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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ice formation and development in aged, wintertime cumulus over the UK : observations and modelling" *by* I. Crawford et al.

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We thank the referee for their comments and suggestions. Our responses to the comments are given below.

Crawford et al. present observations of a certain case of wintertime cumulus over the UK, in which very high ice crystal concentrations were observed despite quite warm temperatures. With the help of different measurements, two models and a number of back-of-the-envelope calculations, the authors search for explanations of this finding. The problem splits up into two aspects: (1) How can the primary ice formation be explained? and (2) How important is secondary ice formation? For neither of these

questions, definite answers can be provided. Most attempts for closure between models and measurements fail at least to some degree. Examples are: surface aerosol and vertical velocity - cloud droplet number; dust and biological aerosols - IN; splinter production. These mismatches are somewhat frustrating for the reader. In my opinion, it can be legitimate to conclude a paper with open questions and future research needs, and that the topic of this manuscript is of high enough interest to be published without providing definite answers. However, I found this manuscript quite difficult to read, because it is long and lacks a clear thread. The organization into sections and subsections does not seem well enough thought through. I recommend a number of revisions, mainly concerning the structure and the discussion of primary ice nucleation

The abstract should be shortened. Here it should become clearer what story the authors want to tell

Response: We agree with the reviewers comment and the abstract has been shortened.

The outline of this manuscript is unclear: 1 Introduction

1.1 Sampling strategy - this doesn't fit into the introduction

2 Shallow convection on the Chilbolton radial

2.1 Meteorological conditions

2.2 Cloud properties

2.3 In-situ aerosol properties

2.3.1 Coupling of observed surface and Airborne Aerosol Measurements – it is not useful to introduce subsubsections if there is only one

2.4 Sensitivity Studies of the HM Mechanism using WRF

2.5 Sensitivity studies of the HM Mechanism using ACPIM - this includes a description of the ACPIM model. However, ACPIM was already introduced in 2.3.1 with a brief model description

2.5.1 ACPIM Parcel Model Results - the title doesn't explain in how far this section is different from 2.5. One would rather expect an inverse order of 2.5 and 2.5.1 (from their titles).

3 Source of the primary ice nuclei - why is this not included under section 2?

4 The Hallett-Mossop secondary production mechanism - HM was already discussed in 2.4 and 2.5, so it is confusing that this is taken up again.

5 Summary of model results - Why only now if modelling was discussed in 2.4 and 2.5? And where is the summary of the observations?

6 Conclusion

The article would benefit from some reorganization, e.g. reordering as follows: introduction - description of instruments and flights - description of the models - observational results - model results – discussion

Response: We thank the reviewer for their suggestion. We have restructured the paper as suggested in the following order:

1 Introduction

2 Sampling strategy

3 Description of models

3.1 WRF

3.2 ACPIM

4 Observational results

4.1 Cloud properties

4.2 In-situ aerosol properties

5 Modelling results

5.1 Sensitivity studies of the HM process using WRF

5.2 Sensitivity studies of the HM process using ACPIM

5.2.1 Aerosol number sensitivity

5.2.2 Kernel sensitivity

5.2.3 Composition sensitivity

5.2.4 IN sensitivity

6 Summary of model results

7 Source of the primary ice nuclei

7.1 IN from the dust fraction of the measured aerosol

7.2 IN from the biological fraction of the measured aerosol

8 The Hallett-Mossop secondary production mechanism

9 Conclusions

p 30801, l 22: It should be mentioned already here that this discrepancy in older data might be due to issues with shattering.

Response: We agree and we have modified the text to the following: "there is strong evidence to show that ice crystal concentrations in this temperature regime frequently exceed this by in some cases 4 orders of magnitude (c.f. Table 1 in Mossop (1978)), however, this may be subject to shattering artefacts (McFarquhar et al., 2007)."

p 30809, l 28: Explain better how the composition was determined. Does this refer to a specific size range only? The dust and primary biological components are not mentioned here, please reconcile.

Response: The sub-micron non-refractory aerosol composition was measured using a C-ToF-AMS. This was then categorised using a Positive Matrix Factorisation technique (PMF). For clarity we have included an additional reference (Lanz et al. (2007)) which gives example spectra from the techniques early field use. Additionally we have renamed the compositional categories to be consistent with the current terminology used by the AMS community. We have also rearranged the paper to summarise all the aerosol measurements in the sampling strategy section which now covers how the dust and primary biological components were determined.

p 30812, l 1: "a detailed modelling study simulating the passage of a thermal bubble": what model does this refer to? This needs to be better explained (or removed)

Response: We have undertaken further modelling studies to respond to reviewers comments. As a result this work is no longer considered to be important and has been removed.

p 30812, l 23: The effect of a small number of sea salt aerosol on the activated droplet number might be significant at small vertical velocities due to the competition effect (Ghan et al., 1998).

