

***Interactive comment on* “The impact of ice uptake of nitric acid on atmospheric chemistry” by R. von Kuhlmann and M. G. Lawrence**

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We would like to thank the referee for his constructive comments on the manuscript. We added more information and/or discussion on each of the points raised and considered all small comments.

Ullerstam et al. studies:

We were indeed unaware of the recent studies by [Ullerstam et al.(2005)] and [Ullerstam and Abbatt(2005)]. A discussion of the impact of these new measurements has been added. We compared calculated fractional coverages using their measured equilibrium constant and with the non-dissociative langmuir uptake expression and

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found that the expression used in our study leads to significantly lower surface coverages, especially at higher HNO_3 pressures.

Estimating large scale impacts of enhanced uptake under growing ice conditions as studied by [Ullerstam and Abbatt(2005)] poses very high demands on the microphysical scheme in chemical transport or global circulation models, which are currently not met. Detailed simulations with cloud resolving models and box-models will be needed to gain more insight. We extended the discussion to include the new experimental findings. Overall, from these studies we find more indications that our standard “LANGM” simulation probably represents a lower limit estimation.

Process oriented analysis:

We have tried using the temperature as a selection criterium for the scatter plots instead of height information. However, this did not lead to clearer results. There is still a similar scatter between model and observations, with overestimate of the model for low measured values and underestimation of the model of high measured values. Using the ice surface as the selection criterion is more difficult, since meaningful ice surface information is not straightforward to extracted from the model. This is because the non-linear processes only take place in the cloudy fractions of the grid cells, while only grid-averaged information can be integrated to time averages and written out. Additionally, it is not clear whether the model averaged predicted ice cloud amounts matches well for the particular times of the campaigns. This kind of analysis would, however, be useful and advised if a campaign-based comparison to model results is done. In this study we tried to rely on the ensemble of campaigns, which gives us a stronger statistical basis. We include a statement recommending more detailed case studies utilizing a cloud resolving model.

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Initial uptake coefficient:

We have discussed our assumption of an equilibrium coverage in Section 3.3, however, without mentioning the effect of a potentially small initial uptake coefficient. This issue is now also addressed in the discussion. Judging the effect of a potentially lower initial uptake coefficient compared to our instantaneous equilibrium approach depends on the lifetime of cirrus clouds, or, more precisely, on the lifetime and fate of the individual particles, which will require future studies with more detailed models and data. If one assumes lifetimes of cirrus on the order of the model timestep of 0.5 hours, however, we estimate that using an equilibrium approach could result in overestimated uptake for low density ice clouds (low surface area density) if the initial uptake coefficient was on the order of 0.01 or smaller.

The small comments have also been considered. There is no clear pattern on the altitude range or season in the model's HNO₃ underestimate, except that the underestimate seems to be largest in the remote regions like the central Pacific. This information is now added for clarity. We also reworded the explanation of differences between the study by [Liao et al.(2003)] and our study. Indeed, the treatment of HNO₃-ice interaction is much more sophisticated in this study.

References

[Liao et al.(2003)] Liao, H., Adams, P. J., Chung, S. H., Seinfeld, J. H., Mickley, L. J., and Jacob, D. J.: Interactions between tropospheric chemistry and aerosols in a unified general circulation model, *J. Geophys. Res.*, 108, 4001, doi:10.1029/2001JD001260, 2003.

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[Ullerstam and Abbatt(2005)] Ullerstam, M. and Abbatt, J. P. D.: Burial of gas-phase HNO_3 by growing ice surfaces under tropospheric conditions, *Phys. Chem. Chem. Phys.*, 7, 3596–3600, 2005.

[Ullerstam et al.(2005)] Ullerstam, M., Thornberry, T., and Abbatt, J. P. D.: Uptake of gas-phase nitric acid to ice at low partial pressures: evidence for unsaturated surface coverage, *Faraday Discuss.*, 130, 211–226, doi: 10.1039/b41718f, 2005.

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