearing (lower Mg/Si) olivine and low-Ca pyroxenes than for condensation of pure forsterite alone. Thus, estimates of GEMS abundances in CP-IDPs based on the above models are almost certainly upper limits. It is not clear whether diopside or gehlenite are as common in IDPs as required by the models. The Al/Si and Ca/Si fractionations in GEMS could be reproduced by a mixture of refractory minerals (e.g., a CAI-like composition) rather than by diopside or gehlenite alone, but at present there are little data to constrain this. Until more detailed information about mineral and GEMS compositions and abundances in IDPs become available, more complex models cannot be justified. The Na contents of GEMS would be particularly useful, although difficult to measure, as Na should be incompatible in almost all minerals that are predicted to condense at high temperatures. The same is true of Ti unless perovskite or Ti-rich pyroxene formed during condensation. Finally, there is also a clear difference in the distributions of the individual GEMS compositions between the results of Ishii et al. (2008), and those of Keller and Messenger (2011) and Messenger et al. (2015). The reasons for these differences will need to be resolved.

The scatter in the GEMS data, the disagreements between the studies, and the fact that even the bulk CP-IDP measurements are not CI like (possibly due to unsampled coarser grained refractory components and/or modification during atmospheric entry and settling through the atmosphere) makes it difficult to estimate the average GEMS fraction in IDPs with any certainty. Nevertheless, assuming that all S in GEMS is in FeS, that all Ni in GEMS is in metal with an Fe/Ni = 1

Table 3. The elemental ratios (at.) of CI chondrites, bulk CP-IDPs, and glass with embedded metal and sulfide (GEMS), as well as model GEMS compositions.

	CI ¹	IDP ²	IDP ³	GEMS ⁴	GEMS ⁵	GEMS ⁶	Model 1	Model 2	Model 3
Mg/Si	1.047	1.015	0.847	0.670	0.651	0.730	0.681	0.689	0.696
Al/Si	0.083	0.070	0.093	0.070	0.113	0.070	0.115	0.120	0.071
S/Si	0.437	0.417	0.409	0.300	0.256	0.310	0.290	0.303	0.297
Ca/Si	0.060	0.047	0.043	0.040	0.058	0.040	0.083	0.065	0.039
Fe/Si	0.871	0.705	0.606	0.560	0.444	0.560	0.485	0.505	0.496
Ni/Si	0.049	0.024	0.022	0.030	0.028	0.030	0.027	0.028	0.028
						Forsterite	22	22	23
						FeS	8	8	8
						Metal	4	4	4
						Diopside		4	
						Gehlenite			3
						GEMS	65	62	62

¹Asplund et al. (2009).

²Schramm et al. (1989), porous particles only.

³Thomas et al. (1993).

⁴Keller and Messenger (2011).

⁵Ishii et al. (2008).

⁶Messenger et al. (2015).

The model GEMS compositions were calculated by fractionating forsterite, FeS, and Fe,Ni metal (Fe/Ni = 10) from the CI composition to roughly reproduce the reported average GEMS Mg/Si ratios (Model 1), along with diopside (Model 2) or gehlenite (Model 3) to reproduce the GEMS Al/Si and Ca/Si ratios. Also given are the volume percent of the phases.

(minimum amount of metal, but results are not very sensitive to this ratio), and that the partial molar volumes of the oxides in the GEMS silicates are similar to those in FeO-bearing silicate melts extrapolated to 1000° C (Lange and Carmichael 1987), the models (Fig. 4 and Table 3) indicate GEMS volume abundances in CP-IDPs of 62–65%. Volume abundances are given here as they are the most readily observed physical quantity in TEM studies. Some GEMS will have been destroyed by the heating that produced equilibrated aggregates (Brownlee et al. 2005), but this will likely result in only a modest correction. Using the earlier compositional estimate of the fraction of GEMS that are interstellar, the estimated abundance of GEMS in CP-IDPs translates to a rough estimate of the abundance of interstellar dust in bulk CP-IDPs of ≤ 2 –13%. Ultimately, this estimate will need to be confirmed by searching for GEMS with solar-like O isotopes.

Combining the estimate of 1–6% of circumstellar GEMS with the upper limit of GEMS abundances in CP-IDPs gives a range of bulk circumstellar GEMS abundances in CP-IDPs of ~ 0.6 –4%. This range is higher than is typically reported for total circumstellar silicate abundances in CP-IDPs (0–1.5%, see Appendix for details). Part of the reason for the wide range in the reported circumstellar abundances in IDPs is the small areas that are typically measured compared to studies of chondrite matrices. Floss et al. (2006) conducted the most extensive study of IDPs to date and obtained an

average circumstellar abundance of 120 ppm, increasing to 380 ppm if only the “primitive” ¹⁵N-rich subgroup of CP-IDPs is considered. Thus, even among IDPs there is a range of apparent primitiveness that must at least in part reflect heterogeneity at the scales sampled, but probably also reflects variable extents of modification in the nebula, in parent bodies, and during atmospheric entry. The average area-weighted circumstellar silicate abundance in IDPs based on all the studies listed in the Appendix is ~ 400 ppm. This abundance of circumstellar silicates is similar to the highest reported chondrite matrix abundance of 240 ± 25 ppm (Nittler et al. 2013), suggesting the intriguing possibility that chondrite matrices might be composed of materials that, like IDPs, had a range of primitiveness.

