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# INTERSTELLAR DUST AS THE SOURCE OF ORGANIC MOLECULES IN COMET HALLEY

J. M. Greenberg

*Laboratory of Astrophysics, University of Leiden, P.O. Box 9504, Niels Bohrweg 2, 2333 CA Leiden, The Netherlands*

## ABSTRACT

Interstellar dust is described as consisting predominantly (by mass) of tenth micron (mean size) silicate core-organic refractory mantle particles which have evolved over galactic time scales of the order of 5 billion years. These particles were incorporated into comets and asteroids in the presolar nebula 4.5 billion years ago. The fragmentation of those primitive bodies gives rise to solar system debris which shows up as comet dust, zodiacal light, IDP's and meteorites. The chemical and morphological structure of comet dust is derived here as fluffy aggregates of interstellar dust. The chemical and morphological structure of the chondritic porous IDP's are then derived from comet dust which has evolved in the solar system. Zodiacal light particles are interpreted as various stages between comet dust and IDP's. Meteorites appear to be a side branch in the evolution from interstellar to solar system particles.

## INTRODUCTION

How far removed in chemistry and morphology are the small particles in the solar system from their progenitors - the interstellar dust? Can we expect to find really close similarities? What we see at the present time in the form of comet dust, zodiacal light, meteors, IDP's, and meteorites must have originated in larger bodies which formed 4.5 billion years ago. Not only could these parent bodies have undergone significant metamorphosis in the aggregation stage but we might expect to find further changes to have occurred both within the parent bodies as well as from chemical and physical changes in the solar system following fragmentation. It is therefore at first glance almost inconceivable that close resemblances with solar system particles are traceable to the original interstellar dust. In some cases - as with comet dust and IDP's - it appears remarkably close.

## SIZE, SHAPE AND COMPOSITION OF INTERSTELLAR DUST

Recent studies of the observations of so-called diffuse cloud dust (dust not in molecular clouds) in the ultraviolet have revealed the fact that there are three populations of dust /1/. There are elongated "large" grains of ~ 0.12 μm in mean radius which provide the major blocking of starlight in the visual. There are also very small carbonaceous particles of $\lesssim$ 0.01 μm in radius which produce a strong absorption feature at about 220 nm /2/. In addition there is an independent population of ~ 0.01 μm silicate type particles. Large carbon bearing molecules (or very small particles) like PAH's /3/ consume a small ($\lesssim$ 5%) of the carbon.

The evolutionary picture of dust which is emerging is a cyclic one in which the particles, before being destroyed or going into solar system bodies, find themselves alternately over many cycles in diffuse clouds and in molecular clouds /4,5,6/. A small silicate core captured within a molecular cloud accretes various ices and gradually builds up an inner mantle of organic refractory material which has been produced by photoprocessing of the volatile ices. Since the silicates which are formed in cool evolved stars are not crystalline, the elongation required for polarization in the 10 μm (Si-O stretch) band must be due to connected more-or-less spherical silicate beads. Representation of the core-mantle particles by concentric cylinders has been a mathematical convenience. The organic refractory mantles are subjected to the highest photoprocessing rates in the diffuse cloud phase - higher by factors of 10,000 or so than in the molecular cloud. Because of the cyclic evolution the organic refractory mantle on a grain is not a homogeneous substance but rather layered like the rings of a tree trunk in which the innermost layers have been the most irradiated and the outermost layer in the most recent molecular cloud phase is first generation organic refractory which is surrounded finally by lightly photoprocessed ices of which $H_2O$ is the dominant component. Since further photoprocessing of organics leads to a greater and greater depletion of O, N, and H, the innermost layers are the most "carbonized" and the most non-volatile.

