Atmos. Chem. Phys. Discuss., 6, S5262–S5275, 2006
www.atmos-chem-phys-discuss.net/6/S5262/2006/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Technical note: A numerical test-bed for detailed ice nucleation studies in the AIDA cloud simulation chamber" *by* R. J. Cotton et al.

R. J. Cotton et al.

Received and published: 11 December 2006

1 Response to first reviewer

I thank the reviewer for the helpfull comments. The following are the replies and changes to the text.

1.1 General comment:

The definition and justification for assumptions made about ice nucleation and crystal growth have been included, in particular the sensitivity to the deposition coefficient and crystal capacitance. The accuracy of using the measured total water content to constrain the crystal growth has been further discussed and justified. The introduction and the discussion section on the sensitivity to different more 'physical' nucleation have been shortened.

1.2 1)Abstract, line 8:

What is a size-resolving parcel model?

This description has been removed.

1.3 2)Introduction:

This section could be clearer in introducing the topic of the paper.

The advance that this model provides over earlier models has been made clearer. The review of ice nucleation has been shortened for conciseness.

1.4 a)Page 9484, line 22:

In the absence of consideration of secondary ice formation processes, I believe it must remain a hyposthesis that the initial ice formation mechanism might determine the concentration and mean size of ice particles...

ACPD
6, S5262–S5275, 2006

EGU

Secondary ice processes will increase the ice number concentration significantly. In the AIDA chamber, during the ice nucleation phase, these will be negligable. In the atmosphere however the point is valid.

The following has been added to the text: In the absence of secondary ice formation processes, such as the Hallet and Mossop (1974) rime-splintering mechanism, the initial ice nucleation formation mechanism determines the ice particle number concentration which changes the mean size and therefore precipitation, microphysical processes such as aggregation and particle growth, and cloud radiative properties.

1.5 b)Page 9485, lines 2 to 3:

Heterogeneous ice nucleation has always been considered the first process for ice formation in clouds warmer than about -38C, has it not?

The text has been changed to clarify this to the following: There is evidence that even for temperatures colder than -38C, not all ice is produced by homogeneous freezing, a small amount of ice is sometimes formed by heterogeneous nucleation.

1.6 c)Page 9485, lines 7 to 9:

Jensen et al (1998) implied, by was of model sensivity studies only, that heterogeneous nucleation can initiate ice in cold wave clouds. However their paper emphasized homogeneous freezing as the dominant factor...

The text has been changed to clarify this to the following:

Aircraft observations of orographic wave clouds have also implied that ice crystals can be initiated by heterogeneous nucleation Jensen98,Field01, although homogeneous freezing is the dominant process.

ACPD
6, S5262–S5275, 2006

EGU

1.7 d)Page 9486, lines 10 to 13:

Many recent papers allude to the results of Durant and Shaw, but as here, this reference may not be appropriate...

Removed discussion on contact nucleation and reference to Durant and Shaw contact freezing hypothesis. This shortens the introduction.

1.8 e)Pages 9486 to 9487:

Just to be clear, perhaps it should be mentioned that the microphysical model treats the chamber as a single bos with one temperature and one ice particle population. These assumptions should be justified...

The model treats the chamber volume as a single 'box' with one temeparature and one ice population. This is justified firstly because of the measured temperature and humidity homogeneity and secondly, in the time-scales relevant for particle fallout.

Because the AIDA aerosol chamber is within a thermally insulated containment which has heat exchanges to maintain its temperature, and in the bottom of the aerosol chamber a large fan mixes the chamber air, the temperature and humidity is nearly homogeneous. Mohler 2006 and Mohler 2003 describe these measurements. They say that even during strong pumping expansion and cooling, the spatial and short-term temperature variability is less than +-0.3K, except for a few centimetres near the chamber wall.

Ice particle loss through fallout is only relevant once particles have grown to significant size, and during the ice initiation period the largest crystal sizes where a few tens of microns.

ACPD
6, S5262–S5275, 2006

EGU

1.9 3)Section 2 a) Page 9487, lines 20 to 21:

Are ice particles also as well-mixed as the thermal environment of AIDA...