The above discussion suggests that Keller and Messenger (2011) overestimated the typical abundance of circumstellar GEMS in CP-IDPs. On average, circumstellar silicate abundances in IDPs appear to be closer to 400 ppm, with circumstellar GEMS abundances being roughly 60% of this, although circumstellar abundances can in some small area studies reach up to 1.5 area%. As an aside, if our estimate for the abundance of interstellar silicates in IDPs is correct, then circumstellar grains make up ≤ 0.4 –2% of interstellar silicates, with crystalline silicates making up roughly a third of this. These circumstellar abundances are significantly lower than was estimated earlier using the 2% crystallinity upper limit for the DISM. In addition, the abundances of circumstellar oxides and SiC would be about a tenth

of the circumstellar silicate abundances (e.g., Leitner et al. 2012; Davidson et al. 2014). This SiC abundance is well below the limit of <5% of the ISM silicate abundance estimated by Whittet et al. (1990).

THE ORIGIN(S) OF CHONDRITIC WATER

If significant abundances of ISM silicates survived the formation of the solar system, the same may also be true for interstellar ices and carbonaceous grains. The water ice accreted by chondritic meteorites (Alexander et al. 2012), comets (Hartogh et al. 2011; Bockelée-Morvan et al. 2012; Lis et al. 2013; Altwegg et al. 2015), and Saturn’s moon Enceladus (Waite et al. 2009) are all significantly enriched in D compared to the bulk solar value. Water ices that formed in cold molecular clouds are very D-rich (e.g., Lerate et al. 2006; Bergin et al. 2010; Liu et al. 2011; Coutens et al. 2012; Persson et al. 2014), but models suggest that water that formed in the solar nebula, even in its cold outer regions, is likely to have a roughly solar D/H ratio (Yang et al. 2013; Cleeves et al. 2014, 2016). Thus, the elevated D/H ratios of water accreted by those planetesimals that have been measured are the products of mixing of water ice inherited from the ISM and water produced in the solar nebula, the latter being the product of high temperatures that re-equilibrated it with H₂ either in the inner disk (Jacquet and Robert 2013; Yang et al. 2013) and/or at high altitudes farther out in the disk (Furuya et al. 2013; Albertsson et al. 2014).

The water D/H ratios in star-forming regions appear to be quite variable ($D/H \approx 3 \times 10^{-4}$ to 10^{-2}) (Lerate et al. 2006; Bergin et al. 2010; Liu et al. 2011; Coutens et al. 2012; Persson et al. 2014) allowing for considerable latitude in estimates of ISM water fractions, although $D/H \approx 10^{-3}$ is a commonly assumed value. We can constrain the D/H ratio of the interstellar water accreted by the solar system somewhat using the properties of comets (Table 4). If D-enriched H₂O cannot be generated in the solar nebula, then a lower limit of $\sim 5\text{--}6 \times 10^{-4}$ for the D/H ratio of the interstellar H₂O is given by the most D-rich comets (67P and F6 Lemmon, Table 4).

It is worth noting that comets contain both solids (crystalline grains) and vapor (low D/H water) that were produced in the solar nebula at elevated temperatures. If we interpret the presence of such materials in the outer solar system as arising from transport or mixing from the inner regions, close to the Sun, this means that the transport process operated on both the gas and solid components in the disk. Transport models largely come in two varieties:

“within-disk” processes (e.g., diffusion, outward advective flows, and gravitational torques) and “above-disk” processes (e.g., X-wind and magneto-centrifugal winds). The “within-disk” models would operate equally well on fine dust and gaseous species, and would thus be consistent with observations. The “above-disk” models, however, would fractionate vapor and solids, as both could be carried away from the disk in gaseous flows, but the solids would eventually decouple from the gas and fall back under the effect of gravity while water vapor, for instance, would remain entrained and be lost from the disk. As “within-disk” models are most consistent with comet properties, this would imply that amorphous silicate fractions provide upper limits for the amounts of interstellar water in comets. This is because outward transport would be more efficient at mixing low D/H water into the outer solar system as this water forms at temperatures as low as $\sim 400\text{--}500$ K as opposed to crystalline silicates that require $\geq 800\text{--}1000$ K and must originate much closer to the Sun. The estimates for the amorphous fractions of silicates in various comets fall in the range of $\sim 30\text{--}80\%$ (Kelley and Wooden 2009), providing upper limits for the amounts of interstellar silicates in comets. One might expect the fraction of interstellar water in comets to be similar to that of interstellar silicates, and the range of estimates of interstellar water abundances in comets made assuming an ISM D/H ratio of 10^{-3} is roughly consistent with the range of amorphous silicate abundances (Table 4), that is, $D/H = 10^{-2}$ would be too high. Thus, for the present we will adopt the commonly used ISM water value of $D/H = 10^{-3}$.

