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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The accommodation coefficient of water molecules on ice-cirrus cloud studies at the AIDA simulation chamber" *by* J. Skrotzki* et al.

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Answer to the Reviews on "The accommodation coefficient of water molecules on ice-cirrus cloud studies at the AIDA simulation chamber" by J. Skrotzki et al.

First we want to thank the reviewers for their constructive comments. In the following we respond to the comments and describe respective changes to the manuscript.

Anonymous Referee #1 (Received and published: 26 October 2012)

This is an excellent summary of experiments to determine, as the title indicates, the accommodation coefficient of water. The experiments are unique, the analysis seems correct, and it is well written. The results are important for understanding cirrus clouds.

I have some general comments: - There is a paper (not mine) just out in the last few days about accommodation on liquid water (Miles et al, J Phys Chem A 10.1021/jp3083858) and one of the points it makes is that for consistent results between different research groups attention must be paid to using consistent values for thermodynamic quantities such as the diffusion coefficient. The authors should put their values in the context of this new work.

Answer: Obviously lower values for the diffusion coefficient of water in air must lead to higher accommodation coefficients in our analysis of the ice crystal growths experiments. Therefore, we choose one of the higher values available in the literature to demonstrate that the values for the accommodation coefficient obtained are still quite close to unity, but missed to include results from both model approaches for the same diffusion coefficient. For a better comparability we repeated our analysis with the SIGMA model with the quite common empirical parameterisation from Hall & Pruppacher (1976) since it results in only about 8% lower values compared to a more recent analysis of experimental data like Massmann (1998). However, values calculated with a Lennard-Jones model are still 20-30% lower (Ghosh et al., 2007). The SIGMA model analysis using the smaller diffusion coefficient resulted in accommodation coefficients of one for most of the experiments. In the modified manuscript the figures and the discussion will be updated accordingly.

- The abstract is somewhat overstated compared to the rest of the paper. In particular, I am not sure that the lack of dependence on the supersaturation is an especially strong result – it is weaker than the overall result that the accommodation coefficient is large.

Answer: We wanted to give the reader the information about the lack of a dependence on supersaturation directly in the abstract, so that this information is readily available, even if is not the most important message of the paper. Hence our preference is to keep it in the abstract.

- I believe that the statement that "An alpha-ice value close to unity also suggests that an enhanced growth at few specific ice particle facets does not play a significant role for the ice particle growth. . ." is too strong. There is an alternative that facets do play a role but that migration of water molecules along the ice surface is sufficiently fast that water molecules that do not directly impinge on a growing facet still contribute to growth by sticking and then moving to the growing spot. This gets into issues about a quasi-liquid layer that are under debate and beyond the scope of this manuscript, but some mention could be given.

Answer: We modified this statement to: “An alpha-ice value close to unity also suggests that an enhanced growth at few specific ice particle facets does not significantly influence the observed ice particle growth that is governed by alpha-ice, i.e. up to a particle size of a few microns.”. The important word here is observed, which is specific to these experiments.

- The manuscript should note that the cloud chamber growth studies will select for particles that grow rapidly. That is, if some particles have higher accommodation coefficients than others, the ice growth and vapor depletion will be a function of the fast particles rather than the ones that don’t grow. This isn’t bad – the atmosphere works this way – but it should be noted.

Answer: The phase of our experiments in which ice may grow differently on different ice nuclei (particles) is not analysed in this work. Only the growths of ice particles by water uptake from a size of about 0.6 μm on was observed and analysed.

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Interactive Discussion

Discussion Paper

- I found the discussion of the two models (SIGMA and ACPIM) more difficult to follow than it needs to be. Although everything is there somewhere in the manuscript, it is somewhat scattered. For example, I didn't immediately realize that the two models were about different aspects of what was happening in the chamber rather than two independent models of the same phenomena. In fact the two models use the same growth equation, something that isn't clear until the model details are given on page 13. I think that using the acronyms less and saying rather "ice water content model" and "bin microphysics model" or something like that would be helpful. Looking back, both the abstract and conclusions mention two independent models with the acronyms but in neither place does it describe in general terms what the models are about. So a reader who only looks at the abstract and conclusions would have no idea what was done with the models. Also, the abstract and conclusions using the word "independent" for the models (line 13 in abstract) is somewhat misleading given that that both models make the same approximations in the mass/thermal transport equation – both use equation (6).

Answer: We think it is reasonable to call the model analysis independent since they were done independently by different people with different models fitting to different observables. It is totally reasonable to expect that an interested reader would consult the whole paper to get the details of the modelling, rather than just read the abstract and conclusions.

Anonymous Referee #2 (Received and published: 14 November 2012)

General comment: In the manuscript the authors investigate ice crystal growth at low temperatures in the AIDA chamber. For the evaluation of the crystal growth and especially of the accommodation coefficient two different models are used in order to re-simulate the chamber experiments. The final conclusion is in agreement with theoretical investigations on the variability of the accomodation coefficient from former

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studies. This is an interesting and important contribution to the issue of ice crystal growth due to diffusion of water molecules in air at low temperatures, which might be suitable for publication in ACP. Although the topic of the manuscript is quite interesting, there are crucial errors in the data evaluation. Thus, I have to recommend major revisions, especially of the data investigations by the models before this manuscript can be accepted for publication. In the following I will explain the major problems.

