

## ***Interactive comment on* “Observations of ice multiplication in a weakly convective cell embedded in supercooled mid-level stratus” by J. Crosier et al.**

**J. Crosier et al.**

j.crosier@manchester.ac.uk

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We thank the referee for their comments and suggestions. Our responses to the comments (along with the original comments in italic) are given below.

*Crosier et al. report measurements of ice crystal concentrations in mixed-phase stratus clouds over southern England. They present a detailed analysis of the cloud microphysics measured during the flights, and corresponding remote sensing measurements of the sampled clouds. Good evidence for the high concentrations of ice nuclei being primarily due to secondary ice crystal production mechanisms,*

*the Hallett-Mossop process in particular, is provided. Seeding of ice crystals from above the measured cloud is also convincingly discounted. Where the analysis and arguments fall apart is in regards to the aerosol concentrations and chemical composition measurements, and the interpreted source of the initial ice nuclei responsible for cloud glaciation at rather warm temperatures  $> -15$  C. The conclusions regarding the sources and efficiency of the likely ice nuclei do not agree with the current understanding of atmospheric ice nuclei. Unfortunately, the Aerosol Mass Spectrometer is not an appropriate instrument for determining possible ice nuclei compositions as it detects only non-refractory aerosol components that evaporate. It cannot detect mineral dust or biological particles, which are the most efficient ice nuclei, particularly at warm mixed-phase cloud temperatures of  $> -15$  C. The manuscript therefore requires major revisions to address these issues before it can be reconsidered for publication. The subject matter of this manuscript fits well with the scope of ACP.*

**The main objective of the paper was to highlight the cloud microphysical processes occurring (primary nucleation and secondary ice formation). The aerosol measurements are presented in a complimentary manner but are not the main focus. The referee does make a valid point regarding the limitations of the Aerosol Mass Spectrometer, in that it can neither detect mineral dust particles nor identify biological particles. Therefore we cannot make any statements regarding the amounts of these particle types. However we do still consider it appropriate to discuss those aerosol measurements which were taken on the aircraft, even if they are not using the best measurement techniques available to the international community. We will insert the following statements into the manuscript to highlight the lack of mineral dust/biological particle information.**

**Section 2.1 In situ measurements, p19385(5), line 26.**

**“The C-ToF-AMS is unable to detect mineral dust particles and has not been shown to be able to identify biological particles in the ambient aerosol population. These have both been reported as efficient ice nuclei (Connolly et al., 2009,**

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DeMott and Prenni., 2010). Therefore the C-ToF-AMS is unable to characterise an important subset of the ambient aerosol population with regards to potential Ice Nuclei.”

Section 2 In-situ aerosol properties, p19394(13), line 3.

“Quantification of dust and biological particle concentrations was not possible due to instrumentation limitations.”

Section 6 Discussion and Conclusions, p19395(15), line 4.

Change

“The AMS measurements showed that these aerosols were a mixture of sulphate and carbonaceous material.”

to the following:

“The C-ToF-AMS measurements showed the bulk submicron non-refractory particulate mass consisted of a mixture of sulphate and carbonaceous material. No estimate of the amount of influence from mineral dust/biological particles (both of which have been shown to be effective ice nuclei) could be determined from the measurements taken.”

*My major issues are in regards to the aerosol measurements presented. First, it would be useful if the size range of aerosol and cloud particles measured by the various detectors was re-iterated when the data are discussed, and also given in the Figure Captions. Section 2.1 states “Aerosol particle size distributions ( $0.1 < \text{Diameter} < 3.0 \mu\text{m}$ ) were measured with a wing pylon mounted Passive Cavity Aerosol Spectrometer Probe (PCASP, PMS).”, but Table 2 shows the smallest particle size*

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detected as  $0.61 \mu\text{m}$ . Which is it?

