INVESTIGATING RAMAN VARIATION ACROSS LARGE CLUSTER INTERPLANETARY DUST PARTICLES.

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Introduction: Interplanetary dust particles (IDPs) collected in the stratosphere are dust from comets and asteroids that have arrived in the Earth's stratosphere via Poynting-Robertson effect after being ejected from their parent body. Chondritic porous IDPs display a range of features indicative of a very primitive nature and are generally believed to originate from comets. While our knowledge is somewhat limited about the internal and near-surface processes affecting non-ice materials within a cometary body, those processes that have occurred will have been different, and probably less pervasive compared to those occurring on the asteroidal meteorite parent bodies.

The abundant organic material present in primitive IDPs may be the result of formation processes that occurred across a large volume of the protoplanetary disk. However, the study of organic material in particles only a few to a few tens of microns across is challenging. Laser Raman microscopy offers a rapid and potentially non-destructive approach for determining some important general characteristics of the organic matter in IDPs. While a number of studies have been conducted to date, the number of IDPs analysed remains relatively small (a few dozen) – particularly when the number of potential parent bodies is considered or the huge volume of the protoplanetary disk that may have contributed to the formation of comets.

Nine IDPs were selected at the Cosmic Dust Laboratory at Johnson Space Center from five large cluster particles on collectors L2005 and L2006. Cluster particles represent large IDPs that broke into smaller pieces on impact with the collector. These particles can be >100 μm across but the individual particles within them are typically 5-15 μm in size. This offers the opportunity to investigate the variability of the Raman signature of the organic matter on a number of scales.

This study is the first part of a larger, on-going project integrating the Raman, mineralogy (ASEM) and isotopic signatures (NanoSIMS) of the particles.

Method: Raman measurements were performed at the Open University with a 473nm excitation laser. A laser power of <70 μW at the sample surface was maintained throughout all measurements so that thermal damage of the sample was avoided [1]. A x50 long working distance objective lens was used giving a spot size of ~2 μm. Analyses were obtained as maps of point spectra (40s integration) across IDPs on glass slides with a step size of 1 μm. The optimum focus for each point was determined using the Raman intensity of the G-band. First order carbon D (disordered) and G (graphite) bands were fitted in the range 850 to 2300 cm^{-1} with a polynomial baseline and Gaussian-Lorentzian profile model. The peak positions (ωD, ωG), peak widths (ΓD, ΓG) and peak intensity (ID/IG) were obtained for each IDP by averaging the peak parameters from all spectra obtained from each particle.

Results: Previous Raman analyses of IDPs and insoluble organic matter (IOM) from meteorites have generally been performed with green lasers (514nm and 532nm). However, it has been shown that the position of the D band is influenced by excitation wavelength [3], and therefore this study was extended to observe the previously unreported effect at 473nm. IOM extracted from Allende, Cold Bokkeveld, Murchison and Leoville meteorites (samples as in [2]) together with 3 of the IDPs were mapped using both 514nm (green) and 473nm (blue) excitation lasers. Both datasets revealed a reproducible shift in ωD with the values from the 514nm laser 10-13 cm^{-1} lower than those from the 473nm laser (Fig.1). There was no significant variation in any other parameters observed for the IOM samples other than a modest shift in ωG (0.2 cm^{-1} Allende; 1.7 cm^{-1} Cold Bokkeveld). However, the results from the 3 IDPs analysed with both lasers generated a ωG ~4 cm^{-1} lower with the 514nm laser than the 473nm laser. The results also reveal narrower peak widths of 4 and 8 cm^{-1} for ωG and ωD respectively when using the longer wavelength laser (Fig.1), consistent with previous work [3], although the scatter in peak width data was considerably greater than the observed mean differences.

In order to compare these data to studies using 523nm lasers, 20 cm^{-1} has been subtracted from ωD values for IDPs measured in this study (20 cm^{-1} results from the 10-13 cm^{-1} difference between 473nm and 514nm excitation plus the 5-8 cm^{-1} downshift demonstrated by [3] for 514nm to 523nm excitation). 5 cm^{-1} was also subtracted from ωG values for the IDPs. No correction was applied to ΓD or ΓG as the uncertainty was much greater than the measured differences.

Discussion: The large ΓD and ΓG, high ωD, and low ωG for all the IDPs in this study indicate that they are all primitive. The samples plot near to the CI, CM and CR field (not resolved from other IOM on Fig.2). The dispersion in ωG with excitation laser is related to the degree of disordering of the carbon [3], and therefore the greater ωG dispersion observed for the IDPs suggests that the organic material in the IDPs may in fact be considerably more disordered than in the IOM. ID/IG varies from 0.9-1.3, which again indicates the

