Siliceous Fragments in Space Micro-dust: evidence for a New Class of Fossil

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Abstract

Collection of Interplanetary dust particles (IDPs) by stratospheric balloon-borne cryosamplers has shown in recent years has shown this to be a superior technique to collection by rockets and aircraft. IDPs in the Cardiff collection from 40km altitude have been studied via scanning electron microscopy and associated X-ray emissions. This paper reports the identification of IDPs containing carbonbaceous-siliceous fibres and whiskers, unlike the mineral silicate particles normally identified with astrophysical silicate. The integration in some cases into cometary agglomerate particles and coatins with salt and other components shows aggregation on the comet. Two examples of fibres have also been found in a carbonaceous chondrite (Tagish Lake) which is thought to have a cometary origin. The fibres and and whiskers may have formed in the comet environment, but their accumulation from the pre-solar dust cloud is not excluded. An astrophysical origin as high temperature condensate in stellar outflows does not however explain the fibre mophologies. We therefore suggest the fibres and whiskers are fragments of fossilized organisms, like some species of terrestrial diatoms.

Keywords: IDPs, stratospheric dust, cryosampling, diatoms, siliceous exoskeleton, extraterrestrial fossils.

1. Introduction

The IDPs of the Cardiff collection have been recovered from the upper stratosphere by balloon-borne cryosamplers flown from Hyderabad (Lal *et al.* 1996; Narlikar *et al.* 1998). Since the sampler drifts with the stratospheric wind, being suspended below the balloon, and the samples are in air accreted by being frozen within steel cylinders, the IDPs are hardly damaged (including fragile low-density cometary IDPs) and make up a representative sample from the stratospheric air (Wallis *et al.* 2002 and 2006; Miyake 2009). Because of care in cleaning and sterilization of the samplers and high cleanliness in filtering out particles from the gas, the collection is found to contain very little laboratory-derived contamination. Collection by balloon above Australia on carbon and metal films (less secure against contamination) up to similar heights (40-45km) show terrestrial particles to be virtually absent (Bigg 2011). Moreover, the various types of IDPs sized 1-30µm are quite distinguishable from spacecraft fragments and terrestrial contaminants - they include types missed by NASA's original collection of stratospheric IDPs (by aircraft at lower altitudes where sulphate particles are common, on oil-covered collectors with a lower degree of cleanliness).

Many particles are composites, including layer silicates presumed of asteroidal origin. but the majority are low density agglomerates and condensates, often high in carbon, characteristic of cometary origin, but including apparently pre-solar crystallites. Near-spherical mineralised shells have been identified (Miyake *et al.*, 2010, Wickramasinghe et al. 2010) which are similar to acritarchs extracted in the 1980s from the Orgueil meteorite (Rossingnol-Strick and Barghoom 1971; nowadays such cabonaceous meteorites are thought to come from comets). This study focuses on the siliceous 'fibres' (exoskeletons of tubular specimens – over 10µm long and down to 0.4µm diameter) and 'whiskers' (finer ones to ~0.1µm diameter) found in the collection, which were briefly reported earlier (Miyake *et al.*, 2009). These samples were obtained in 2001 at 40km altitude, extracted in Cardiff.

Sample preparation and analysis is descibed in the Appendix. The scanning electron microscopy (SEM) reveals the IDPs are sparsely distributed on the air filter, some with adjacent fragments of similar type, implying fragmentation on impacting the filter (Miyake 2009; Miyake *et al.*, 2010). The impact of larger (~10µm) particles was sufficient to cause observed damage to the acetate surface, confirming they are not laboratory contamination. The SEM with EDX and transmission electron microscopy (Wallis *et al.*, 2002 and 2006; Miyake, 2009; Miyake *et al.*, 2010; Rauf *et al.*, 2010) show substantial carbonaceous components as well as mineral materials that could be meteoritic or processed in comets. Mineral materials composed of silicate are indicative of condensation during an energetic phase of the early sun or from pre-solar stellar outflows that fed the interstellar dust complex. The present images show siliceous fibres and whiskers are mostly embedded in or associated with either acritarchs or loose agglomerates and fit with neither origin.

There is a long history of claims of fossil life in meteorites; such extraordinary claims require an extraordinary level of evidence, so 20th century claims were dismissed when contamination could not be excluded. However, modern techniques particularly in the Mars meteorite work (McKay et al. 1996, 2009) including chemical analysis of the specimens and substrate, give us much stronger cases. As Michael Engel said (Engel 2011) in welcoming the latest work on cyanobacteria-like structures in the Orgeil meteorite (Hoover 2011), it is essential to continue to seek new criteria more robust than visual similarity to clarify the origin(s) of these remarkable structures. Our IDPs have an advantage in being newly collected (uncontaminated by their terrestrial landing environment and handling) and uncompact so less damaged components. There are numerous specimens compared with the few suitable carbonaceous chondritic meteorites, so reproducebility is no more an issue than in paleontology. Similar to that science, our interpretation uses the body of knowledge on IDPs and a concept of the comet environment where mineralisations and agglomeration occurs, as well as chemical composition and association with adjacent components.