**All size ranges for all cloud probes are summarised in Table 2, which is accurate. The PCASP (which is an aerosol spectrometer and not a cloud probe) has been purposefully omitted from this table. Its size range is  $0.1 - 3 \mu\text{m}$ . The referee has confused the size range of the PCASP with that of the CAS, which is  $0.61 - 50 \mu\text{m}$  as summarised in table 2. The PCASP size range is already stated in the text several times. It is also stated in any figure captions where the data appears in the corresponding figure (e.g. Figure 14). We apologise for the similarity in some of the acronyms, but feel they should not be altered as these are standard names as used in the literature.**

*On a related topic, the CDP particle concentrations are displayed in several figures. This instrument detects particles with  $2.0 < \mu\text{m} < 50$ . It will therefore miss the majority of aerosol particles present, as number concentrations maximize at submicron sizes. These undetected particles are also a large potential source of ice nuclei concentrations (DeMott et al., 2010). The manuscript should be clarified to make these issues clear to the reader.*

**We feel the referee has miss-understood the use of the CDP data. The CDP size range covers that of typical cloud droplet sizes. The data are used as an indicator for in cloud conditions. It is not being used to quantify large aerosol particles.**

*It does not appear that any measurements of submicron aerosol concentrations were performed here, which severely limits any interpretation of likely ice nuclei sources and efficiencies responsible for cloud glaciation.*

**We feel this is not true. We present/discuss both mass and number concentrations of submicron aerosol in several parts of the paper (see Figures 13 and 14 and associated text).**

*In Fig. 13 the AMS submicron aerosol mass concentrations are shown along*

*with the CDP supermicron particle number concentrations. It should be clarified that the two instruments do not detect the same size range of particles. The AMS's transmission efficiency falls off rapidly above 600 nm. This likely explains why the two data traces are clearly uncorrelated in Fig. 13, perhaps even anti-correlated.*

**We agree that the CDP and AMS data are not correlated. This is no surprise. This is because the AMS data is submicron mass from what is effectively an interstitial inlet, whereas CDP is cloud droplet number concentration (it is not a measure of aerosol particle number concentration). We feel this is clearly written in section 5, paragraph 1. However, we intend to add the following as clarification.**

**Section 5, p19393(13), line 16.**

**“The CDP data are included on Fig. 13 to inform the reader when in-cloud conditions were encountered.”**

*Why is no AMS data shown in Fig. 13 just above 1 km when the CDP counts maximized? It is important to specify that the AMS measures non-refractory aerosol mass. Therefore, no mineral dust, elemental carbon, bioparticles, or other components that don't vaporize are detected, and these particle compositions are the most likely source of ice nuclei. Why wasn't aerosol composition obtained for the below cloud runs? Was sampling suspended when flying through precipitation?*

**There is no AMS data in the section of the omitted cloud in the plot, as the AMS inlet was closed to prevent ingress of large amounts of water which can damage the instrument. This is also the case for in/below cloud runs R2/R3+R4. The following will be added to the manuscript. The comments regarding mineral dust and biological particles are dealt with later.**

**Section 5 In-situ Aerosol Properties, p19393(13), line 16.**

**“No data from the C-ToF-AMS is shown in figure 13 during the profile ascent**

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between 1 - 1.5 km altitude, as sampling was suspended to prevent instrument contamination with water. The C-ToF-AMS sampling was also suspended during runs R2-R4 for the same reason.”

*Remotely-sensed cloud-top and base temperatures are used to infer the temperature that both primary and secondary ice nucleation occurred at, between -12 and -10 C.*

**Temperatures were predominantly determined from the in-situ measurements on the aircraft. The satellite data were used simply to provide the reader with an idea of the size of the features associated with the cloud.**

*Ice nucleation efficiency at  $T > -15$  C is typically quite low for mineral dust particles. The only known possible source of ice nuclei at this temperature would be certain bioparticles such as some bacteria (DeMott and Prenni, 2010). Do you have any evidence to support or reject this?*

**We agree with the referee that bioparticles are a candidate for being involved as IN. We have no information to confirm/rule out their role. We will add the following text to highlight this fact.**

**Section 6, Discussion and Conclusions, p19395(15), line 7.**

**“The instrumentation used in this study could not confirm/rule out the presence of either mineral dust or biological particles. Both of these particle types have been shown to be efficient ice nuclei at colder temperatures (Connolly et al 2009, DeMott et al 2010). Biological particles could also be responsible for heterogenous ice nucleation at relatively warmer temperatures ( $>-15$  C, DeMott and Prenni, 2010). Studies have demonstrated the ability of some organic compounds to form a glassy phase at low temperatures which can act as ice nuclei (Murry et al., 2010). Glassy organic aerosol particles have recently been detected at room temperatures (Vertanen et al., 2010), and these could also be**

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a potential source of efficient ice nuclei at relatively warm temperatures. There is no evidence of dust outflow from Africa based on 7 day back trajectories generated from ECMWF wind field data.”