A schematic representation of grains in the various regions of space is shown in Fig. 1. Theoretical calculations of core-mantle particles have been shown to match the observed

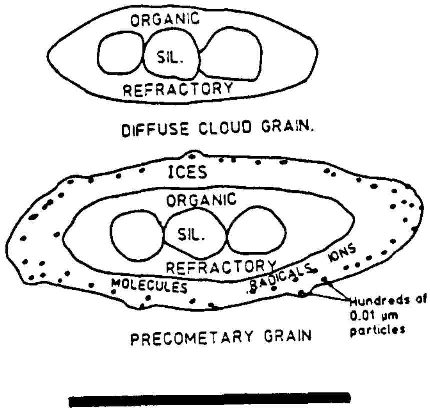


Fig. 1. Interstellar grains as core-mantle structures. The solid bar is 1 μm.

extinction and polarization as well as the albedo of interstellar dust /8/. In the final stage of cloud condensation we may expect that all remaining (condensable) molecules will have accreted onto the dust. In addition, the very small (≲ 0.01 μm) particles will be collected and trapped within the outer volatile icy mantle.

Our focus here will be on establishing a relationship between the chemical and morphological structure of presolar interstellar dust and comet dust, interplanetary dust, and meteorites.

## DUST AGGREGATION AND MORPHOLOGY

In our solar system all of the planets and satellites have incorporated into their bodies at least the most refractory components of the interstellar dust which existed in the pre-solar nebula. Comets, appear likely to have preserved their original composition best including their volatiles not only because the volatile molecule $S_2$ may be traced back to the photochemical evolution of the interstellar dust /9/ but also because of the observed $CH_4/H_2O$ ratio /10/.

As a first approximation, therefore, we consider a comet nucleus as if its chemical composition and morphological structure are directly related to interstellar dust. Table 1 shows the relative fractions of the various chemical constituents which have been obtained by an extrapolation from the molecular cloud dust phase /11,12/.

TABLE 1 Suggested mass distribution of the principal chemical constituents of a cometesimal based on the dust model. Parentheses refer to very small particle components ($a \lesssim 0.01$ µm). /7/.

| Component | Mass Fraction |
|---|---|
| Silicates | 0.14+(0.06) |
| Carbon (carbonaceous) | (0.06) |
| Nonvolatile Complex Organic Refractory | 0.19 |
| $H_2O$ | 0.37 |
| CO | 0.05 |
| Other Molecules + Radicals ($H_2CO$, $NH_3$, $OCN^-$, $CO_2$ HCO, $S_2$, $CH_3OH$...) | 0.13 |

In forming the nucleus we assume that first clumps of grains form, and then clumps of clumps, and so on, until finally we reach the size of the comet nucleus. If we should start with the interstellar dust tightly packed and then remove all the volatiles (along with the trapped super small particles) the resulting mean density of the remaining core-organic refractory grains skeleton is about 0.5 g $cm^{-3}$ /13/. It is however observed that meteors (which are what is left after the original cometary volatiles have evaporated) have a characteristic density much lower than this, often being even less than 0.1 g $cm^{-3}$. This leads to a packing factor of 0.2; i.e., a comet is about 80% empty space! A model of such an open aggregate of 100 typical precometary grains is shown in Fig. 2a.

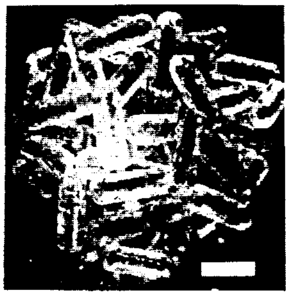

Fig. 2a: A piece of a fluffy comet: Model of an aggregate of 100 average interstellar dust particles each of which consists of a silicate core, an organic refractory inner mantle and an outer mantle of predominantly water ice in which are embedded the numerous very small (< 0.01 µm) particles responsible for the interstellar 216 nm absorption and the far ultraviolet extinction (See Fig. 1). Each particle as represented corresponds to an interstellar grain ½ µm thick and about 1½ µm long. The mean mantle thickness corresponds in reality to a size distribution of thicknesses starting from zero. The packing factor of the particles is about 0.2 (80% empty space) and leads to a mean mass density of 0.28 gm $cm^{-3}$ and an aggregate diameter of 5 µm.