2. Images of Fibres and Whiskers

SEM microscopy reveals IDPs sparsely distributed on the air filter, some showing adjacent particles of similar type – implying fragmentation on impacting the filter (Miyake 2009; Miyake *et al.*, 2010). The impact of larger (~10µm) particles was sufficient to cause observed damage to the acetate surface, confirming they are not laboratory contamination. Some IDPs are associated with siliceous 'fibres' and fine 'whiskers'. Figure 1A shows a single fibre 15µm long and 0.35µm diameter (probably a hollow cylinder - see caption; further examples shown in Miyake *et al.*, 2010). The finer whiskers are largely embedded in surface coats (Fig.1A). Figure 1B shows a complex bundle of fibres. The EDX spectrum (Fig.1D), taken at spot-S1 of figure 1C, shows Si and C with Na+Cl coating (crusting at the top).

The finer whiskers were found associated with cometary mineral particles of aqueous alteration and compaction. Figure 2A shows a 4µm long whisker, 1µm in diameter (arrow), firmly attached to the porous aggregates of carbonaceous IDP which resemble cometary condensates. Figure 2B shows a number of various rod-fibres and whiskers (arrows) deeply embedded in the carbonaceous magnetite composite of a 5x7µm loose agglomerate. Figure 2C shows an example of a 4x8µm IDP with an embedded submicron-width whisker. EDX data (Fig.2D) reveals that the IDP has magnetite-like composition (spot-S1) and the whisker has elevated Si and O (spot-S2).