With this value for the ISM water, recent estimates of the D/H ratio of water accreted by CI and CM chondrites (Alexander et al. 2012) would require that they contained roughly 5–8% ISM water. The ungrouped carbonaceous chondrite Tagish Lake, which has been linked spectroscopically to the very primitive D-type asteroids (Hiroi et al. 2001), would require 3–5% ISM water. The CR chondrites have water D/H ratios that are slightly above terrestrial, and the ordinary and R chondrites have ratios that are similar to comets (McCanta et al. 2008; Alexander et al. 2012). It is possible that the more D-rich compositions of the water in these meteorites, compared to the CIs, CMs, and Tagish Lake, reflect higher ISM water contents (e.g., Deloule et al. 1998; Piani et al. 2015). However, there is no evidence that they accreted higher abundances of presolar circumstellar grains or the possibly presolar organic material than the CIs, CMs, and Tagish Lake. A more mundane explanation for the elevated D/H ratios in their water is that there was D/H fractionation associated with the oxidation of metal by

Table 4. The D/H ratios of water in Oort Cloud (OCC) and Jupiter Family (JFC) comets, the estimated fractions of interstellar water (F_{ISM}) they contain assuming that they are mixtures of water with solar (D/H = 2×10^{-5}) and interstellar (D/H = 1×10^{-3}) H isotopic compositions, and the fractions of their silicates that are amorphous (F_{amor}) based on spectroscopic observations. The estimated D/H ratios for the solar nebula, bulk Earth, and water in Enceladus are also provided for comparison.

	D/H	F _{ISM}	F _{amor} .
OCCs			
1P/Halley ¹	$3.16 \pm 0.34 \times 10^{-4}$	0.30	
1P/Halley ¹	2.12×10^{-4}	0.20	
C/1996 B2 Hyakutake ²	$2.90 \pm 1.0 \times 10^{-4}$	0.28	
C/1995 Hale-Bopp ³	$3.30 \pm 0.8 \times 10^{-4}$	0.32	0.2–0.4
C/2002 T7 LINEAR ⁴	$2.50 \pm 0.4 \times 10^{-4}$	0.23	
8P/Tuttle ⁵	$4.09 \pm 1.45 \times 10^{-4}$	0.40	
C/2009 P1 Garradd ⁶	$2.06 \pm 0.22 \times 10^{-4}$	0.19	
C/2001 Q4 Neat ⁷	$4.60 \pm 1.4 \times 10^{-4}$	0.45	0.3
153P/Ikeya-Zhang ⁸	$<2.50 \times 10^{-4}$	0.23	
C/2014 Q2 Lovejoy ⁹	$1.40 \pm 0.4 \times 10^{-4}$	0.12	
C/2012 F6 Lemmon ⁹	$6.50 \pm 1.6 \times 10^{-4}$	0.64	
JFCs			
103P/Hartley 2 ¹⁰	$1.61 \pm 0.24 \times 10^{-4}$	0.14	
67P/Churyumov-Gerasimenko ¹¹	$5.30 \pm 0.7 \times 10^{-4}$	0.52	
45P/Honda-Mrkos-Pajdušáková ¹²	$<2.00 \times 10^{-4}$	<0.18	
Other			
Solar ¹³	$2.1 \pm 0.5 \times 10^{-5}$	0	
Earth ¹⁴	$1.49 \pm 0.03 \times 10^{-4}$	0.13	
Enceladus ¹⁵	$2.9 + 1.5/-0.7 \times 10^{-4}$	0.28	

¹Eberhardt et al. (1995), Brown et al. (2012).

²Bockelée-Morvan et al. (1998).

³Meier et al. (1998), Harker et al. (2004).

⁴Hutsemékers et al. (2008).

⁵Villanueva et al. (2009).

⁶Bockelée-Morvan et al. (2012).

⁷Altwegg et al. (2015), Wooden et al. (2004).

⁸Biver et al. (2006).

⁹Biver et al. (2016).

¹⁰Hartogh et al. (2011).

¹¹Altwegg et al. (2015).

¹²Lis et al. (2013).

¹³Geiss and Gloeckler (1998).

¹⁴Lécuyer et al. (1998).