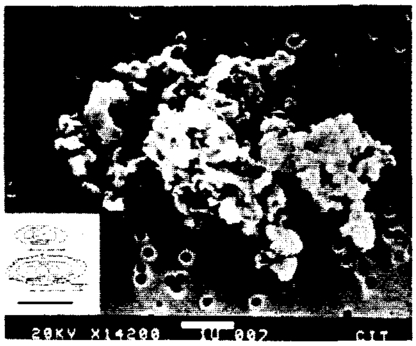


Fig. 2b: A highly porous chondritic IDP /35/. Note that the bird's nest particle (Fig. 2a), the IDP (Fig. 2b) and the average interstellar core-mantle particle (Fig. 2b insert) are equally scaled to 1 µm.

## CHEMICAL COMPOSITION OF COMET HALLEY

Of course, $H_2O$, was the most abundant molecule deduced in the coma of comet Halley. The next most abundant species is CO. For example Krankowsky et al. /14/ found a ratio $Q_{CO}/Q_{H_2O} = 0.03$ and infrared data gave $Q_{CO_2}/Q_{H_2O} = 10^{-2}$ while IUE observations /15/ gave $Q_{CO}/Q_{H_2O} = 0.1 - 0.2$. These values are more or less within the ranges suggested by the volatile composition of interstellar dust /16,17,18/. There are two possible sources of CO. One of these is, of course, as part of the ice which evaporates from the grains. Another is the photodissociation of the more volatile molecules of the organic refractory component. The existence of carboxylic acid groups in laboratory first generation organic residues and, by inference, in the outer parts of the organic dust mantles, makes such a source highly plausible. The existence of an extended CO source in Halley /19/ associated with the dust provides support for the fact that a large fraction of CO comes off as the dust fragments and releases small grains from which the not-so-refractory organics evaporate and are photodissociated (see section d for other gas components from dust). There is no definite evidence for the presence of $NH_3$ in the ion mass spectra /20/ and there may even be a lack of nitrogen in the coma gas. This is yet to be definitely confirmed but one possible reason could be that nitrogen is strongly bound in the organic refractories and is rather part of the dust than directly in volatile forms like $NH_3$ and $N_2$. Although $NH_3$ had earlier been suggested to be a substantial component of interstellar dust the observational evidence /21/, as well as theoretical arguments lead to generally rather small amounts of $NH_3$ in grain mantles and possible more $N_2$ /22/.

It was noted by Balsiger et al. /20/ that the C/O ratio is about half of the cosmic abundance ratio. This had earlier been called the missing carbon mystery by Delsemme /23/ and had been attributed to the "hiding" of a large fraction of the carbon in the organic refractory component /12/. The dust mass spectra where the carbon to oxygen ratio is much higher than cosmic abundance confirm this prediction /24/.

The dust impact mass analyzers on Vega 1/2 (PUMA) and on Giotto (PIA) showed a predominance of the light elements H, C, O, N (organics) relative to the heavier elements Si, Mg, Fe (rockies) in the dust /25,26/.

Kissel and Krueger /24/ have derived a molecular analysis of the comet dust and in particular its organic component. Masses between $2 \times 10^{-15}$ and $10^{-11}$g were measured with the masses of most of the particles estimated to be in the range $10^{-12}$-$10^{-13}$ g with "systematic error within an order of magnitude". Their typical total relative atomic abundances in their molecules (of the organic refractory) show a significant lack of oxygen just as is predicted by the interstellar dust model. A four-fold enhancement of carbon was predicted relative to oxygen /11,12/. The ratio of organics to silicate mass deduced by K+K is $m_{OR}/m_{sil} = 1:2$ which, not surprisingly, is less than that in the interstellar dust because of the expected evaporation of the less refractory organics at solar system temperatures.

Table 2 shows the distribution of atomic constituents in the various precometary interstellar dust components based on the values in Table 1. Normalizing to Si = 100 comparison may be made with the comet dust data deduced by Kissel and Krueger. This comparison is shown in Table 3. The only major discrepancy which can not be readily explained is the underabundance of N. For the rest it is seen how similar the atomic composition of organic refractory mantles of interstellar grains are to the comet dust organics.

