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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.

Anonymous Referee #2

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Review of

The accommodation coefficient of water molecules on ice-cirrus cloud studies at the AIDA simulation chamber

by Skrotzki et al.

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 accomodation 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 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 Ω_{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. $\Omega_{AB} = 1$. Only with this assumption, they can derive the formula (2) in their text, ending with

$$D_w = D_0 \frac{p_0}{p} \left(\frac{T}{T_0} \right)^{\frac{3}{2}} \tag{1}$$

   where D_0 denotes the value at $T_0 = 273.15\,\mathrm{K}$ and $p_0 = 1013.25\,\mathrm{hPa}$. 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 Ω_{AB}, which depends on $\frac{k_B T}{\epsilon_{AB}}$ with the Boltzmann constant k_B and the Lennard-Jones energy

ACPD
12, C9322–C9328, 2012

ϵ_{AB}, 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:

$$D_w = D_0 \frac{p_0}{p} \left(\frac{T}{T_0} \right)^{1.94} . \quad (2)$$

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 α the difference in the data evaluation might be quite large, for smaller values of α 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.

2. Unrealistic ice crystal concentrations and pressure conditions:

ACPD
12, C9322–C9328, 2012

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 $T \sim 196\,\mathrm{K}$. These combinations seem to be quite artificial and it is not clear to me, if the results would change when realistic conditions (e.g. $T \sim 220\,\mathrm{K}$ and $p \sim 300\,\mathrm{hPa}$) 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 $10\,\mathrm{cm}^{-3}$ are quite rare. Thus, some explanation for this experimental setup is needed, too.

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 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 $\pm 0.5 - 1\,\mathrm{K}$) on their results.

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.

ACPD
12, C9322–C9328, 2012

Minor points:

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

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

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ACPD

12, C9322–C9328, 2012

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ACPD

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