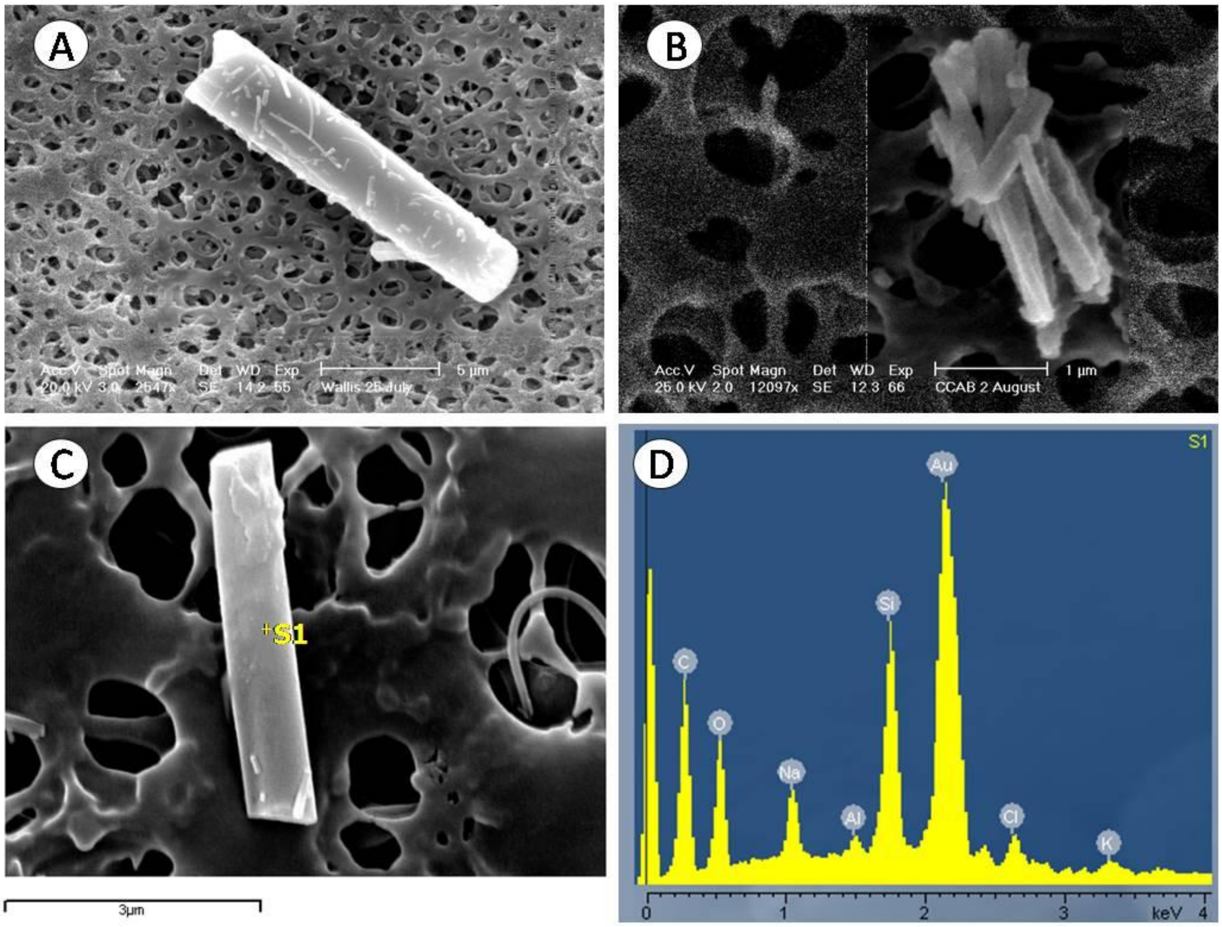

Figure 1:

[A] SEM image of a large siliceous fibre (15x3.5µm). Note the rounded lower end and fractured upper end, implying a hollow cylinder with top broken off. The short protrusion at the lower end would be a fibre fragment, adhering to the underside. The fine markings may be whiskers embedded in a salt coating as seen in other specimens.

[B] SEM image of a bundle of 0.3µm whiskers (~3µm long).

[C] SEM image of a single siliceous fibre (1x3.5µm). S1 is the location for the EDX spectrum (Fig.1D).

[D] EDX data of specimen (Fig.1C) shows the main Si peak with some Na, K and Cl (C appears strong but has an uncertain contribution from the acetate background). The Au peak comes from the gold conductive coating.