¹⁵Waite et al. (2009).

water in the chondrite parent bodies (Alexander et al. 2010). This effect would have been largest in the chondrite parent bodies with the lowest H₂O/metal ratios at the time of accretion, and when corrected for, albeit with large uncertainties, the water compositions become closer to those of CI and CM chondrites (Sutton et al. 2013). Hence, we consider that the ISM water fractions estimated for the CI and CM chondrites to be the most reliable.

ORIGIN OF REFRACTORY ORGANICS IN CHONDRITES AND COMETS

Whether the IOM in chondrite matrices is interstellar or solar is a matter of considerable debate (Robert and

Epstein 1982; Yang and Epstein 1983; Alexander et al. 2007b; Gourier et al. 2008). We know that carbonaceous dust is abundant in the DISM, and the isotopic evidence suggests that some interstellar water survived in the early solar system (see previous section). Erasing the interstellar O and H isotopic signatures in water through gas phase reactions would probably occur at comparable or lower temperatures than those required to modify or destroy refractory organic matter in dust. Thus, it would seem almost unavoidable that some interstellar carbonaceous dust survives in chondrites, IDPs, and comets. Assuming that the IOM in chondrites was originally interstellar, Alexander (2005b) used the abundance of IOM in CIs to infer that $\geq 75\%$ of CI material was thermally processed in the solar

system. More recent estimates of the Sun's composition (Asplund et al. 2009) and the diffuse ISM dust composition (Compiègne et al. 2011; Jones et al. 2013) warrant a recalculation of this estimate.

In the Compiègne et al. (2011) dust model for the DISM, ~50% of the cosmic C is in dust (45% 10–500 nm in radius, 5% 1–10 nm in radius) and a further 24% is in free PAHs. It is interesting to note that nanodiamonds with a median radius of ~1.3 nm (Daulton et al. 1996) make up ~3–5% of the C in IOM residues (Alexander et al. 1998). Assuming that the Mg and Si are completely condensed in the DISM, this means that the solar-normalized atomic C/Mg and C/Si ratios for the DISM dust are 0.5, and 0.74 if the PAHs are included. Much of the carbonaceous dust in the ISM appears to be much finer grained than is generally reported in the meteorites—roughly half the mass is in particles that are ≤ 130 nm in radius. However, this apparent difference between the interstellar and meteoritic particle sizes may in part be due to the spatial resolution of most of the techniques used to study the meteoritic material. Recent TEM studies suggest that there is significant finely dispersed C in primitive meteorites, although the fraction of the C that this may represent has not been quantified and some or all of it may be soluble organic C (Le Guillou et al. 2014). It is also possible that processes in the presolar molecular cloud (e.g., Hirashita and Li 2013), in the solar nebula, and in the meteorites caused the dust, and possibly even the PAHs, to aggregate to form larger grains.

To place limits on how much interstellar carbonaceous dust may have survived solar system formation, we assume that (1) the condensed C contents of the DISM were those of the ISM material from which the solar system formed, (2) that the organic C contents of CIs (i.e., excluding carbonates) provide an upper limit for the fraction of ISM carbonaceous dust that survived thermal processing at least in the inner disk, and (3) that in the DISM and the CI formation region Mg and Si were fully condensed. With these assumptions, we can compare C/Mg and C/Si ratios in CIs and the DISM, as well as in IDPs and comet Halley dust (Table 5). The CI ratios depend on whether one uses a C content of ~2 wt% based on the amount of IOM that is typically extracted (e.g., Alexander et al. 2007b), or the bulk C content of ~3.5 wt% after subtracting the small amount of C in carbonate. The 3.5 wt% C includes the mystery C that is insoluble in solvents but is lost during IOM isolation (Smith and Kaplan 1970; Alexander et al. 2015). The mystery C is not carbonate, is present in all primitive chondrites, and appears to be compositionally and isotopically similar to IOM (Alexander et al. 2015). This mystery C is

Table 5. The atomic C/Mg and C/Si ratios in CI chondrites, CP-IDPs, Halley dust, and models for diffuse ISM dust.

	CI ¹	IDP ²	IDP ³	Halley ⁴	Halley ⁵	ISM ⁶	ISM ⁷
C/Mg	0.74	2.35	2.39	8.14	4.12	3.98	5.90
C/Si	0.77	2.39	2.00	4.40	1.98	4.17	6.17

¹Total organic C = 3.5 wt%, otherwise from Lodders (2003).

²Only the porous particles of Schramm et al. (1989).

³Thomas et al. (1993).

⁴Jessberger et al. (1988).

⁵Schulze et al. (1997).

⁶Model DISM dust, excluding PAHs (Compiègne et al. 2011).