Response: This has now been removed from the CCN discussion which has now been significantly reworked to concentrate on aerosol compositional effects, see next response.

30811 and 30812: It is unclear to me how all the points discussed in this section fit together. ACPIM simulates 350/cc with the modal vertical velocity, observations show peak values of 150/cc (why not give the modal value as well?), but the aerosol concentrations entering ACPIM are apparently to be scaled by a factor 1/6. Nevertheless, it is stated that "the aerosols measured at the ground are strongly linked to the aerosols at 750m" (which is just below cloud base).

Response: We have reviewed this work again, concentrating on the aerosol composition used in the model as constrained by the ground based AMS and HTDMA measurements. We have shown that when the largest aerosol modes were an internal mixture comprised of ammonium sulphate, ammonium nitrate and fulvic acid , and the smallest mode is pure fulvic acid we get good agreement with the measured CCN without the need to scale the aerosol number concentration. We have included a full discussion of this work in the manuscript, in which several different compositional cases were trialled.

p 30813, l 28: What is an "acceptable simulation"? Were any objective criteria applied?

Response: Although strict objective criteria were not applied, we did conduct a qualitative comparison of the simulated reflectivity with that observed from the vertically-pointing Chilbolton radar (see plots below, figs. 1 & 2). This showed that the model captured the timing of the main frontal rainbands well, and gave confidence that the model provided a realistic simulation of the general meteorology on the day. Although the signature of the shallow convective cloud is clearly visible in the model reflectivity field at approximately the correct time of day, it is important to point out that the origin of the cloud is slightly different in the model as it spins up too early over the SW peninsula, and advects eastwards over the UK until it reaches the Chilbolton area. During this time precipitation is maintained as the simulated cloud is continually replenished as it heads eastwards. This is different to the history of the convective cloud provided by the NIMROD rainfall radar, which shows a later spin-up. Possible explanations for the replenishing effect in the model are considered later in response to a later comment.

p 30814, l 12: The change of the model output interval is trivial and doesn't need to be mentioned.

Response: Given the length of the paper we have omitted this sentence from this section.

p 30814, l 20: "insufficient vertical resolution" - how many levels are below 2km?

Response: We use 81 vertical levels in total, with 14 levels up to 2km.

p 30816, l 4ff: This discussion of the Cooper and Meyers ice nucleation parameterization in the WRF model is of little relevance for this case study. Their relative contributions don't say much about the actually occurring ice formation process. It is important to note that neither Cooper nor Meyers were developed for such warm temperatures.

Response: We argue that this discussion is necessary for clarity of the model set-up and to allow reproducibility of results. Whilst admittedly neither Meyers nor the Cooper scheme were developed for such warm temperatures, we use these schemes in the absence of a better alternative. What the results of this section show is that when IN are not being depleted (because WRF does not use prognostic aerosol), the nucleation schemes act to maintain a steady ice crystal concentration that has the effect of continually replenishing the cloud and sustaining precipitation in an unrealistic way. Thus we have identified a weakness inherent to mesoscale models when simulating shallow convection at relatively warm temperatures, which may be improved through a prognostic treatment of IN in future work. We have made this point clearer in the revised paper.

Why isn't the DeMott et al. (2010) parameterization employed in WRF, same as in ACPIM? This would make the simulations more comparable.

Response: The DeMott scheme depends on the number of aerosol above 500nm, but the WRF simulations do not use a prognostic treatment of aerosol, so we use the purely temperature based nucleation parameterizations as used in the standard Morrison microphysics scheme. This is to show how a typical mesoscale model performs in this instance, and thus the aims of the WRF modelling are quite different to those of the APCIM modelling, which is more focused on the specific details of the role of aerosol and ice-phase processes within the cloud.

p 30818, l 18: No discussion about the different bin schemes needed here.