We also need to add the following to the Acknowledgements section: “We also acknowledge the British Atmospheric Data Centre for the back-trajectory data (<http://badc.nerc.ac.uk/community/trajectory/>).”

*The mixed sulfate/carbonaceous aerosol measured by the AMS is not a likely source of heterogeneous ice nuclei, based on the body of laboratory and field studies of ice nucleation. The known sources of efficient IN deserved further discussion with reference to other papers in the literature. Soot is a poor ice nuclei even at -30 C, and even worse at > -15 C (Karcher et al., 2007; Phillips et al., 2008). If the carbonaceous signal is from biological particles this could explain ice nucleation at these warm temperatures > -15 C. At this temperature the most likely sources of IN are biological particles and perhaps mineral dust particles, neither of which are detected by the AMS. It does not appear that the authors can make any convincing attribution as to the source of the ice nuclei with the presented measurements; it would be pure speculation. Mineral dust could likely be ruled out based on back-trajectory analysis, MODIS images, and the measured aerosol size distributions (which omit the submicron aerosol modes).*

**This comment has been addressed by the previous response.**

*Page 14: “Roughly 1 in 500 particles entrained would need to be efficient IN at temperatures > -12.0 C.” This is a preposterous statement completely unsupported by our current understanding of ice nuclei sources and efficiencies (DeMott et al., 2010). Only at lower temperatures of < -20 C do some mineral dust types display this high ice nucleation ability via immersion-freezing (Connolly et al., 2009; Welti et al., 2009). Biological particles are the only known source of ice nuclei that could exhibit this high*

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*ice nucleation efficiency (DeMott and Prenni, 2010), but bioparticles acting as the ice nuclei are not discussed here. The fact that a large fraction of the total CN is also not being measured produces another large error in this estimated 1 in 500 IN efficiency.*

**Our estimates are based on PCASP aerosol number concentration (0.1 - 3  $\mu\text{m}$ ), and not CDP concentration (2-50  $\mu\text{m}$ ) as the referee earlier suggested. Therefore our estimate of the CN concentration will be somewhat low as it misses out sub 100 nm particles. However, DeMott et al (2010) have shown a strong correlation between ice nuclei concentrations and particles greater than 500 nm, demonstrating a lack of sensitivity to the smallest particles. We have already addressed the comment regarding mineral dust and biological particles in a previous comment.**

*In Section 4 the cloud temperatures of R1 and R2 are stated to be above zero degrees. Is a negative sign missing?*

**The manuscript already has negative signs for the cloud temperatures.**

*Please justify using a collision efficiency between drops and ice crystals of 1. Was the sensitivity to this parameter tested?*

**The use of a collection efficiency of unity between drops and ice crystal was deemed reasonable due to the low updrafts in the system, and also the low estimated fallspeed of the ice crystals. Comparison with collection efficiencies from the study of Beard and Glover (1974) have already been performed and are already included in the manuscript (p19398(18), lines 14-15), as has the reference.**

*“Relaxing the constraint where only drops with  $D > 24 \mu\text{m}$  allow splinter production to occur...”, does this mean that the constraint was completely removed, or was the size threshold moved to a lower particle size? The sensitivity to the size cutoff for this constraint should be tested.*

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The constraint was completely removed so drops of all sizes can produce splinters. The sensitivity of the result of this calculation is very sensitive at around  $15\mu\text{m}$  diameter, as this is the typical droplet size. We will reword the sentence as follows:

**“Removing the constraint where only drops with  $D > 24 \mu\text{m}$  allow splinter production to occur (to a situation where all droplets measured by the CDP can allow splinter production).....”**

*Most of the Figures, especially Figs. 4-9, are too small and difficult to read.*

**We feel these figures are acceptable if they are given enough space in the final manuscript.**

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