The Accommodation Coefficient of Water Molecules on Ice and its Role for Cirrus Clouds

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I. MOTIVATION

One of the parameters governing the growth of ice crystals in cirrus clouds is the accommodation coefficient of water molecules on ice. It describes the sticking probability of water molecules colliding with the surface of an ice particle. It is relevant for the ice growth in the kinetic regime, i.e., for sub-micron ice crystals.

However, the magnitude of α is still unclear. Experimental results vary between unity and values below 0.01 [Haynes et al., 1992].

Model calculations suggest that values for α between 0.1 and 1 do not have a significant impact on ice growth in cirrus clouds. Lower values however could explain the observation of unexpectedly high ice number concentrations and supersaturations within cirrus clouds [Giersen et al., 2003; Lohmann et al., 2008].

II. THEORY OF ICE GROWTH

For spherical ice crystals, the mass increase per time can be described by the following formula [Pruppacher and Klett, 1997]:

\[
\frac{dn}{dt} = \frac{4 \pi \varrho (S - 1)}{r \cdot \rho_i \cdot \rho_m + LH}
\]

where \( r \) is the ice particle radius, \( S \) the saturation ratio, and \( \rho_i \), \( \rho_m \), and \( \rho_H \) the gas constant, absolute temperature, and molar weight of water, respectively. LH describes the growth-impeding effect of the latent heat of deposition.

The accommodation coefficient α enters in the modified diffusivity of water vapor in air:

\[
D_{\alpha}^{*} = \frac{D}{1 + \frac{\alpha \cdot \rho \cdot T}{\rho_i \cdot T}}
\]

where \( \Delta \) is the vapor jump distance.

III. AIDA CIRRUS EXPERIMENTS

Dedicated experiments examining the ice crystal growth for deposition nucleation in the temperature range from 190 K to 230 K were carried out at the cloud simulation chamber AIDA [Möhler et al., 2003].

As aerosols, hematite particles and graphite-spark generator (GSG) soot were used.

Example experiment of the AIDA studies
- Dotted line: start of the experiment, i.e. start of pumping
- Dashed-dotted line: ice onset

Water vapor and total water are measured by two tunable diode laser (TDL) hygrometers [Fahey et al., 2009]. From the difference of these two measurements, the ice water content is derived.

The ice number concentration \( N_i \) is measured by an optical particle counter (WELAS).

IV. MODELING

Two models are used to derive the accommodation coefficient α from experimental data:

- The Aerosol-Cloud-Precipitation Interaction Model (ACPIM) [Connolly et al., 2009]
- The Simple Ice Growth Model (SIGMA)

V. UNCERTAINTY ESTIMATE

Accuracies of the experimental data sets used for a Monte Carlo (MC) uncertainty analysis:

<table>
<thead>
<tr>
<th>Source</th>
<th>Accuracy</th>
<th>ACPIM</th>
<th>SIGMA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total water</td>
<td>5%</td>
<td>5%</td>
<td>5%</td>
</tr>
<tr>
<td>Ice number concentration</td>
<td>20%</td>
<td>20%</td>
<td>20%</td>
</tr>
<tr>
<td>Aerosol number concentration</td>
<td>10%</td>
<td>10%</td>
<td>10%</td>
</tr>
<tr>
<td>Aerosol size distribution</td>
<td>10%</td>
<td>10%</td>
<td>10%</td>
</tr>
<tr>
<td>Temperature</td>
<td>0.3 K</td>
<td>0.3 K</td>
<td>0.3 K</td>
</tr>
</tbody>
</table>

VI. RESULTS

Temperature dependent values of α for a set of 16 individual AIDA experiments. The error bars are obtained from the MC uncertainty estimate described previously.

- Both models in good agreement with each other with all best-fit points in the range 0.1-1
- Lower values than 0.1 excluded by the error estimate, independent of the aerosol type (SIGMA)
- Temperature averaged value \( \alpha = 0.9 \pm 0.1 \) (SIGMA)

VII. CONCLUSIONS

- The ACPIM and the SIGMA model are in good agreement despite their different approaches in determining α.
- In the temperature range from 190 K to 230 K, values between 0.1 and 1 for α are preferred by both models.
- The uncertainty analysis excludes α-values below 0.1 with a temperature averaged value \( \alpha = 0.9 \pm 0.1 \) (SIGMA).
- These results suggest that α does not have a significant impact on ice growth in cirrus clouds.

ACKNOWLEDGEMENTS

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