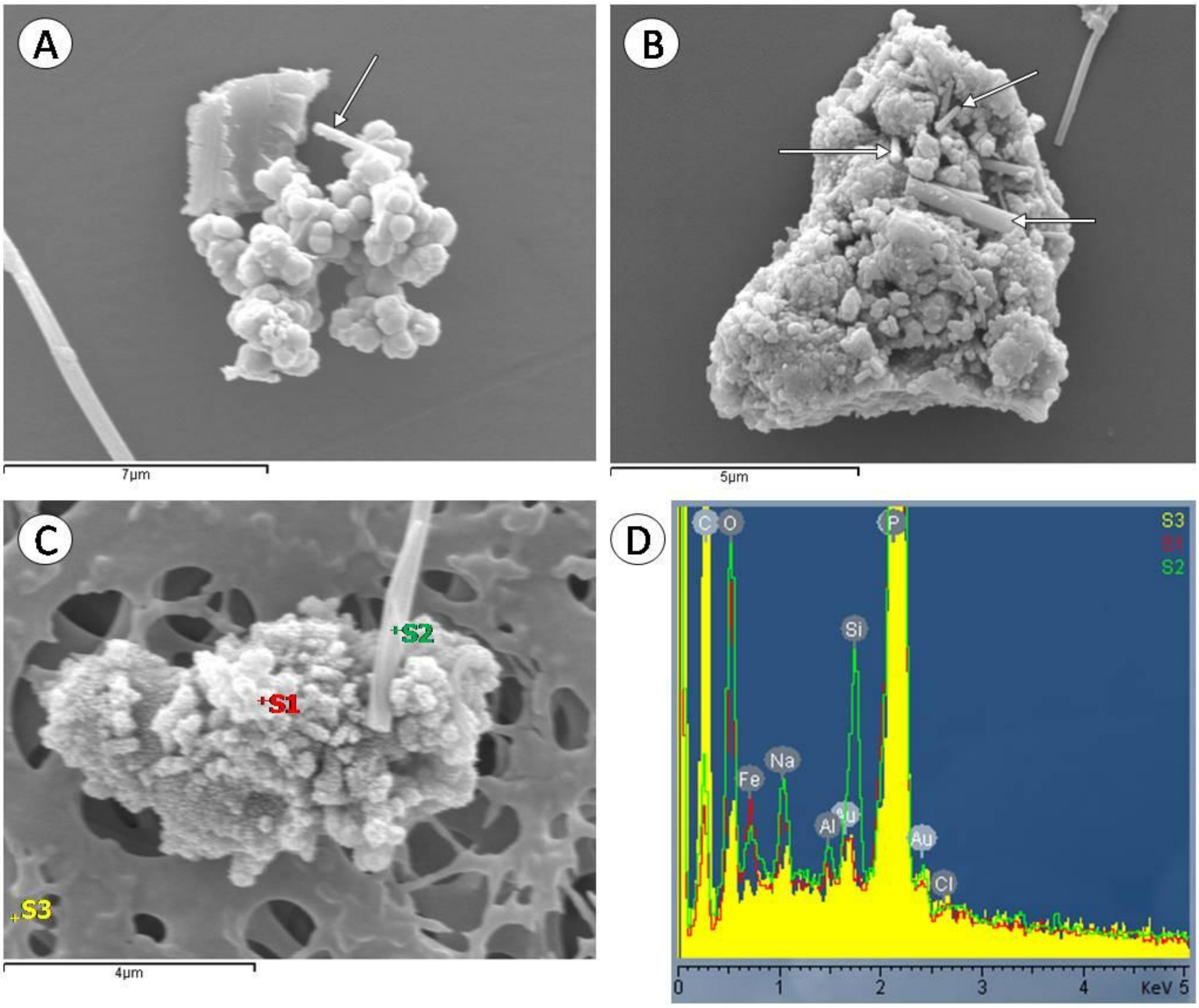

Figure 2:

[A] SEM image of a probable carbon-rich condensate of 0.5~2.0µm aggregate with higher mineral fraction in the particles (Cr ~10.5%, Fe ~22.5%) and a 4 x 1µm whisker (arrowed).

[B] SEM image of 5x7µm carbonaceous IDP with magnetite (C ~ 15%, Fe ~39%, O ~ 30%) with various sub-micron rods and whiskers embedded, as arrowed (as thin as 0.1µm).

[C] SEM image of 4x8µm IDP with an embedded submicron wide whisker. S1 is the location of the red coloured line in the EDX spectrum (Fig.2D). S2 for the green line and S3 for the background (yellow) spectrum.

[D] EDX data of specimen (Fig.2C) that shows the magnetite-like nature of the IDP (red line; Fe ~33%, O ~ 30%) and elevated Si and O in the whisker (green line - as the EDX spot size ~1mm, the Fe may be all in the underlying particle). The yellow profile is a background reading (spot-S3) of the acetate filter and its gold conductive coating (Au and Pd peaks).

Our study also found a 12µm long and 2~3µm diameter siliceous fibre associated with cometary ROCK particles (Fig.3A). EDX data (Fig.3B) shows the IDP is a Fe-Al-Si mineral (spot-S1) with a deposit of Na+K chlorides (spot-S2) that also covers the fibre (spot-S3).

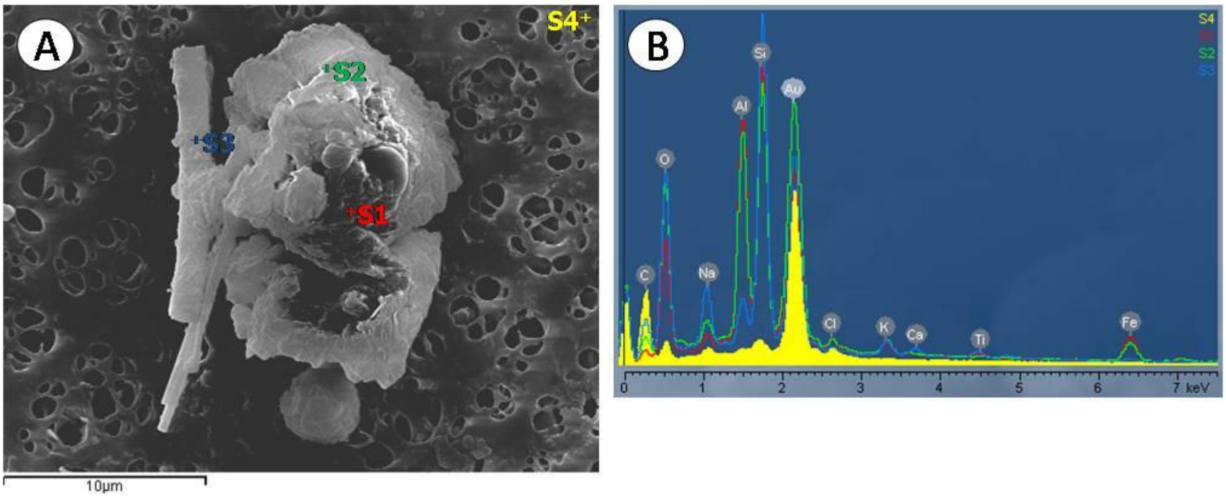

Figure 3:
[A] SEM image of 10x20µm IDP associated with 15µm length and 2µm diameter fibres. S1 is the location of the red line in the EDX spectrum (Fig.3D); S2 - green line, S3 - blue line and S4 - yellow background.
[B] EDX data of specimen (Fig.3A) shows the IDP to be a Fe-Al-Si mineral particle with alkali Na, K chlorides (red line); the silicate fibre is covered with similar minerals (blue line) proving that they came together in an aqueous environment. The yellow profile is the background reading (spot-S4) of the acetate filter and gold conductive coating (Au and Pd peaks).