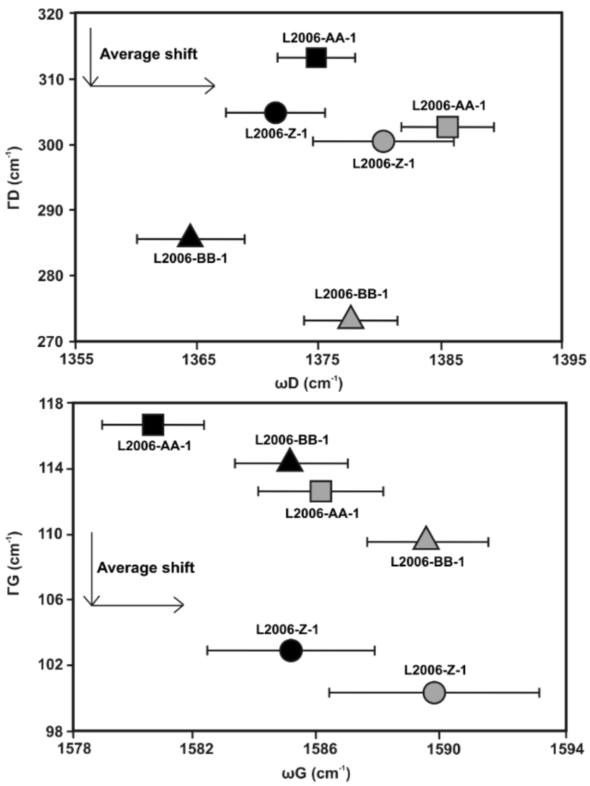

Figure 1: ωD versus ΓD and ωG versus ΓG for 3 IDPs from separate cluster particles (different symbol shapes) analysed using the 473nm (grey) and 514nm (black) lasers. Arrows indicate the average shift in band position and width.

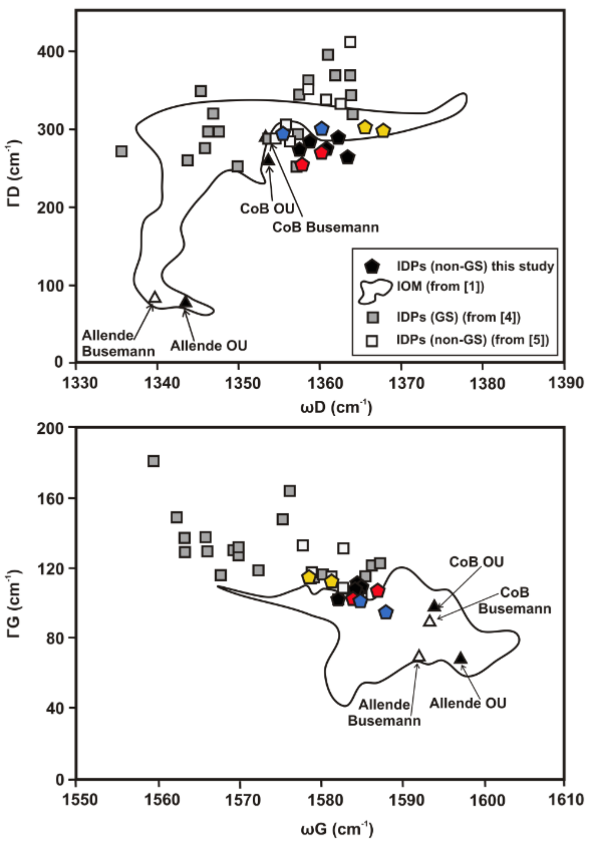

Figure 2: ωD versus ΓD and ωG versus ΓG for IDPs of this study, other non-GSC [5] and GSC IDPs [4,5]. The field for meteoritic IOM [1] is also shown plus some specific IOM comparisons from this study and [1]. Individual particles from clusters coloured differently. All data corrected to 523nm excitation values.

primitive nature of the samples, particularly when their large ΓD is taken into account. Peak metamorphic temperatures, calculated after [1], are low for all IDPs with values <220°C. The IDPs selected for this study do not appear to be as primitive as the Grigg-Skjellerup Collection (GSC) particles [4] but are comparable to other non-GSC IDPs [5]. The sample set is composed of multiple fragments of the five cluster particles. The range of ωD and ωG values from any one cluster is relatively limited, comparable to meteoritic IOM from CMs analysed as part of this study. For particles within individual cluster particles ωD varies by <6 cm^{-1} and ωG by <3 cm^{-1} (25 and 6 cm^{-1} respectively for ΓD and ΓG). While great isotopic and structural variability exists within individual particles at the sub-micron scale (*e.g.* [4]), these results indicate that the organic material within IDPs is reasonably homogeneous at the few to tens of micron scale, and that there is greater variability between particles than within. The variability between particles most likely reflects accretion from different reservoirs, potentially on the scale of individual comets given the distinctive signatures from GSC IDPs [4]. Secondary processes on the comet, in interplanetary space or during atmospheric entry may also play a role, although peak metamorphic temperatures were low (<220°C).

It is concluded that the results of these Raman analyses support the concept that IDPs originate from a primitive source such as a comet. The OM of the IDPs is primitive and the variation in D and G band properties within particles is smaller than that between particles – potentially reflecting heterogeneity across the protoplanetary disk. Future analysis of these particles will help to better constrain the origin of these features and allow investigation into the compositional and isotopic heterogeneity exhibited by cluster IDPs.

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