TABLE 2. Atomic constieunts of various cometary components as fraction of the cosmic abundance, based on the dust model. The hydrogen are estimated as follows: 1 for each carbon in the organic refractory, 2 for each oxygen in $H_2O$, 1 for each carbon in "Other", 1 for each nitrogen in "Other" and 1 for each oxygen in "Other". Superscript a indicates that the figure is based on graphite (which we know is not valid); b indicates that the figures are particularly uncertain. This is because nitrogen is relatively more abundant in the organic refractory than oxygen, so the fractional (by cosmic abundance) nitrogen value could be significantly higher and that of oxygen significantly lower. These changes affect "Other" accordingly.

| Element | Silicate | Organic refractory | Small carbonaceous | $H_2O$ | CO | Other |
|---|---|---|---|---|---|---|
| H | - | $1.7 \times 10^{-4}$ | - | $4.7 \times 10^{-4}$ | - | $4.4 \times 10^{-4}$ |
| C | - | 0.45 | ≤0.27[a] | - | 0.10 | 0.17 |
| N | - | 0.25[b] | - | - | - | 0.75 |
| O | 0.09 | 0.13[b] | - | 0.65 | 0.05 | 0.80 |
| Mg | 1.0 | - | - | - | - | - |
| Si | 1.0 | - | - | - | - | - |
| Fe | 1.0 | - | - | - | - | - |

TABLE 3. Abundances in comet dust relative to Si (= 100) (Kissel and Krueger Nature 326, 755 (1987) compared with presolar interstellar dust (ISD).

| Elemental abundance Organic mantles | (K+K) | (ISD) | Elemental abundance | Choudritic (K+K) | Silicate cores (ISD) |
|---|---|---|---|---|---|
| H | 400 | 531 | C | 100 | |
| C | 500 | 520 | O | 300 | 200 |
| N | 20 | 97[b] | Na | 2 | |
| O | 100 | 275[b] | Mg | 70 | 94 |
| S | 10 | ?[a] | Al | 5 | |
| | Water (K+K) | "Ice" ISO | Si | 100 | 100 |
| H | 300 | (2840)f[c] | S | 40 | |
| O | 150 | (1630)f | Ca | 4 | |
| S | | (<87)[a]f | Fe | 70 | 81 |

a The cosmic abundance value of S used is 87. The interstellar dust model presumes a large fraction of this to be in the volatiles (ices).

b "First" generation organic refractory. Subsequent UV radiation reduces O but N should not be so reduced, i.e. N/O should be higher than Cosmic Abundance in organic refractory which has been further photoprocessed in diffuse clouds.

c f is the fraction of all volatiles remaining at the time of impact. A value f = 0.1 does not seem unreasonable.

## COMET DUST

The 3.4 µm and 10 µm excess emission in comet dust provide evidence not only for the basic chemical ingredients - as given in the mass spectra - but also for the morphological structure /27/. It turns out that pure silicates no matter how small do not achieve high enough temperatures to produce the observed 10 µm emission. At, for example 1.11 AU the required temperatures needed to keep the total mass of the emitting particles at all reasonable is T > 430 K. Absorbing organic refractory mantles - such as those on interstellar silicate cores - are absolutely required to raise the compound grain temperatures high enough to make the 10 μm peak observable. Furthermore, the T > 430 K temperature constraint leads to a most probable silicate core radius ~ 0.05 μm and a mantle thickness ≳ 0.02 μm. i.e. an organic to silicate mass ratio $m_{OR}/m_{Sil} \approx 0.9$ which, within the uncertainties, is like that deduced from comet dust mass spectra. If only such small particles ($m \lesssim 10^{-13}$g) could produce the 10 μm (and 3.4 μm) emissions their fluxes would have been more than 10,000 times higher than observed. It is, only by considering them to be in fluffy aggregates that the integrated fluxes come into reasonable resemblance to the particle impact detector data /28/ - although still by a factor of about 25 too high for masses $\lesssim 10^{-9}$g.