Major points: 1. Use of diffusivity coefficient: The major error in the data investigation results from the use of an inappropriate diffusivity coefficient. In the manuscript the authors use the general approach for binary diffusion of a substance A (water vapour) in a substance B (air), as given by the Chapman-Enskog theory (e.g. Chapman and Cowling, 1970). The crucial parameter in the theory is the collision integral AB between the substances. The authors assume here (at least implicitly, because it is nowhere stated in the text) the hard sphere approximation, i.e. AB = 1. Only with this assumption, they can derive the formula (2) in their text, ending with

Dw = D0*p0/p*(T/T0)**1.5 (1)

where D0 denotes the value at T0 = 273.15K and p0 = 1013.25 hPa. However, this hard sphere assumption is not appropriate for water molecules in air. The standard approach for water vapour is the use of a 6-12-Lennard-Jones potential for molecular interactions. This theory leads to a collision integral AB, which depends on kBT/ïĄęAB with the Boltzmann constant kB and the Lennard-Jones energy ïĄęAB, and even to some corrections for the polar water molecule. In a recent study by Ghosh et al. (2007) the correct use of the collision integral was presented (see their formulae (7) and (8)), leading to a much more complex description of the diffusivity of water vapour in air. Indeed, the qualitative behaviour of the diffusivity as derived by correct Chapman-Enskog theory is very similar to the old but still valid empirical relation by Hall and Pruppacher (1976), as given here:

Dw = D0*p0/p*(T/T0)**1.94 (2)

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The different exponent could lead to differences up to 20% between the two equations. As Sölch and Kärcher (2010) recently pointed out, the difference between the complicated diffusivity given in Ghosh et al. (2007) and the fit by Hall and Pruppacher (1976) is quite unimportant, at least for LES studies. However, for the data evaluation the difference between eq. (1) as used in the manuscript and eq. (2), used as a reference, is quite important. I made some simple calculations on my computer and it seems that for large accomodation coefficients ïĄą the difference in the data evaluation might be quite large, for smaller values of ïĄą the differences are not that crucial. Thus, it seems that the qualitative result of this manuscript will not change, but this must be checked. Nevertheless, I have to insist to redo all model simulations with a correct treatment of diffusivity, no matter if the authors use the approach by Ghosh et al. (2007) or the old but still valid fit by Hall and Pruppacher (1976). The actual approach cannot be justified. Finally, I would like to express my astonishment that although the AIDA group is closely working with groups at Leeds and DLR, who investigated the diffusivity of water vapour for ice crystal growth quite in detail, they are obviously not aware of these developments of the last few years.

Answer: Since the accommodation coefficients from our analysis must be larger for smaller water diffusivity values we choose a large diffusivity in order to demonstrate that the accommodation values are close to unity even for a quite high diffusivity of water in air. As suggested by the reviewer the model simulations were also done with the smaller diffusivity as described in the empirical parameterisation by Hall and Pruppacher (1976). The SIGMA model analysis using the smaller diffusion coefficient resulted in accommodation coefficients of one for most of the experiments. In the modified manuscript the figures and the discussion will be updated accordingly. Use of the smaller diffusivity values from the Lennard-Jones model (Ghosh et al., 2007) would lead to accommodation coefficients even closer to one. In fact, it was an oversight on our part but the ACPIM modelling in the current paper already uses the Hall and Pruppacher equation for the diffusivity, which the referee accepts is reasonable. The reason this wasn't spotted in the manuscript preparation is that they are almost the same and the two model studies, SIGMA and ACPIM, were independently conducted.

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2. Unrealistic ice crystal concentrations and pressure conditions: The AIDA experiments were carried out at quite high pressure conditions, i.e. approximately surface pressure is used, but in combination with low temperatures down to T= 196 K. These combinations seem to be quite artificial and it is not clear to me, if the results would change when realistic conditions (e.g. T = 220K and p= 300 hPa) would be used. The authors should explain, why they can use such unrealistic combinations for their study and how the results would change in realistic setups. Additionally, the ice crystal number concentrations are quite high. As we know from in situ measurements (see e.g. Krämer et al., 2009) ice crystal number concentrations above 10 cm−3 are quite rare. Thus, some explanation for this experimental setup is needed, too.

Answer: A simulation chamber experiment will never perfectly simulate the real atmosphere. We choose the experimental parameters to have highest sensitivity for the accommodation coefficient of water on the growing ice crystals, within the constrains of our experiments. This sensitivity should indeed be higher for lower pressures in the chamber. Test experiments at about 200 hPa showed that the achievable cooling rates and therefore supersaturations and ice crystal number concentrations were smaller compared to 1 atm which unfortunately overcompensated the potential gain expected for lower pressures. Experiments with smaller ice crystal number concentrations and supersaturations would have increased the uncertainty on these parameters and on ice water content significantly.