⁷Model DISM dust, including PAHs (Compiègne et al. 2011).

probably either very fine-grained IOM (<100 nm) and, therefore, hard to recover, or is associated with IOM but converted to soluble/volatile material when exposed to strong acid. The CI ratios in Table 5 use the 3.5 wt% C value, but below we also consider the possibility that the ~2 wt% of C in recoverable IOM is more appropriate. As already mentioned, in the DISM dust model of Compiègne et al. (2011) ~45% of the total C is in 10–500 nm grains, 5% in 1–10 nm grains, and 24% is in free PAHs. Since nanodiamonds are a component of the C in CIs, the first ISM dust composition in Table 5 includes all the model DISM dust. The IOM is also rich in PAHs, so as an upper limit on the C content of the dust from which the solar system formed, the second ISM dust composition includes the PAHs. From these compositions, we can estimate that the CI C contents of 2–3.5 wt% would be consistent with survival of ~11–19% of ISM dust if PAHs were not part of the ISM dust, and decreases to ~7–13% if the PAHs were incorporated into the ISM dust.

RECONCILING THE VARIOUS ESTIMATES

The estimate that ~7–19% of the original interstellar carbonaceous dust has survived in CI chondrites (previous section) is more or less consistent with our estimates of the fractions of interstellar water (5–8%) surviving in chondrites and of interstellar silicates surviving in CP-IDPs ($\leq 13\%$) (Fig. 5). This adds plausibility to the argument that chondritic organics are largely interstellar in origin. However, the average C contents of IDPs are 2.5–3 times that for CI chondrites (Table 5), which if taken at face value implies between at least ~50% and ~80% survival of interstellar grains in them. Thus, there is an inconsistency in the extent of survival of interstellar material in IDPs depending on whether one assumes that all ISM silicate grains have roughly CI-like compositions or whether one assumes

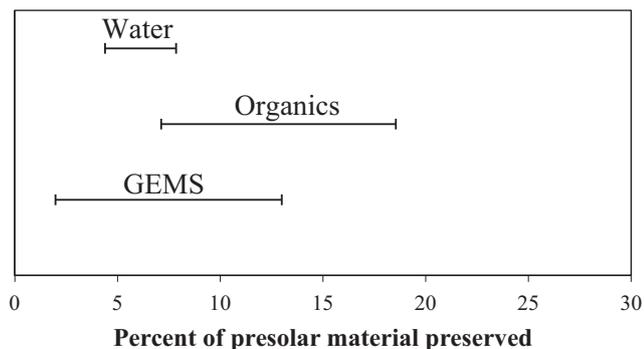


Fig. 5. A summary of the ranges estimated for the fractions of presolar material accreted by CI chondrites, chondrite matrices, and IDPs. These estimates are based on the H isotopic compositions of water, the insoluble organic C contents, and the abundances of GEMS in IDPs with roughly CI bulk compositions. While significant uncertainties remain and there is no reason that these different materials should have been preserved to exactly the same degree, the survival of ~5–10% of the original interstellar material seems a fairly robust conclusion.

that the C in IDPs is interstellar. One possible explanation for this inconsistency is that to survive atmospheric entry without being severely heated biases the IDP population toward lower density particles with high C contents, that is, IDP C contents are not representative of their bulk parent bodies.

When measuring bulk IDPs, Schramm et al. (1989) and Thomas et al. (1993) studied particles that were typically 6–20 μm across. Interplanetary dust particles in this size range also tend to be enriched in highly volatile elements compared to both CI and large cluster IDPs by on average a factor of four (Flynn et al. 2009). It is unclear whether C is also enriched in small IDPs compared to large cluster IDPs, but Flynn et al. (2009) attributed the highly volatile element enrichments in smaller IDPs to the presence in the cluster IDPs of >70% of large, volatile-poor mineral grains (e.g., forsterite, enstatite, FeS, etc.). If correct, this would mean that the C contents of the more representative cluster IDPs are similar to those of CI chondrites. It would also require reducing our estimates of interstellar silicate abundances in IDPs by about a factor of three to $\leq 0.6\text{--}4\%$. However, the formation of so much forsterite and enstatite is hard to reconcile with the compositions of GEMS (see Origin of Amorphous Silicates—Circumstellar, Interstellar, and Solar section), unless GEMS are also volatile-poor and a significant fraction of them adhere to the large crystals or are otherwise underrepresented in the small IDPs. This does raise the issue of what the carrier(s) of the highly volatile elements in IDPs might be, but that is beyond the scope of this study. GEMS grains, including

circumstellar ones, being associated with the large crystals might also help alleviate a disagreement between circumstellar grain abundances in IDPs and chondrite matrices since otherwise the mean circumstellar grain abundance found in small IDPs (see Origin of Amorphous Silicates—Circumstellar, Interstellar, and Solar section) will have to be reduced from ~400 ppm to ~130 ppm.

While acknowledging that a number of uncertainties remain, we believe that the estimates of interstellar grain abundances in CI chondrites, chondrite matrices, and the IDP parent bodies of 7–19% from CI organic C contents and $\leq 2\text{--}13\%$ based on GEMS elemental compositions in IDPs are valid. These estimates, along with the estimated 5–8% ISM water in chondrites, suggest a most likely abundance of preserved interstellar matter in these materials of ~5–10% (Fig. 5). However, there is no reason that interstellar water, carbonaceous dust, and GEMS should have been preserved at exactly the same levels, and it is possible that the different ranges reflect to some extent their relative degrees of robustness in the disk.

