ROUGH AND IRREGULAR ICE CRYSTALS IN MID-LATITUDE CLOUDS

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1. INTRODUCTION

Cloud feedbacks are the largest source of uncertainty in climate models. In particular, uncertainties exist concerning the radiative forcing of clouds containing ice crystals, most notably cirrus. Indeed, whether cirrus clouds warm or cool the Earth's surface depends on ice crystal morphology. Reducing this uncertainty requires detailed *in situ* characterization of cloud particles. Also, detailed knowledge of the scattering properties of cloud particle types is needed for accurate retrieval of cloud microphysical properties from remote sensing. One of the main barriers to achieving these goals is the inability of cloud probes to determine the contribution of small ice crystals (that is crystals smaller than about 50 μm) to the total distribution. This is partly due to their inability to resolve the geometric structure of small ice crystals because of the conflicting demands of high optical resolution and large sample volume (Ulanowski et al. 2004, Connolly et al. 2007, Kaye et al. 2008).

There is also growing evidence, largely indirect, that atmospheric ice crystals have shapes departing from idealized geometries based on perfect hexagonal prisms (Garrett 2008, Baran 2012). Korolev et al. (2000) concluded that the majority of ice particles in mid-latitude stratiform clouds observed during several campaigns were of irregular shape. Best fits to data obtained by Lampert et al. (2009) in Arctic ice cloud were consistent with deeply rough hexagonal ice crystals. Gayet et al. (2011) found prevalent particles with imperfect or complex shapes at trailing edge of mid-latitude frontal cirrus. Measurements using the Polar Nephelometer (Shcherbakov et al. 2006) indicated that the surface of Antarctic ice crystals was deeply rough. It is important in this context that particle roughness can dramatically alter the scattering properties of ice crystals. For example, it can significantly reduce the asymmetry parameter (Yang et al. 2008, Ulanowski et al. 2006). Roughness may also account for the relative rarity of ice halos (Ulanowski 2005). Therefore it is important to quantify fine detail of ice crystal geometry, currently largely beyond the reach of imaging cloud probes, although there are indications that a new generation of probes using incoherent light may be able to resolve at least some of the detail (Schön et al. 2011).

It is possible to circumvent the optical resolution limitations of imaging probes by acquiring light scattering "patterns" instead of images. Such patterns can be obtained from relatively large sample volumes, as there is no sharply-defined image plane to limit resolution. Several light scattering cloud probes, jointly known as Small Ice Detectors (SID) have been developed over the last decade at the University of Hertfordshire. Successive models obtain scattering patterns with progressively higher angular resolution. The earlier designs rely on multi-element detectors measuring mainly the azimuthal scattering, while the most recent, collectively known as SID-3, acquire high-resolution two-dimensional (2D) scattering patterns (Kaye et al. 2008). 2D scattering patterns offer high potential for detailed particle characterization. It is possible to recover the shape, size and orientation of small ice particles by comparing such patterns to models such as the Ray Tracing with Diffraction on Facets (RTDF) scattering model (Clarke et al. 2006, Kaye et al. 2008). Ice particle roughness can also be obtained, as evidenced by experimental patterns from ice analogue crystals with smooth and rough surfaces, which show distinct differences: while the former have sharp, well-defined bright arcs and spots, the latter have much more random, "speckly" appearance, but with greater azimuthal symmetry (Ulanowski et al. 2006). Here we focus on the application of 2D scattering patterns to retrieving the roughness ice particles encountered in mid-latitude cirrus and mixed phase clouds.

2. METHODS

First *in situ* cloud data from the SID-3 probe was obtained during the Met Office CONSTRAIN campaign in Scotland in Feb. 2010 with another case in south England in

Sept. 2010. SID-3 was flown in a PMS-style canister on the FAAM research aircraft. The probe has “open” geometry similar to SID-2, to minimize ice particle shattering. Particle triggering, incident illumination (532 nm wavelength laser beam) and sample volume definition are also similar to SID-2 (Cotton et al. 2010). However, the main detector of SID-3 is an intensified CCD camera with a nominal resolution of 780 by 582 pixels (Kaye et al. 2008). The camera produces 2D scattering patterns from single particles at rates up to 30 per second, depending on configuration. Receiving optics collect the scattered light over an annulus covering scattering angles from 6° to 25°, sufficient to encompass the 22° halo scattering from ice prisms, but with the central low-angle area obscured by a beam stop.