Figure 4A shows an example of acritarch with whiskers ~4µm long and ~0.3µm diameter, indicated by arrows attached to its surface, representative of several given by Miyake *et al.* (2010). Acritarchs often crack in the SEM electron beam, with cracks widening over 5-10 minutes, so revealing as in this case that they are hollow shells.

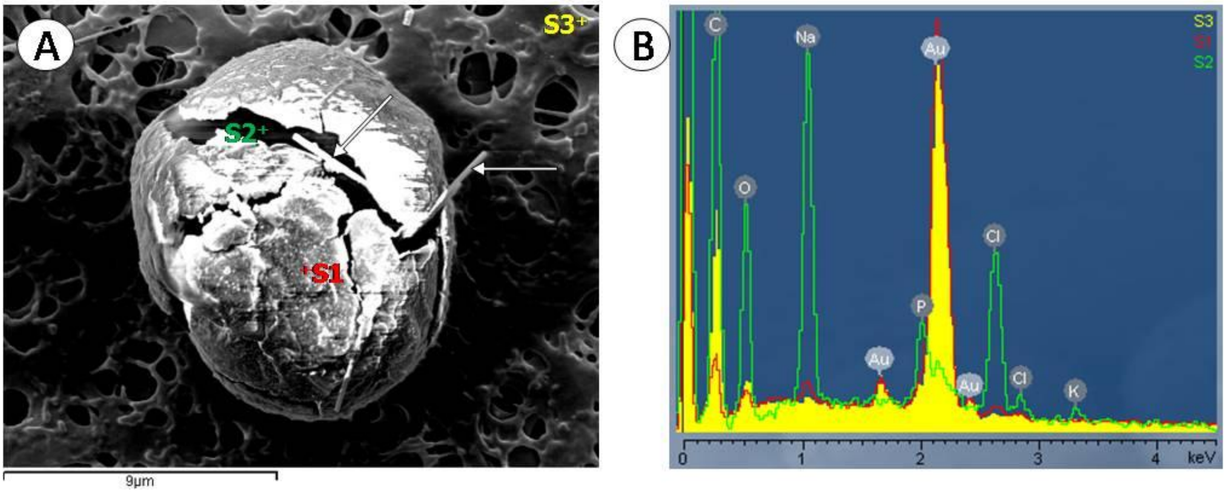

Figure 4:
[A] SEM image of a acritarch with attached whiskers (arrowed: Miyake *et al.*, 2010). The large crack reveals it is a hollow sphere. S1 is the location of the red line in the EDX spectrum (Fig.4D); S2 - green line and S3 - yellow background..
[B] EDX data of specimen (Fig.4A) showing it is high in carbon. S2 on the crack (S2; the absence of the Au peak shows the gold coating is cracked too) shows particularly high C (55%) and O (20%) in the interior, with Na (10%), Cl (10%), P (3%) and K (1.2%). S1 shows the shell's mineral coating contains O, Na and Cl (as well as shell C but no P.

Our example of detached siliceous fibres as well as ones attached to the IDP (Fig.1) shows some particles were loosely attached on the parent body. Other fibres and finer whiskers are firmly attached or even embedded within the aggregate IDPs. They were found associated with three types of space dust particles. The first association is with porous aggregates of carbonaceous IDP, which could be cometary condensates (Fig.2A). The second association (as Fig. 3) of siliceous fibre is with cometary ROCK particles. A third association is attached to acritarchs as shown in Fig. 4 (further examples are given in Miyake *et al.*, 2010).

The association with low density aggregates and salt crusts confirm their deriving from a comet. Whether the fibres and whiskers reached the comet after originating elsewhere has to be discussed. Note that Rossingnol-Strick and Barghoom (1971) found siliceous frustules in their Orgueil meteorite samples. Other fibres have been recently found in the Tagish Lake carbonaceous chondrite (Fig. 5).