At least for small ice particles in the SID size range (up to a few tens of microns) where fallspeeds are small this should to be true because of the large mixing fan.

1.10 b) Page 9488, line 6:

What are the heat sources in the chamber?

The main heat source within the chamber is the gas mixing fan. This means that initially the gas temperature is slightly higher than the wall temperature, and therefore all experiments start at just below ice saturation.

1.11 c) Page 9488, lines 11 to 13:

Is there a reference for the strength of updrafts in the convective regions of cirrus generating cells? I would guess that 1 to 2 m/s is tops.

The text was not clear, the higher updrafts are for wave clouds where up to 5 m/s (Jensen98,Field01) has been observed.

The text has been changed to the following: The equivalent ascent rates are from 1 m/s to 10 m/s, and enable both orographic wave clouds, where updrafts of up to 5 m/s (Jensen98,Field01) have been observed, and the lesser convective updrafts in cirrus generating cells up to to be simulated.

ACPD
6, S5262–S5275, 2006

EGU

1.12 4)a) Section 4.1, page 9490, lines 9 to 11:

Since all ice crystals probably do not enter the hygrometer tube efficiently, under what circumstances is total water measured in this manner considered reliable at all? This becomes a critical sissue in later discussion and my most significant concern.

As long as the total ice water content is small compared to the total water content the error from not sampling all the larger ice particles is small. For these expansions this is the case because the total number concentration of ice particles is low. Additionally, the important model phase is the ice nucleation time-period and the ice particles are small (but growing). The sampling tube size cut-off is also not an exact value.

Figure 4 shows an expansion where during the nucleation interval, most ice particles are less than 7 microns and also the total condensate mass mixing ratio is an order of magnitude less than the water vapour mixing ratio. Figure 5 shows for the second ice nucleation mode, many ice particles above 7 microns, but the total condensate mixing ratio is still an order of magnitude less than the water vapour. Figure 6 shows lots of large ice particles present (just after the ice nucleation), and the relative humidity will be underestimated. The possible underestimate of relative humidity, however, does not change the conclusions regarding the ability to model the onset of ice nucleation.

1.13 b) Section 4.4, page 9491, lines 6 to 7:

How are ice crystals sampled from the chamber into the SID sampling volume? If there is a sample tube, does flow through this tube impact sampling in any manner? As regards the 1.4 correction factor for presumed spherical ice, how does one know that these particles are in fact ice crystals. Is the baseline value that seems always present subtracted and might this baseline change if unactivated liquid particles are present?

ACPD
6, S5262–S5275, 2006

The SID probe sampled horizontally from near the base of the chamber using an air pump to achieve a flow speed of 5 m/s, which is a sample of 1 cm^3 per second, (described in more detail in Field06). The sample tube was insulated, kept short and had a relatively large cross-section in order to minimise any particle loss.

In Figure 2 the coincidence of the increased asphericity and the broad swath of diameter measurements indicate when ice nucleation and particle growth is occuring. This is well below water saturation so that all growing particles will be ice particles. The relatively few unactivated aerosols were subtracted first. The size calibration for SID is only valid for liquid water drops, the sizing of aerosols is not correct.(The calibration was obtained by comparing the bulk liquid water content from a hot-wire probe to the SID estimate from measurement of liquid water drops in a stratocumulus cloud.)

1.14 5)a) Page 9492, lines 21 to 26:

In line with the last part of the previous comment, I wonder if it is truely valid to state the general assumption that the CCN are only important for warmer temperature expansions. Are unactivated drops possible in sumulations, even at sizes above 1 micron?

All chamber expansions were modelled with this CCN distribution, and were allowed to deliquesce and if the humidity is high enough activate to drops. The SID probe will not necessarily detect all the larger aerosols because the size threshold for detection mentioned in the text is for for liquid drops, not dry aerosol. The effect of including the CCN was only important for expansions 18–21 (listed in Table 1) where liquid drops were observed before any ice particles.