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3. Errors in measurements due to inhomogeneities in the chamber: In the study, the authors claim that the conditions inside the chamber are mostly homogeneous, such that the local inhomogeneities leads to just small errors. This statement from Möhler et al. (2006) has just been repeated, however, there is only slight evidence from former

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studies (e.g. Möhler et al., 2003) that the inhomogeneities were measured inside the vessel. Since the vessel is quite big and the mixing procedure is carried out with a simple fan, the statement of small temperature errors due to inhomogeneities is hard to believe. The authors should carefully explain, how this error was estimated and additionally investigate the impact of a (probable) higher temperature error (maybe up to $\pm 0.5-1$ K) on their results.

Answer: Temperatures are measured during each experiment at various positions throughout the AIDA simulation chamber proofing the temperature homogeneity of ± 0.3 K within the well mixed part of the chamber (Möhler et al., 2006). Larger deviations can occur mainly in the uppermost section of the vessel due to insufficient mixing. However, since the measurements show the largest part of the chamber to be well mixed the temperature uncertainty can be neglected compared to those parameters used in the uncertainty analysis (cf. Table 4). As there is, according to our knowledge, no evidence that the accommodation could significantly depend on pressure or ice crystal number concentration we are confident that our results are representative for cirrus ice particle growth in the atmosphere. Especially our experiments which cover a factor of five in ice number concentration show no evidence for a significant dependence of alpha on ice number concentration.

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4. Errors due to size distribution: Since it is probably not possible to maintain a mono disperse ice crystal size distribution during the whole experiment, the question arises how an evolving distribution might also influence the growth of the particle. Did the authors measure the size distribution of the ice crystals inside the chamber? If so, do the models represent this evolving size distribution in a sufficient way in order to include possible size effects on the results? This issue should be clarified.

Answer: ACPIM does simulate the evolving size distribution of the particles; hence, with this model we are not assuming a mono disperse ice distribution a-priori, and so

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in principle this could be compared to data. The SIGMA model uses 20 growth bins to simulate the size distribution of the ice particles. However, ice crystal growth was observed with optical particle counters (WELAS, Palas). Since these instruments assume spherical particles to calculate the particle size a quantitative comparison to the modelled ice crystal sizes is unfortunately of limited value. In actual fact the ice distributions simulated by ACPIM are approximated well by a mono-disperse distribution, so this turns out to be a good approximation.

Minor points: 1. Page 24354: What is the difference between parcel models and box models?

Answer: We modified the sentence to: "It is included in the formalisms for cloud ice formation in general circulation models (Kärcher et al., 2006; Kärcher and Lohmann, 2002a, b, 2003; Morrison and Gettelman, 2008) but also in more detailed models (Haag et al., 2003; Cotton et al., 2007; Spichtinger and Gierens, 2009)."

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2. Page 24354: The scheme developed by Spichtinger & Gierens (2009) is also used in 2D/3D models on high resolutions in order of O(100m).

3. page 24359: The use of the electrostatic analogue (after Jeffreys, 1918) is quite problematic. This approach assumes that the shape of ice crystals is smooth, such that no strong changes in the concentrations can occur. Especially, edges and corners are not allowed; however, these surface effects might be important for the investigation of the kinetic uptake coefficient, as pointed out e.g. by Wood et al. (2001). Please explain why this approach is meaningful for your investigations and does not lead to errors in the estimations.

Answer: As explained in section 4 of the manuscript the ice particles had small aspect ratios and are assumed to be spherical. The assumption of spherical ice particles is

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justified, since ice particles smaller than 20 μm in diameter have been observed to be compact and nearly spherical in cirrus cloud measurements (Korolev and Isaac, 2003; Mitchell et al., 2011) as well as in laboratory studies (Abdelmonem et al., 2011; Earle et al., 2010) for temperatures below -35°C. The maximum size to which the ice particles grew stayed below 20 μm. Furthermore this analogy is used within the vast majority of atmospheric models and thus comparisons to data are useful validation. In addition Westbrook and Heymsfield (2011), JAS, 68 2416-2429 validated the capacitance model for a wide range of conditions, showing it to perform reasonably well; although these were for larger crystals for which the deposition coefficient is not important, in contrast to this paper.

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4. page 24360: The INTACC field study (Field et al., 2001) is not representative for cold cirrus, since the measurements were taken at high temperatures, i.e. T > $-$41_C. For a better reference for orographic cirrus clouds, see e.g. the INCA campaign (Gayet et al., 2006).

Answer: We still think we have chosen characteristic experimental conditions but cite also Gayet et al., 2006.

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5. On many occasions in the text, the use of the accomodation coefficient in climate models is mentioned as a motivation or even as final goal. However, climate models should not be first candidate for implementing sophisticated ice physics, since they have major problems in representing clouds in a meaningful physical way because the hierarchy of scales in dynamics is not represented. Maybe a good intermediate step would be the implementation into LES models, cloud resolving models or maybe also regional models.

Answer: We fully agree with this statement and added regional models to the conclusions.

Technical comment on fig. 4: It is hard to distinguish between different curves. Maybe the lines could be thicker.

Answer: We increased the line thickness in Figure 4.

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