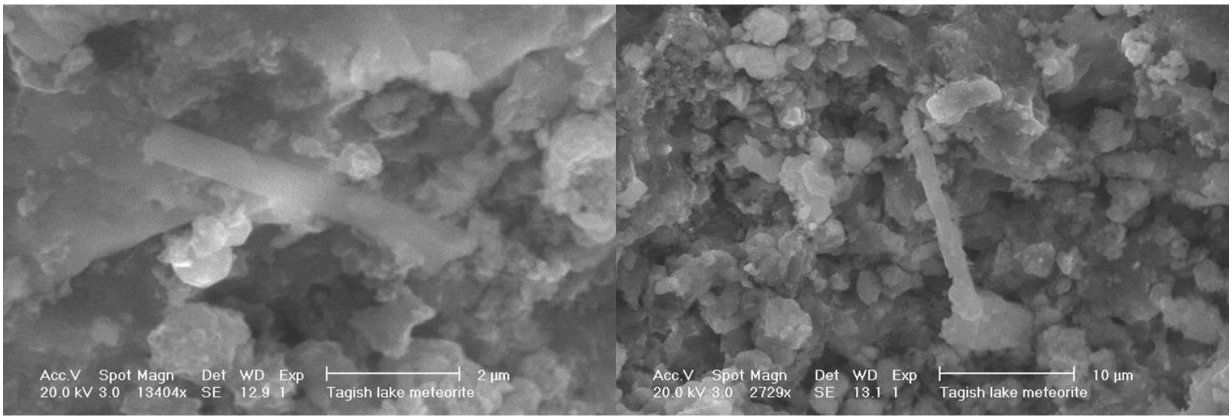

Figure 5: Diatom-like fibres in the Tagish Lake meteorite (SEM images courtesy of Kani Rauf, CCAB Cardiff). Scale bars on the left 2µm and on the right 10µm. The right-hand example shows surface deposits on the fibre similar to the surrounding mineral material and indicating integration with the substrate.

3. Potential Sources of Silicate Material

3.1. Astrophysical Silicates

SiO_2 is not a plausible condensate from stellar mass flows, supernova ejecta or protoplanetary nebula. Thermochemical calculations for an expanding solar outflow have shown the highest temperature condensates to be TiO_2, Al_2O_3, ZrO and CaO forming at temperatures of 1800-1700K, followed by Fe and Ni at 1500K. Magnesium silicates (eg. olivine) condense at temperatures below 1400K. The strong resonant peaks of crystalline silica (quartz) at 8.7 and 12.7µm are not seen in the interstellar medium or comets (Hoyle *et al.*, 1982).

3.2. Cometary Silicates

Dust collected from comet Wild-2 (Stardust) showed the presence of nanoscale high-temperature minerals coexisting with organics in the same particle, which indicated condensates in the outflow from an early active sun that underwent mixing with material of the solar nebula and volatiles condensing in the outer solar system (Brownlee *et al.*, 2006).

Comet Hale-Bopp gave spectral evidence of crystalline olivine mixed with much organic polymeric material (Fig.6 *Left*: Crovisier *et al.*, 1997). Fine phyllosilicate clays were discovered in comet Tempel-1's dust ejected by the Deep Impact collision (Lisse *et al.*, 2006; Fig.6 *Right*: Napier *et al.*, 2007). Our siliceous fibres and whiskers fit with neither the condensate nor the clay (aqueous-altered) origins.

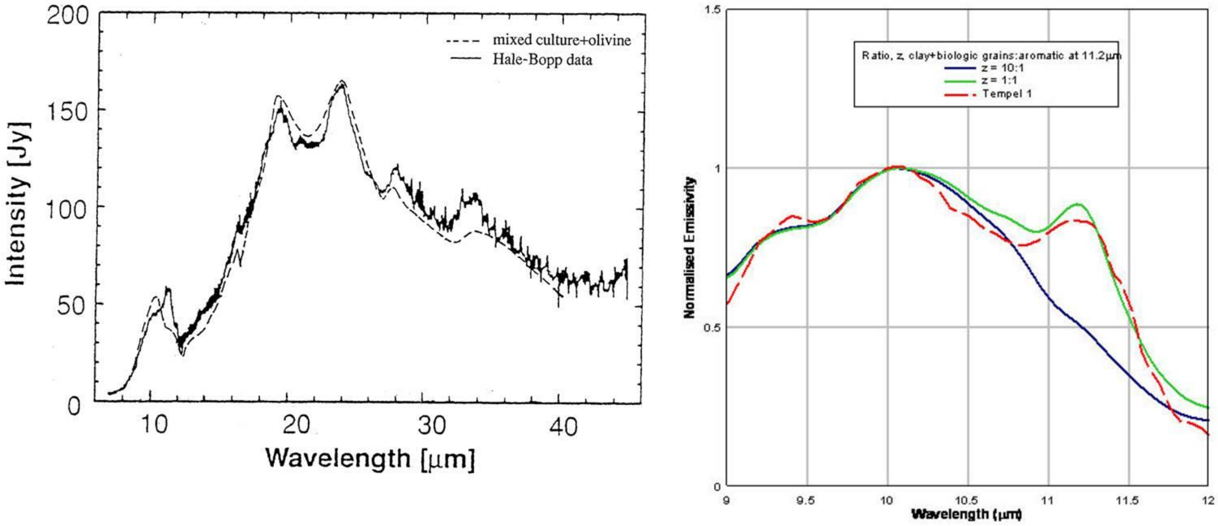

Figure 6:
Left – infra-red (IR) spectrum of comet Hale-Bopp shows a mixture including crystalline olivine (magnesium silicate) rather than SiO_2 fibres (data from Crovisier *et al.*, 1997, model fit from Wickramasingh). *Right* – part of an IR spectrum (0-12μm) of Comet Tempel-1 (red line) along with models with clay to biologic material weightings of 10:1 (blue line) or 1:1 (green line). Crystalline silicate (SiO_2) emissions have further structures not shown in the data. The further IR to >40μm also supports clays. The close fit of the green line supports the clay interpretation and indicating ~50% contribution in a clay-organic mix.