1.15 b) Section 5.2, page 9494, lines 1 to 13:

The method selected to parametrise the wall vapour flux is the method using the MBW

ACPD
6, S5262–S5275, 2006

hygrometer as an accurate measure of the total water content. This raises the critical issue of particle sizes entering the MBW. The paper presently states that the total water is only 100% accurate for 7 micron and smaller ice crystals. How often does that describe the chamber ice distributions during the critical measurement periods? I would expect not very often, except below say -50C and close to nucleation time periods.

Using the MBW hygrometer to estimate the total water content is valid when there are few large particles present or when the total condensate mass mixing ratio is insignificant. This is reasonable since significant errors are expected only when the total condensate mass is large compared with the water vapour mixing ratio. So the presence of large ice particles does not necessarily invalidate this method. In all of the expansions, especially during the ice initiation phase, the total condensate mass mixing ratio is an order of magnitude less than the water vapour mass mixing ratio and therefore any error in not sampling large particles is negligable. While Figure 5 and Figure 6 both show large ice particles and therefore a significant condensate mass mixing ratio, it is mostly after the ice nucleation phase. Here the MBW sampling inefficiency of large ice particles leads to the small discrepency in condensate mass mixing ratio, but this does not affect the conclusion regarding the ability to model the expansion during the ice initiation phase.

1.16 c) Section 5.3, page 9494, lines 18 to 90:

If most of the aerosol particles are larger than 0.1 microns, why are ice particles initiated at 0.1 microns? Does it make a difference if they are initiated at their actual sizes at nucleation. What is assumed regarding the crystal capactitance and water vapour accommodation coefficient?

0.1 micron is the first ice bin in the model, and while initiating into this bin is arbitrary,

ACPD
6, S5262–S5275, 2006

EGU

the actual aerosol subset which acted as IN are also unknown. The aerosol radius indicated by the SID probe is also not valid. Model runs indicate that the choice of 0.1 or 1.0 micron initial size does not significantly change the ice growth during the ice initiation phase. Because the ice crystals are less than a few tens of micron diameter and there is no habit information, the crystals are assumed to be spherical. The capacitance is always assumed to be 1.0 and the ice density is 1.0 g cm^{-3}. The ice deposiition coefficient is 0.24 following Sassen88. The sensitivity to the capacitance and deposition coefficient is shown later in the paper.

The following has been added to the text: The capacitance is always assumed to be 1.0 and the ice density is 1.0 g cm^{-3}. The ice deposiition coefficient is 0.24 following Sassen88. The sensitivity to the capacitance and deposition coefficient is shown later in the paper.

1.17 a) Section 6, page 9495:

Defining nucleation regimes seems an artificial construct that may not have explicit meaning. The figures suggest that nucleation could follow more than two modes and may in fact represent a continuum of behaviour in regimes II and III.

The expansions are divided into four temperature regimes with one example taken from each purely for organistional purposes rather than assuming different ice nucleation modes are occuring in each regime. This follows the Field06 division. The nucleation modes do represent a continuum of behaviour in regimes II and III.

Added to the text the following: This division in regimes is done just for organisational purposes and no assumption is made that different ice nucleation modes occur in each regime, and in fact there is a continuum of behavior in regimes II and III.

ACPD
6, S5262–S5275, 2006

1.18 b) Page 9495, lines 27 to 28:

The condensate mixing ratio depends both on the model microphysics and on the assumption made on the validity of the total water measurement. It seems possible that the agreement with the TDL could be a convoluted and fortuitous result.

The agreement with the TDL could indeed be a fortuitous result, but there is good agreement for all of the expansions listed in Table 1 and this gives confidence in the wall heat and vapour flux correction. If no agreement is possible even when inserting ice crystals at the appropriate time, then no other physically based ice nucleation will agree either.

1.19 c) Page 9497, section 6.2:

This paragraph touts the agreement between model and measured RH,but one wonders if this is expected given that ice crystals larger than 10 microns, not efficiently sampled by the MBW, are already present at the point that the second nucleation mode is observed. Could the actual RH be higher? Also, since the seperation between nucleation modes becomes more evident in secondary expansions, does this imply that the most efficient IN are lost from the total aerosol (sedimentation and other losses)? Has this been considered and does the model account for it al all?