Response: We disagree and we feel this is useful information for the modelling community given the possibility of higher numerical diffusion in the other schemes.

p 30820, l 5: It would make more sense to multiply the value of ns(T) by varying factors > 1 instead of shifting the temperature. Strictly speaking, the temperature shifts would lead to ice nucleation at T > 0C (which I'm sure was suppressed)

Response: We thank the reviewer for the suggestion and we have performed the analysis using the DeMott et al. (2010) scheme multiplied by several factors (INx1, INx10 and INx100) which replaces the temperature shifted analysis and discussion in the manuscript.

p 30821, l 12: Is this for the "low" of the "high" aerosol run?

Response: This is for the Low aerosol run.

p 30821, l 15: Does this refer to droplet or ice nucleation? If droplet activation, why wasn't this discussed in 2.3.1?

Response: This refers to droplet activation, however, this has now been removed from the manuscript.

p 30823, eq. 2 and the following discussion: I strongly recommend to disregard this equation and the attempt to derive IN numbers at a given temperature from it. As noted later, the droplet volume dependence included in this equation is not reasonable for the case of particles including only one particle. The whole discussion would be more conclusive if only ice active fractions were discussed.

Response: We have removed the equation and discussion of it from the manuscript as the reviewer has suggested and we have concentrated the discussion on studies based on ice active fractions.

p 30824, l 10: 5x10ˆ6: The correct number from Levin & Yankofsky (1983) is 5x10ˆ6 bacteria per 1mm drop (page 1965, second sentence of their section 3). However, note that the 50% freezing from their Figure 1(b) refers to droplets of radii of 220 to 360 μm (see their section 2a).

Response: It isn't explained clearly in the Levin and Yankofsky paper. However, section three "droplets of the same size [1 mm] containing 5e6 dried cells". This was at -5C and they rapidly froze, suggesting that about 1 in 1eˆ7 particles may initiate freezing. The paper doesn't actually say how many particles the 220 to 360 micron drops in the wind tunnel contain unfortunately. With these numbers our estimates for the bio-in concentration can be revised as follows:

bio-aerosols measured with WIBS = 0.1 cmˆ-3

active bioparticle fraction is approximately 1 in 5eˆ7 cells.

yielding an IN contribution from bacteria of 1eˆ-5 Lˆ-1.

INA fractions of bacteria: Only one study is cited here to derive the estimate of bacterial IN. This is problematic, as INA fractions vary greatly between different bacterial species, strains and can even change as a function of the sample preparation. For alternative estimates, see Phillips et al. (2008) and Hoose et al. (2010). For recent measurement of INA fractions, see e.g. Möhler et al. (2008) or DeMott et al. (2011).

Response: We cite the Möhler et al. (2008) paper and use the suggested INA fraction contained within in conjunction with the WIBS PBAP measurements. We accept that its use in the paper isn't clear and this is expanded upon in a later response.

INA fractions of pollen: Because the number of pollen per droplet in the immersion freezing experiments of Diehl et al. (2002) is very uncertain, I recommend using the condensation freezing results shown in Diehl et al. (2001) instead. These refer to individual pollen grains.

Response: We have discounted pollen as a potential IN as the case study was performed in January, where the pollen concentration would be negligible. Additionally there were very few particles greater than 10 μm classified as biological by the WIBS,

further indicating that there was no pollen present. We therefore have removed the discussion about pollen acting as IN from the manuscript.

p 30824, l 16: Number of bacteria per particle: I disagree with the statement that it is likely that the bacteria occurred as single cells. Burrows et al. (2009b) and Huffman et al. (2010) find that bacteria are likely to occur in clumps. Individual cells are mostly around 1 μm

Response: Using the assumption that each biological particle only contains a single bacterium yields an IN concentration of 1x10ˆ-5 L-1 for the case considered, suggesting that bacteria is not a likely source of IN when compared to the contribution from dust of 1x10ˆ-2 Lˆ-1. WIBS PBAB measurements showed the biological aerosol to be of a few microns in size so in the event that each particle contained approximately 10 bacterium this would yield an IN concentration of 1x10ˆ-4 Lˆ-1, which is insignificant when compared to the dust contribution. So while the assumption may not be realistic it has a minimal impact upon the conclusions presented.

p 30824, l 23: pollen diameter: The pollen grains studied by Diehl et al. (2002) had diameters around 30 μm. Here, 50 μm are assumed, while on p 30826, l6, it is stated that "most pollen particles range in size from 15-40 μm diameter". Please harmonize.