3.3. Siliceous Micro-organisms

EDX spectra show significant levels of carbon as would be present in siliceous cell walls of diatoms (Hoover *et al.*, 1986; Miyake *et al.*, 2010). Diatoms have siliceous exoskeletons, some of a basic cylindrical form, while marine species in particular, tend to possess numerous whisker-like extensions ('pili'-Fig.7A and 7B). Hoover *et al.* (1986) pointed out that diatoms could model the 'astrophysical silicate' indicated by infrared spectra of interstellar dust and toward the galactic centre source. Diatoms also exhibit an absorbance peak near 2200 Å, agreeing with the observed ultraviolet absorbance properties of interstellar grains.

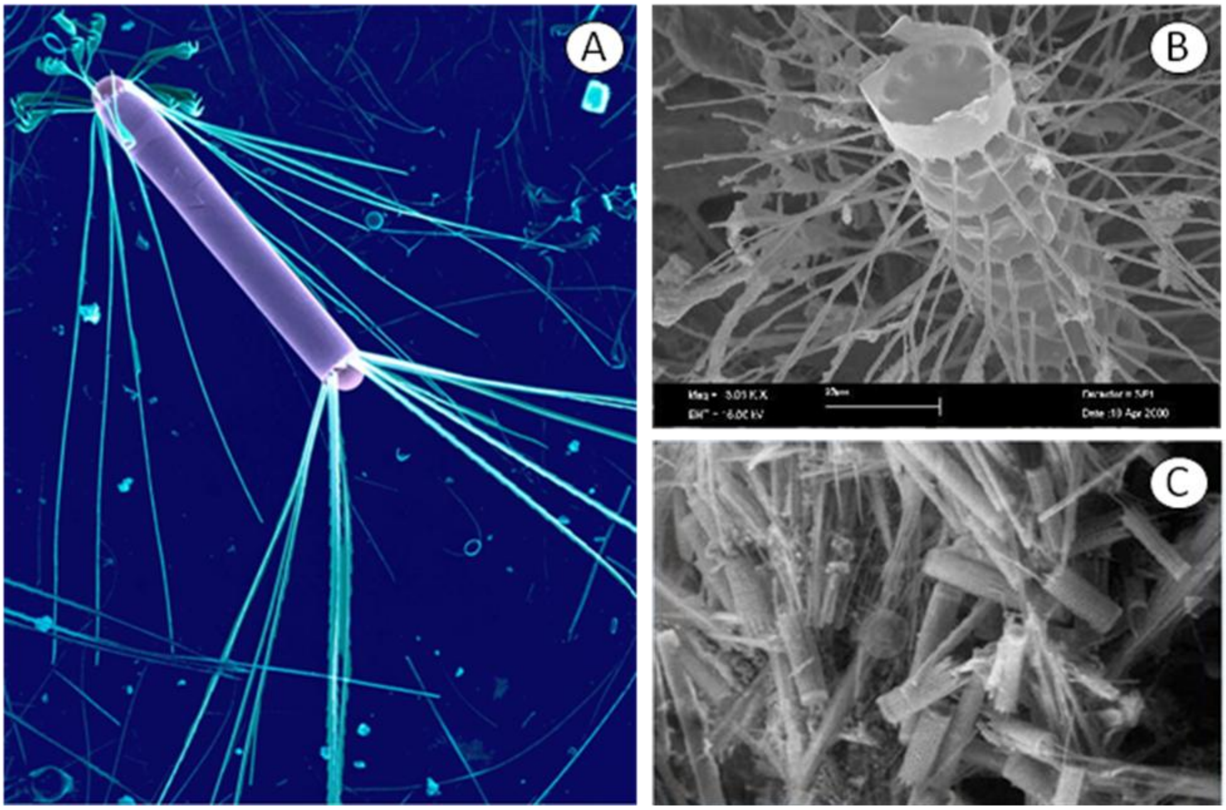

Figure 7:
[A] Antarctic marine diatom (reproduced by permission © Dee Breger, Drexel University); an SEM image (false colours) of a Corethron species of diatom, showing submicron width ‘whiskers’ and a siliceous exoskeleton (hollow cylindrical ‘fibre’).
[B] SEM image of a branched marine diatom collected at Rocky point, Mexico, scale bar - 25µm (© Boyer, Northern Arizona University).
[C] SEM image of lake sediments, showing a range of sizes of siliceous diatom fragments (© K. Wetmore of University of California, Berkeley Museum of Paleontology).