The number of ice crystals is generally smaller in the first nucleation event than the second, so the total condensate mass is still much less than the water vapour mass mixing ratio at the onset of the second mode and therefore any error in not sampling all of the large ice crystals is small. Field06 speculates on the physical cause of the seperation between nucleation modes sometimes observed. Sedimentation and SID sampling will remove some of the more efficient IN from the aerosol sample. For each further expansion, the model starts with fewer aerosols.

ACPD
6, S5262–S5275, 2006

EGU

1.20 d) Page 9498, section 6.3:

Temperature regime III is said to contain two nucleation modes. Yet the first mode is similar in magnitude to the continued rise in the ice signal between modes in temperature regime II. This is what I mean by the artificiality of the multiple 'mode' construct. It looks like it could be a continuum behavior.

For all expansions in temperature regime III, the two ice nucleation modes are not clearly seperated apart from an increase in ice nucleation rate. This increase in nucleation rate is really the only justification for descibing it as two modes.

1.21 e) Page 9498, section 6.4:

The fact that ice formation appears to require water saturation/liquid cloud conditions says little about the exact ice formation mechanism, even whether or not it rules out deposition, as suggested. It seems enough to simply say that water condenstion appears to be involved based on the conditions of activation.

The text has been replaced with the following: The ice particles are produced at some time during this liquid dominated interval, not before which implies that water condensation is required for ice initiation for these expansions.

1.22 f) Pages 9498-9499, section 6.5:

I have a major comment on this section. One must question the validity of the line in Figure 8. indicating the 'critical ice saturation for freezing of ice in aqueous ammonium sulphate drops' for defining heterogeneous freezing conditions...It should probably be stated that this line may not be well anchored for the specific dusts used...The consequence is that I believe one has little justification in concluding that the second ice

ACPD
6, S5262–S5275, 2006

nucleation mode is 'probably' immersion freezing rather than homogeneous freezing. Is there any ability to conclude so when considering the validity of the Zuberi et al. results and within the stated RH uncertainty of measurements in this study? Also is this not a central topic of the Field paper already published?

The conclusion regarding the second nucleation mode is removed from the text. The possible nucleation modes has been discussed in Field06.

Added to the text the following: This line is specific for certain size drops and dust concentrations which are not the same as in these expansions.

1.23 g) Page 9500, section 6.6:

This discusion is unconvincing for distinguishing heterogeneous and homogeneous ice formation mechanisms...

The section which describes model-observation comparisons using homogeneous freezing has been removed. Only the sensitivity to the deposition nucleation remains, but an additional section which shows the sensitivity to ice deposition coefficient and crystal capacitance has been included.

1.24 7) Figures 5 and 6:

Reflecting earlier comments, although there are two distinct modes of ice formation in Figure 5, the signal is not really constant between them. It is no more constant than for the first mode identified in Figure 6.

This two broad swaths of ice particle diameters indicate two modes in Figure 5. While there is also some ice initiation occuring between these modes it is at a much lower rate.

ACPD
6, S5262–S5275, 2006

1.25 8)

The SID size threshold is variuosly stated or used in different places in the text and figure captions as anywhere from 1 to 4 microns.

The actual SID size threshold is around 1 micron diameter. Because of noise on the SID photo-detectors however, the asphericity value does not discriminate between spherical and non-spherical particles when the particle diameter is less than around 3 microns.

The text has been altered so that the SID size threshold is constistently 1 micron. The text has been replaced with the following:

The modeled ice particle concentration, where the diameter is greater than 3 μ m and for all sizes above 1 μ m, are shown by the two thick grey lines. The SID A_f does not discriminate the particle phase for diameters below around 3 μ m.

1.26 9) Figure 8:

The line show for water saturation conditions is not identified. The caption should also note that the dashed line is immersion freezing for micron-sized kaolinite particles in the 20 to 50 micron ammonium suplhate drops. The caption mention of the homogeneous freezing lines hould reiterate that these are for two specific dry particle sizes (0.5 and 5 microns).

Added to caption.

1.27 10) Figure 9:

The caption mentions variations of C but does not say what the mass accommodation

ACPD
6, S5262–S5275, 2006

coefficient was fixed at for ice mass growth calculation.

Added to caption. But removed this figure anyway.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 9483, 2006.