The dust from comet Halley seems to be even more C-rich than small IDPs, although the abundances are very uncertain as the two different analyses in Table 5 illustrate. Indeed, based on the estimates of their C content, the Halley dust could be almost entirely interstellar in origin. However, Halley does contain crystalline silicates (Bregman et al. 1987; Campins and Ryan 1989). That the fraction of surviving interstellar material varied as a function of radial distance from the Sun and/or time should not be too surprising. In fact, variable abundances of interstellar material would seem to be required by the higher D/H ratios in the water of many comets (Table 4) compared to chondrites. Much of the heating in the disk would almost certainly have occurred in the inner disk and the processed material then transported outward, naturally generating a radial gradient. Also, accretion of molecular cloud material onto the disk will have continued for some time after formation of the disk, and the amount of accreted material will have varied as a function of radius (e.g., Dullemond et al. 2006; Visser et al. 2009; Yang et al. 2013).

THE HEAT SOURCE FOR EARLY PROCESSING OF DUST

The very early establishment of dust with $\Delta^{17}\text{O}$ near 0‰ is supported by the recent suggestion that the parent bodies of iron meteorites formed 0.1–0.3 Myr after CAIs (Kruijer et al. 2014). These iron meteorite parent bodies all had O-isotopic compositions that were close to terrestrial. Many of them also contained abundant volatile elements (e.g., Ga and S), which is

consistent with the low ambient midplane temperatures that would be necessary to condense water ice and concentrate it along with the silicate dust. The fact that this processed dust was widely dispersed in the solar nebula is indicated by the nature of the dust returned from comet Wild 2 (e.g., Gounelle et al. 2008; Ishii et al. 2008; Zolensky et al. 2008) and the presence of crystalline silicates in other comets (e.g., Wooden 2008; Hanner and Zolensky 2010).

What was the heat source responsible for producing the crystalline silicates and the majority of GEMS? The apparently common mixed ortho- and clinomicrostructure of low-Ca pyroxene in IDPs, chondrite matrices, and comet Wild 2 samples suggest that cooling rates were relatively rapid when they formed, requiring a local rather than global heating mechanism (Scott and Krot 2005; Alexander et al. 2007a). However, there have been no systematic studies to confirm this. Nevertheless, transient heating events would seem to be required if ambient temperatures were low enough for water to be condensed so that it could be concentrated, along with silicates, relative to the gas.

Chondrule formation is one possible mechanism for producing the fine-grained material (Scott and Krot 2005). However, if it is true that chondrite matrices, IDPs, and comet dust are all genetically related, there is no obvious relationship between the abundances/compositions of the fine-grained material, and the abundances/compositions of the chondrules in the same samples (e.g., their Mg#s). In addition, chondrules are chemically fractionated, most dramatically in the depletions of their volatile elements (e.g., Grossman and Wasson 1983). However, as already discussed, the matrices of chondrites at the time of their accretion appear to have been dominated by a roughly CI-like component, including highly volatile elements, organic matter, and circumstellar grains (Alexander et al. 2001; Alexander 2005b; Zanda et al. 2009, 2011, 2012); that is, they are neither chondrule-like nor the volatile enriched complements to chondrules as has been suggested by some (e.g., Alexander et al. 1989a; Palme et al. 2015). Also, most chondrule formation seems to have occurred 1–3 Ma after the formation of the solar system (e.g., Kurahashi et al. 2008; Villeneuve et al. 2009; Connelly et al. 2012; Ushikubo et al. 2013), making the dispersal of the fine-grained material throughout the nebula hard to explain.

The formation of most or all refractory inclusions occurred very early in solar system history and the formation timescales of type B CAIs, at least, were relatively brief (Stolper and Paque 1986). However, refractory inclusion formation produced even more extreme elemental fractionations than chondrules, and refractory inclusions are ^{16}O -rich. Hence, the ^{16}O -poor composition of the fine-grained material rules out any genetic relationship with the refractory inclusions.

Thus, it is more likely that the heating that produced the fine-grained crystalline silicates involved a process(es) that was distinct from chondrule and CAI formation, but also active in the early phases of the solar protoplanetary disk. Heating during the very earliest phase of disk formation seems unlikely as accretion from the molecular cloud envelope would have been close to the protosun where temperatures were high and there would have been little opportunity for dust + ice fractionation from the gas. The FU Orionis type outbursts that are thought to occur slightly later in disk evolution are a more attractive possibility. Multiple outbursts are thought to occur in the first 10^5 – 10^6 yr of a disk's life and last for 10–100 yr (Hartmann and Kenyon 1996; Zhu et al. 2009; Liu et al. 2016). FU Orionis outbursts are thought to result from the disk becoming gravitationally unstable as material builds up in it. The sudden burst of accretion onto the star leads to heating of much of the inner ~ 1 AU of a disk (Zhu et al. 2007, 2009). Presumably, further out in the disk shockwaves associated with spiral density waves would produce brief, more localized heating. While most mass transport is inward in a gravitationally unstable disk, there can be some outward transport as well (Boss et al. 2012; Boss 2013).