## ZODIACAL LIGHT DUST

Interplanetary dust has classically been observed via its scattering of sunlight - the zodiacal light. The addition of infrared observations has revealed some significant physical distinctions between particles as a function of distance from the sun. Those which are within 1 AU scatter visible light much more effectively than those which are beyond 1 AU. At the same time, those which are farther out are more effective emitters of infrared radiation. This implies a difference in kind as well as number with increasing solar distance /29/. The most obvious explanation of this phenomena is that the radial decrease of the albedo of the zodiacal light particles is produced by a decrease in material density, just as the albedo of cometary dust is decreased because of its fluffiness. The interplanetary particle probe results of Pioneer 10/11 were also interpreted in terms of a radial decrease of particle density /30/.

It has been suggested that the zodiacal light is predominantly produced by particles which started out as comet dust /31/. The alternative point of view is that interplanetary particles result from asteroidal collisions /32/. Probably something in between may be true although, if some asteroids are just inert comets, the distinction may be academic. That asteroids play only a minor role as a dust source /33/ appeared to be confirmed by the Pioneer 10/11 data which did not show any dust increase in the asteroidal belt /34/. With the assumption that most interplanetary particles start out as fluffy low albedo comet dust particles (like that in Fig. 2a), Mukai and Fechtig /35/ proposed a mechanism by which solar heating would lead to a gradual compaction of the initially fluffy dust by evaporation of the volatiles in what they called "Greenberg particles" leading to more compact and higher visual scattering particles like the "Brownlee particles" (Fig. 2b).

## COLLECTED INTERPLANETARY DUST PARTICLES (IDP's)

Although the mean density of the chondritic porous IDP's collected in the stratosphere is low it is much higher than the initial cometary dust. But, as has been pointed out by Brownlee himself /36/. there is no evidence of a bird's nest structure in the IDP's (Fig. 2b). What we see in Fig. 2b is an aggregate of more or less spherical particles of about 0.1 μ diameter whose infrared signature is that of silicates. When the interstellar dust is scaled like the IDP we see how its silicate core segments - which are hidden in the bird's nest model (Fig. 2a) - are like the silicates in the IDP. But where are the organic refractory mantles in the IDP's? In the original (interstellar dust) comet nucleus material the ratio of O.R. mass to silicate mass is given as about 1.5:1 (Table 1). However, already in the comet dust, the loss of the more volatile O.R. molecules has led to a reduction of this ratio by about a factor of 3 to about 1:2 (K+K). While the organic mantles are not "seen" in the IDP electron micrographs they become immediately apparent with Raman spectroscopy /37/. It appears that every silicate particle is covered by some organic mantle. The fact that the mean silicate particle size is like that of the interstellar core pieces and each silicate or clump of silicates has an O.R. coating is certainly suggestive of the interstellar origin while the bird's nest morphological structure is lost because of the removal, during the passage from the comet to the earth, of a further part of the original comet dust O.R.

Additional indications for the cometary to interplanetary dust evolution may be seen in the lower density of meteors whose aphelion distances are beyond 5.4 AU as compared with those which spend more time closer to the sun /38/.

## METEORITES

How do meteorites and their parent asteroidal bodies fit into the cosmic dust connection? Since the formation region for the asteroids was certainly at a higher temperature than that for comets we do not expect the interstellar dust to be nearly as well preserved. Within the framework of the theory of Ruzmaikina and Maeva /39/ the temperature of the pre-solar nebula relevant to the asteroidal belt was 250-300 K which was sufficient to evaporate all the dust volatiles while preserving a fraction of the organics. One factor which appears to provide a basis for believing the connection lies in the preservation /36,40/ of the pre-solar isotopic abundances of the heavy noble gases Ar, Kr and Xe in the carbonaceous component. These elements are presumed to have been trapped in the interstellar organic refractory mantles and retained during asteroid formation. Thus, although meteorites may be identified with the same interstellar dust ancestors as comets, they are like cousins rather then siblings.