Image texture can be quantified using statistical measures, e.g. the gray-level co-occurrence matrix (GLCM), which deals with spatial relationships of pairs of gray values of pixels. Previously, GLCM was applied to retrieving surface roughness from laser speckle images (Lu et al. 2006). Initially, four GLCM features were chosen: contrast, correlation, energy and homogeneity. In addition, image entropy and two measures relating to image brightness distribution, rather than texture, were examined: the ratio of root-mean-square (RMS) brightness to its standard deviation (*RMS*/*SD*, Jolic et al. 1994), and kurtosis. The measures were calculated for 2D patterns from a range of test particles: smooth and rough ice analogues, and mineral dust grains and correlated with a semi-quantitative measure of particle roughness. We also examined the sensitivity of the same measures to potential bias sources, including image noise, particle size, shape and orientation, and detector gain. Various image normalization and averaging schemes were compared too, and the chosen one involved scaling mean image brightness to 10 on the 0 - 255 scale.

Energy was found to have the strongest correlation with roughness and robustness with respect to the potential bias sources of all the GLCM measures. It is relevant that the GLCM energy also shows good correlation with surface roughness and is most robust with respect to variation of "the setup configuration, the position, and the orientation of the surface to be measured" in the context of laser speckle (Lu et al. 2006). Similar performance characterized the remaining measures, entropy, *RMS*/*SD* and kurtosis, but different measures showed sensitivity to different sources of bias. For example, entropy was slightly sensitive to gain and particle size. A combined feature was therefore defined, composed of the most robust measures, energy *E*, *RMS*/*SD* and the logarithm of kurtosis *K*, as follows:

$$RMS/(200SD) - 10E - \log(K)/5 + 0.7.$$

The combined roughness measure is weighted so that individual measures contribute to it approximately equally and it is centred on zero for the particles examined, with approximate bounds of ±1.

Scanning electron microscopy (SEM) images of test particles were taken using JEOL-5700 environmental SEM. Images of ice were obtained in the presence of water vapour in the SEM chamber. Ice was allowed to grow on a metal substrate on a Deben Ultra Peltier-cooled cold stage.

3. RESULTS AND DISCUSSION

Fig. 1 shows a typical selection of scattering patterns from ice particles from the flight campaign, in comparison with test particles. Both cirrus and mixed phase patterns are qualitatively similar to the rough rosette pattern. The roughness measures were calculated for a random selection of several hundred patterns from marine cirrus and mixed phase flights, as well as one cirrus flight in a continental airmass – the frequency distributions of the combined roughness measure are shown in Fig. 2. Roughness measures were also calculated for a range of test particles, including ice analogue crystals (Ulanowski et al. 2006) and mineral dust grains, all representing a range of surface roughness – the results are given at the bottom of Fig.2, with example test particles shown in Fig. 3. It can be seen that smoother test particles correspond to the tails of the roughness distributions, which are better represented by the rougher particles in the selection. Scattering from ice analogue rosettes very similar to those used here was previously characterized using a levitation technique. It was found that the transition from smooth to rough geometry for these large crystals lowered the asymmetry parameter from 0.81 to about 0.63 (Ulanowski et al. 2006). It is worth noting that such a large change corresponds to almost doubling the reflectivity of a cloud composed entirely of such particles.

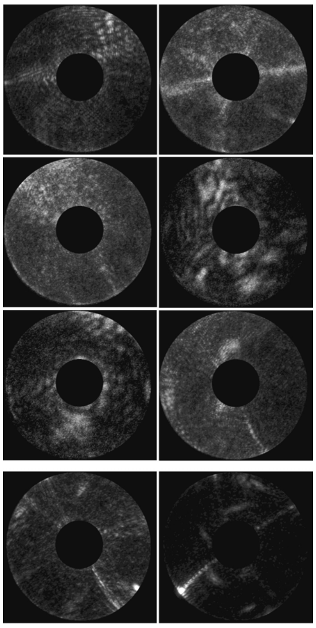

Fig. 1. Top 3 rows show 6 randomly selected SID-3 patterns from ice particles during mixed phase (left) and marine cirrus (right) flights, compared to patterns from ice-analogue rosettes with moderately rough (bottom left) and smooth surfaces (bottom right).

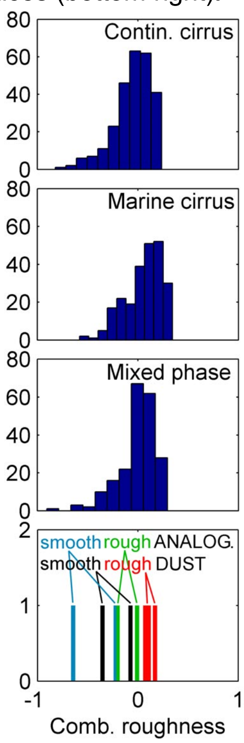

Fig. 2. Distributions of combined roughness measure from 2D SID3 patterns in continental cirrus, marine cirrus and mixed phase flights, compared to test particle roughness.