SUMMARY AND CONCLUSIONS

Evidence for the reworking of interstellar materials (e.g., the presence of crystalline silicates) can be seen in protoplanetary disks and in primitive solar system materials. This reworking must be a fundamental process in disk formation and evolution, but how, when, and to what extent it occurs is still largely unknown. Here, we have used various features of primitive solar system materials to try to constrain these unknowns.

Most extraterrestrial materials (meteorites, IDPs, comet Wild 2 samples) have a relatively restricted range of mass-independently fractionated O-isotopic compositions that lie near the terrestrial value ($\Delta^{17}\text{O} = 0\text{‰}$) rather than the solar value ($\Delta^{17}\text{O} \approx -29\text{‰}$). The principal exceptions to this are refractory materials (e.g., CAIs, AOAs, and refractory oxides) that have $\Delta^{17}\text{O}$ values that are much closer to the solar composition. Here, we argue that the nonsolar and near-terrestrial O-isotopic compositions were established in nebula dust very early by heating of regions of the disk that, compared to expectations for a solar elemental composition, were modestly enriched (≥ 5 – 10) in silicates ($\Delta^{17}\text{O} \approx -29\text{‰}$) and water-dominated ice ($\Delta^{17}\text{O} \approx 24\text{‰}$) relative to the gas. Such dust enrichments can be produced by grain growth and settling to the midplane in a moderately turbulent disk. This scenario requires

that ambient conditions in the affected regions of the disk were below the sublimation temperature of water ice (~150–170 K). Heating of dust in regions where ambient temperatures were above the sublimation temperature of water ice would have produced materials with more solar-like O-isotopic compositions, possibly explaining the compositions of refractory materials. We suggest that the episodic heating of the disk associated with FU Orionis outbursts (or their equivalent in Class 0 protostars) in the first few hundred thousand years of solar system history was the likely cause of the heating. The gravitational instabilities that are thought to be responsible for the outbursts can transport material efficiently through the disk. While most mass transport during an outburst is inward, some fraction of material is transported outward to the comet-forming regions. Given that this thermal processing must happen very early in solar system history, it seems more likely that the ^{16}O -depleted composition of the ice was inherited from the protosolar molecular cloud rather than generated in the solar nebula.

The presence of circumstellar grains in chondrites, IDPs, and comet Wild 2 samples show that this thermal processing did not destroy all interstellar material. Models suggest that large D enrichments in water cannot be generated in disks. Therefore, the nonsolar D/H ratios of water in chondrites and comets point to the survival of some D-rich interstellar water. The D/H ratio of interstellar ice is typically assumed to be $\sim 10^{-3}$, in which case the water accreted by the CI and CM chondrites (and Tagish Lake) contained $\sim 5\text{--}8\%$ interstellar water. The higher D/H of water in CR, O, and R chondrites could indicate that they accreted higher proportions of interstellar ices. However, these chondrites do not contain higher abundances of circumstellar grains in their matrices, so we prefer a parent body explanation for their elevated D/H ratios. On the other hand, the elevated D/H of comets cannot be explained in this way and their interstellar ice contents must be higher than in chondrites.

If some interstellar ices survived solar system formation, then presumably other more refractory materials did as well. The two major components of diffuse interstellar dust are amorphous silicates and carbonaceous dust. Adopting recent models for the amount of C in diffuse interstellar dust and assuming that all insoluble organic matter is interstellar in origin, we estimate that 7–19% of interstellar carbonaceous dust survives in CI chondrites and chondrite matrices. Again the amounts of interstellar C that survive in comets may be significantly higher. The amounts of interstellar silicates that survived in the nebula were the most difficult to estimate. Nevertheless, assuming that interstellar silicates have roughly CI compositions, we estimate from the range of published GEMS compo-

sitions that $\leq 2\text{--}13\%$ of interstellar silicates survive in IDPs. Amorphous silicates were important components of chondrite matrices at the time of their accretion and there are clear genetic links between IDPs and chondrite matrices. Hence, we argue that interstellar silicate abundances would have been similar in chondrite matrices and IDPs.

Taken together, these estimates suggest that at the time of accretion the CI chondrite and IDP parent bodies were composed of $\sim 5\text{--}10\%$ of pristine interstellar material. The matrices of all chondrites included roughly similar interstellar fractions. Whether this interstellar material avoided the thermal processing experienced by most dust because it came from far out in the disk or was accreted by the disk after the FU Orionis outbursts ceased to be important remains to be established.