Based on the observations of the largely amorphous, carbonaceous coatings in the Allende (C3V) meteorite /41,42,43/ Huss /44/ has suggested that the matrices in the parent bodies of the C3V, C3O, and type 3 ordinary chondrites probably accreted from presolar dust that had lost the icy mantles. On the other hand he proposed that CI (C1) chondrites and the matrices of C2 chondrites probably accreted as bulk samples of presolar dust with <u>some</u> icy mantles intact - almost cometary. Parent body heating (<u>not</u> characteristic of comets) then caused the icy mantles to react with the fine grained dust to produce the hydrothermal mineral assemblages now observed. The icy mantles in comet dust <u>evaporate</u> rather than melt so that, although we should not be surprised by seeing <u>some</u> resemblance between CP IDP's and CI chondrites, the differences should also not be a surprise - there are no hydrated silicates in low density IDP's. If IDP's are remants of comet dust they should more resemble the chemical and physical composition of the latter in which the $H_2O$ evaporated rather than melted. There is a monotonic sequence of carbonaceous content from interstellar dust to comet dust to IDP's to meteorites.

In Fig. 3 we summarize the relationship between interstellar dust, interplanetary dust and meteors and meteorites as conceived of here.

## CONCLUDING REMARKS

We have to look to future space missions to recover comet material much more pristine than we can infer from flyby or even rendezvous missions. If the comet nucleus material can be retrieved from its depths and maintained intact cryogenically for laboratory studies, we may hope to study not only its atomic and molecular compositions but also its morphology. Microprobes are being developed /45/ which will make investigations possible of submicron structures. If it should turn out that the interstellar dust model is correct, individual grains whose mean lifetime before becoming part of a comet is about $5 \times 10^9$ yr will reveal cosmochemical evolution not only of the solar system but dating back a further 5 billion years before the earth's beginning - back to the earliest stages of the chemical evolution of the Milky Way. Dramatic differences in isotopic abundances could be expected on scales of microns. The next twenty to thirty years should be exciting ones indeed for studies of our origins.

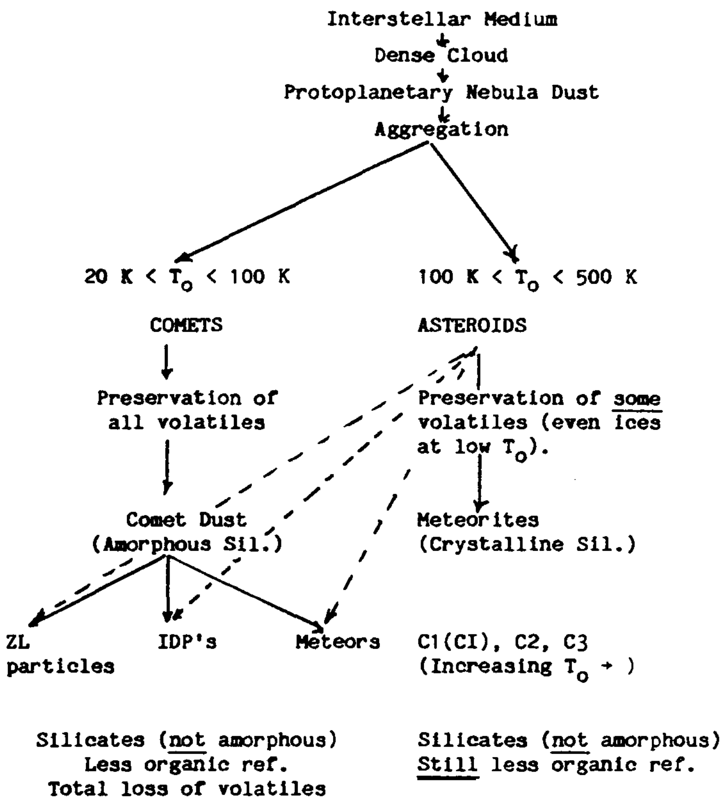


Fig. 3. Decrease of organics and increase of silicate crystallinity according to aggregation temperature, $T_O$, and thermal history.

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