4. Discussion

Hoover *et al.* (1986) were the first to suggest siliceous diatoms in the space context, pointing out that diatoms could model the ‘silicate’ feature in infrared spectra of interstellar dust (Trapezium nebula) and toward the galactic centre source GCIRS 7. They hypothesised that cometary ice could provide a habitat for diatoms, analogous to arctic and antarctic ice, so these organisms would contribute to the cometary silicate feature. In this context, the siliceous fibres and whiskers of varying sizes could be naturally interpreted as remnants of fossil diatoms.

Diatoms can live at low light levels and are well adapted to polar ice ecosystems (Pituka *et al.*, 2007). Cryophilic ice diatoms respire at extremely low rates in the dark, ‘hibernating’ as intact organisms rather than entering some type of resting spore state. They revive readily after months of exposure to winter darkness, photosynthesizing rapidly when light and nutrients become available. Some species live in the total dark of deep-sea sediments and employ heterotrophic nutritional modes (Hoover, 2006). Primitive organisms other than diatoms do develop siliceous skeletons, though diatoms (and radiolarians) use a special mechanism for building their cell walls on a protein template layer (Hoover *et al.*, 1999). Identification with diatoms has to be tentative, pending the use of more powerful techniques on the samples.

It's becoming accepted that the aqueous alteration in crystalline state of many IDPs takes place within comets, including those picked up by *Stardust* from comet Wild-2 (Berger et al. 2011). Embedding of siliceous fibres in aggregate IDPs and attachment to acritarchs, with deposits of salt on the whole, would occur in the same cometary envrionment. Our siliceous fibres and whiskers could have been accreted into the comet from elsewhere, but we'd favour for simplicity an origin from diatoms on the comet itself. If alternatively, the diatoms populated the martian polar caps and/or solar system icy satellites, and were ejected to space by large impacts, these could be accreted by comets. In that case the IPDs would contain these ejecta as well as ones processed through comets. We can therefore conside 3 possible sources for the fibres: first martian ices, particularly the polar caps, for which ejection to space by large impacts could occur. Second, the sub-surface or near-surface cometary lakes (the site for aqueous alteration of minerals) likely to give embedded or firmly attached fibres, and third the sublimating surface of a comet where the fibres attach loosely to other particles as the icy volatiles sublimate away (Wickramasinghe *et al.*, 2010).

Identifying fossil matter in meteorites (McKay et al. 2009, Hoover 2011) is complex, as the biomatter would necessarily have been strongly mineralised in fossilisation processes. McKay et al talk of 'biomorphs' for which little more than the shape remains. IDPs are in principle contain less altered microfossils. The acritarchs are one new group of fossils, whose progeny requires study. The carbonaceous-siliceous material that we tentatively identify with diatoms is another type of fossil worthy of further study.

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APPENDIX Materials and Methods

Sample preparation

The recovery technique used at Cardiff (Harris *et al.*, 2002) extracted the particulate matter on 0.45μm micropore cellulose acetate filters, both directly via venting of the high pressure gas and via filtering water washings from the cylinders.

Some samples were transferred to silicon wafers (Miyake 2009), in order to accurately quantify the carbon from X-ray data. A portion of the filter was placed on top of a sterile 4 mm^2 silica wafer and gently tapped, which sufficed to transfer some loosely attached particles. The procedure being carried out swiftly in an aseptic laminar air flow cabinet throughout. To check on possible contamination, a control membrane was subjected to identical extraction procedures and SEM-analysis.

Scanning Electron Microscopy

In each case, the acetate filter (cut into 25 mm^2 pieces with a sterilised razor blade) or silica wafer was attached onto metal stubs with double-sided sticky carbon tape, then stabilised with a thin deposit of gold that creates a conducting surface, which prevents charging by the electron beam. These sample preparation procedures were again carried out in an aseptic laminar air flow cabinet. High resolution imaging was conducted with an environmental scanning electron microscope (Philips ESEM-XL30), operated at 20 keV accelerating voltage and 40μm condenser aperture.

X-ray nanoprobe Analysis

The samples as prepared above were analysed using an X-ray detector (EDX, EM-400 Detecting Unit, UK) fitted in the scanning electron microscope. The following operating conditions were maintained constantly throughout the analyses: electron accelerating voltage (10 keV), condenser aperture (50μm), and analysis time (200 cps). Both qualitative and quantitative analyses of the spectra were performed using the EDAX Genesis software.