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APPENDIX

Table A1. Summary of the results of all published presolar grain searches of IDPs conducted with NanoSIMS instruments. The averages were calculated by weighting the reported presolar abundances by the total areas analyzed.

Study	Subset	Nos. IDPs	Area (μm^2)	Nos. grains	Silicates (ppm)	Oxides (ppm)	SiC (ppm)
Messenger et al. (2003)	All	6	308	6	5519		
Messenger et al. (2005)	All	1	–	1	n.d.		
Floss et al. (2006)	All	26	10432	10	121		
	Primitive	15	3339	10	377		
Stadermann et al. (2006)	All	1	320	2		600	60
Busemann et al. (2009)	All ¹	4	2262	11	454		
	G4	1	36	7	15083		
	E1	1	138	4	3514		
Messenger et al. (2009)	All	2	181	5	2197		
	#13	1	152	3	1300		
	#4	1	29	2	6900		
Floss et al. (2010)	All	3	495	5	233		
	Adric ²	1	165	5	700		
	Perse + Quas. ¹	2	330	0			
Davidson et al. (2012)	All	8	565	13	2234	552	53
	GS ¹	1	173	1	919		
	Other	7	392	12	2814	796	77
Average	All	44	14563	52	396	35	3

¹Studies of particles that may have come from comet Grigg-Skellerup.

²Study of particle that may have come from comet Temple-Tuttle.

n.d. = not determined.

Table A2. Summary of presolar grain searches in the matrices of the least aqueously altered and thermally metamorphosed chondrites. Only studies conducted with NanoSIMS instruments were included to ease the comparison between studies of chondrites and IDPs

Meteorite	Study	Area (μm^2)	Nos. grains	Silicate (ppm)	Oxide (ppm)	SiC (ppm)
C ungrouped						
MIL 07687	Davidson et al. (2014)	31,000		n.d.	n.d.	57
MIL 07687	Floss and Brearley (2014)	33,900	74	110 \pm 15 ¹		85 \pm 20
Adelaide	Floss and Stadermann (2012)	34,600	69	53 \pm 8	17 \pm 6	8 \pm 3
Adelaide	Davidson et al. (2010)	2,600	8	180	73	
Acfer 094	Vollmer et al. (2009)	70,500	158	163 \pm 14	26 \pm 6	
Acfer 094	Nguyen et al. (2007)	~16,000	19	90 \pm 35 ²	55 \pm 20 ²	
Ningqiang	Zhao et al. (2011)	19,800	16	127	3	
CO						
DOM 08006	Nittler et al. (2013)	22,500	90	~210	~30	40 \pm 13
DOM 08006	Haenecour et al. (2014b)	14,900	51	216 \pm 30 ^{1,3}		\leq 31 \pm 13
ALHA 77307	Nguyen et al. (2010)	42,880	115	177	11	
ALHA 77307	Bose et al. (2012)	17,100	113	119 \pm 14	22 \pm 9	110 \pm 27
LAP 031117	Haenecour et al. (2014a)	30,650	112	204 \pm 24 ^{1,3}		60
CR						
QUE 99177	Nguyen et al. (2010)	21,170	52	152	8	49 \pm 14
QUE 99177	Floss and Stadermann (2009)	8,500	33	220 \pm 40	<5	n.d.
MET 00426	Floss and Stadermann (2009)	9,200	28	160 \pm 30	4	n.d.
MIL 090657	Davidson et al. (2015)	12,000	33	112 \pm 25 ¹		40 \pm 20

Table A2. *Continued.* Summary of presolar grain searches in the matrices of the least aqueously altered and thermally metamorphosed chondrites. Only studies conducted with NanoSIMS instruments were included to ease the comparison between studies of chondrites and IDPs.

Meteorite	Study	Area (μm^2)	Nos. grains	Silicate (ppm)	Oxide (ppm)	SiC (ppm)
NWA 852	Leitner et al. (2012)	20,700	39	78	38	160
GRV 021710	Zhao et al. (2013)	46,900	112	165 ± 29	9 ± 6	n.d.
GRV 021710	Zhao et al. (2013)	10,700	28	n.d.	n.d.	189 ± 34
L/LL						
MET 00526	Floss and Haenecour (2016)	19,200	39	275 ± 50^1		65 ± 25

¹Did not differentiate between silicates and oxides.

²These data are uncorrected for detection efficiency so that they can be compared to all other studies since they did not make such corrections.

³This is the average composition for matrix. Haenecour et al. (2015) reported large spatial variations in O-anomalous grain abundances that appear to reflect varying degrees of aqueous alteration, with the highest abundance in LAP 031117 matrix of 291 ± 54 ppm.

n.d. = not determined.

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