It is interesting to observe that, while rough particles dominated in all three cloud types, marine cirrus and mixed phase clouds were similar, but cirrus in a continental, polluted airflow had lower roughness. We speculate that this is due to higher concentration of ice nuclei in the last case. High-resolution cloud modeling shows that lower ice supersaturations can be found in continental than in marine cases (Flossmann and Wobrock 2010). This raises the possibility that the observed roughness may be the outcome of faster ice growth.

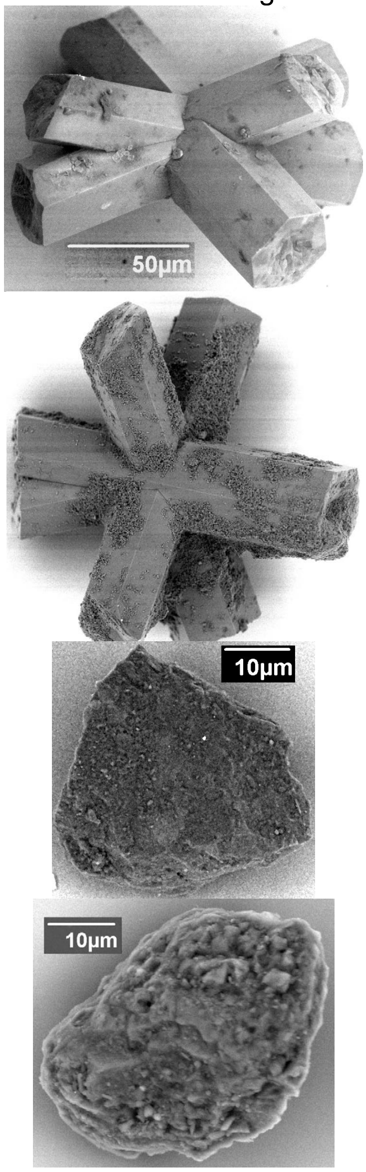

Fig. 3. SEM images of test ice analogues and mineral dust grains with roughness measure shown in Fig. 2. Max. dimension and combined roughness are given, top to bottom: 150 μm rosette –0.22, 160 μm rosette –0.01, 41 μm dust -0.07, 47 μm dust 0.17. The scattering patterns from the rosettes are shown in Fig. 1.

Roughness was similar in growth and sublimation zones of cirrus. This suggests that the currently prevailing view that ice crystals merely become rounded as they sublimate (Nelson 1998) may be incorrect. Indeed, laboratory experiments show that ice surfaces can become rough during fast sublimation (Cross 1969, Pfalzgraff et al. 2010,Ritter et al. 2012). Sublimating ice crystals seen at high resolution is shown Fig. 4. While rounded crystals are frequently observed using imaging cloud probes, optical resolution limitations conceal fine detail associated with roughness (Ulanowski et al. 2004, Connolly et al. 2007).

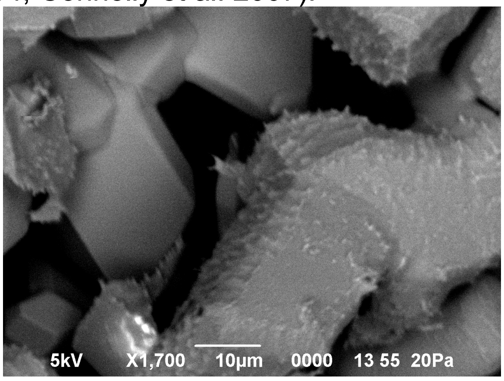

Fig.4. Scanning electron microscopy image of ice crystals sublimating near –40°C. Note that surfaces deeper within the sample, where subsaturation is expected to be weaker (due to lower temperature and/or higher local water vapour pressure), remain smooth while the exposed surfaces are rough.

4. CONCLUSIONS

SID-3 probe was flown on the FAAM aircraft in mid-latitude clouds. Unlike most earlier results from cloud chambers, where SID-3 2D scattering patterns typically displayed characteristics attributable to idealized geometric crystal shapes, the majority of the cloud patterns showed random, “speckly” appearance. Lab experiments show that this is typical of particles with rough surfaces or complex structure. Quantitative comparison of lab and cloud data was done using pattern texture measures, originally developed for surface roughness analysis using laser speckle. The results are consistent with the presence of strong roughness in the majority of cirrus and mixed phase cloud ice crystals, at levels similar to those found in rough ice analogue and mineral dust particles used for reference. Similar roughness was found in the growth and sublimation zones of cirrus, suggesting that roughness can be maintained or possibly even reinforced by sublimation. Slightly weaker roughness was present in cirrus in polluted airmass of continental origin than in marine cirrus, possibly as an indirect outcome of higher concentrations of ice nuclei in the former.

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