Response: The section discussing pollens contribution to IN has been removed as discussed in a previous response.

p 30825, l 9: Here it is argued that the observed biological particles in the size range of a few micrometers could be fragments of pollen, and these are assumed to have the same ice nucleation properties as the intact pollen grains. This is highly unlikely, as ice nucleation scales with the surface of particles, and therefore smaller particles should be worse ice nuclei.

Response: The section discussing pollens contribution to IN been removed as discussed in a previous response.

Why don't you estimate the pollen IN based on the number of particles > 10μm?

Response: The section discussing pollens contribution to IN has been removed as discussed in a previous response.

p 30825, l 11: This paragraph comes back to the discussion of bacteria. Please move this part to the previous discussion of bacteria. This would then deal with my previous comment.

Response: This now flows from the discussion of bacteria as the pollen discussion has been removed.

p 30825, l 13: "could account for IN concentrations of around 0.01 L$\hat{}-1$": Where does this number come from? Is an INA fraction determined from the experiments used?

Response: In the conclusions of Möhler et al. (2008) an INA fraction of 10ˆ-4 is suggested for the examined biological particles over their active range of temperature -7 to -11C. We accept that this isn't clear in the manuscript and we have changed: "However, it was found by Möhler et al. (2008) that pseudomonas Syringae bacteria were active as ice nuclei at -8C and could account for IN concentrations of around 0.01 Lˆ-1 although we do not have any measurements specific to this species of bacteria" To: "Möhler et al. (2008) determined the ice active fraction of several biological aerosol where they derived a typical ice active fraction of 10ˆ-4 over the examined bio aerosols active temperature range of -7C and -11C. Applying this suggested ice active fraction to the average WIBS biological particle number concentration (~0.1 cmˆ-3) yields an IN concentration of 0.01 Lˆ-1."

p 30825, l 13: "we do not have any measurements specific to this species of bacteria": This is also a problem when using the Levin and Yankofsky (1983) data.

Response: We accept this limitation and we used the general biological particle INA fraction presented in Möhler et al. (2008). We have also removed the quoted text from the manuscript to reflect this generality.

p 30825, l 17: This discussion is indeed inconclusive and somewhat confusing. Putting all the information together, it could be tried to obtain estimates of the possible contributions of pollen, pollen fragments, bacteria and dust to the total IN number, as a function of temperature. This could look somehow like the sketch which is attached. I'm not sure whether the result will look as in my sketch, but if it does, then it would support your claim that biological IN could be significant at the temperatures of the observed cases

Response: We agree that the original manuscript is confusing and this section has been reworked. When the Levin and Yankofsky study was used we yielded an IN concentration much lower than was found for dust, suggesting that dust was the likely candidate IN. However, using the Möhler et al. (2008) study yields an IN concentration which was comparable to that of dusts IN contribution. We have highlighted this in the discussion and we have suggested that more laboratory studies of biological IN are required.

Fungi, lichen and plant fragments have also been suggested as biological IN. These should be mentioned as well.

Response: We agree and the above has been added to the discussion of candidate biological IN. "Further, Conen et al. (2011) found that soil particles which consisted of a mixture of mineral and biological material were sometimes able to act as ice nuclei at temperatures as high as $-7°$C" Has been changed to: "Further, Conen et al. (2011) found that soil particles which consisted of a mixture of mineral and biological material were sometimes able to act as ice nuclei at temperatures as high as 7°C and fungi, lichen and plant fragments have also been shown to act as ice nuclei (Després et al., 2012).

It should be emphasized in this section that in-situ IN measurements are not available for this case, and that there is no way to further constrain the estimated biological and non-biological IN concentrations.

Response: This has been clarified in the text. We have included the following in the final paragraph of the biological IN section: “While inconclusive, due to the lack of aircraft in-situ biological particle data to constrain the estimated biological and non-biological IN concentrations, calculations based on laboratory data and the observed bioaerosol concentrations at the surface, allow us to suggest that it is entirely plausible that a significant fraction of the primary ice nuclei at the observed temperatures could be of biological origin.”

Figures 3, 4, 5: Please explain the variable "Nround" and how it is to be interpreted (in the text).

Response: A description of the classification scheme used is given in appendix B of Crosier at al. (2011) as referenced in the text. The total area (A) and perimeter (P) of each particle is determined from the raw image. These parameters then used to determine the particle circularity (C) by: $C=P^2/4\pi A$ Where particles with a circularity greater than 1.25 are considered to be ice and less than 1.25 are considered to be liquid (round). Particles with less than 16 pixels are classified as small due to having insufficient resolution to determine shape. In this case Nround is the number concentration of the round classification and should be interpreted as liquid droplets/drizzle.

Figures 10 and 11: Not much can be seen on these plots. Show the difference or relative change.

Response: We agree and difference plots have been added to the figures.

Figure 11: The precipitation is shown for a 5-minute interval. This could mean that the precipitation enhancement is rather noise in the disturbed simulation than a consistent signal.

Response: We thank the reviewer for their comment and we have now used a 20 minute interval to improve the signal-to-noise ratio.

Figure 12: "lower aerosol concentration reduced by a factor of 6": Isn't the lower

aerosol concentration given by the high aerosol already divided by 6? Or is this even further reduced here?

Response: This is the low aerosol concentration. This analysis is no longer in the paper as this section has now been reworked.

Figures 7, 13, 14: There is too much discussion of the results in the captions. This belongs into the text; the captions should only describe what is plotted

Response: These figures have been changed in light of the new analysis.

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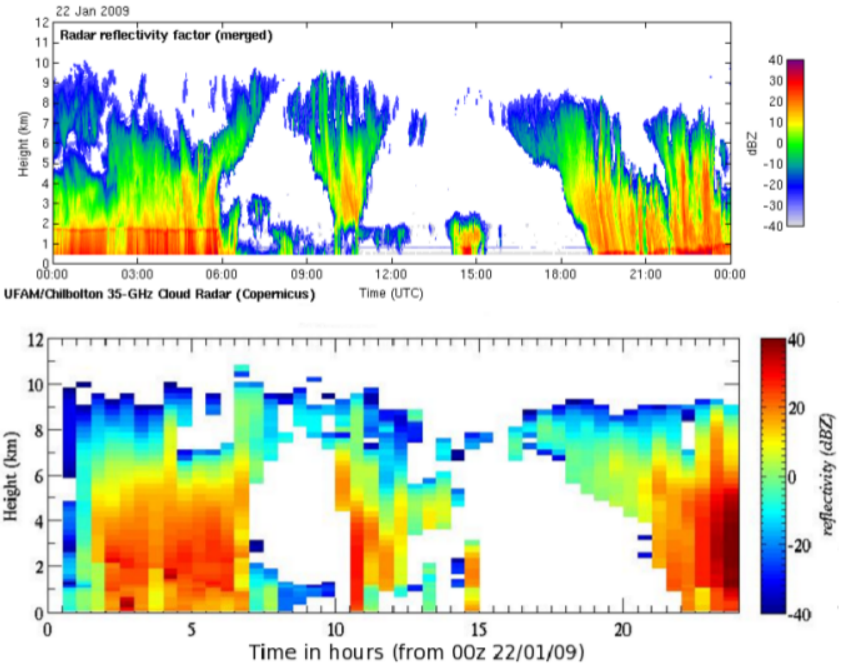

Fig. 1. comparison of vertical pointing radar and WRF simulated reflectivity

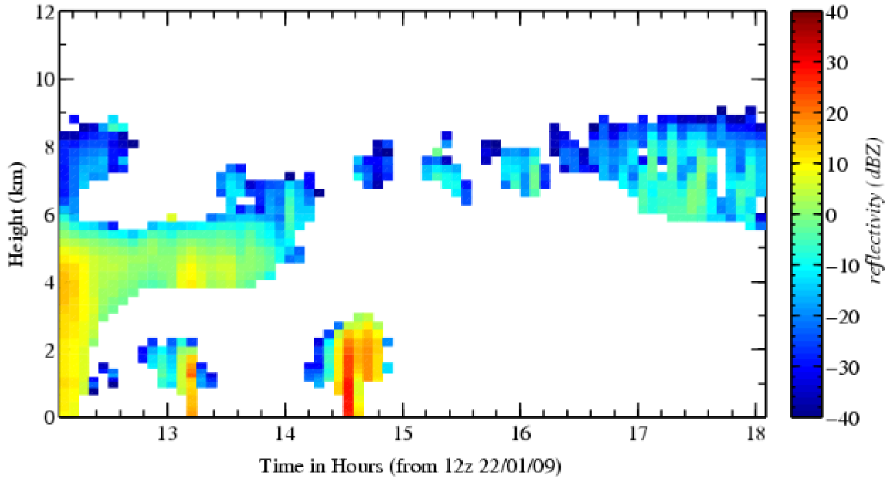

Fig. 2. same as fig.1 but at higher temporal resolution