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Interactive comment on "A Parameterization of Heterogeneous Hydrolysis of N$_2$O$_5$ for 3-D Atmospheric Modelling: Improvement of Particulate Nitrate Prediction" *by* Ying Chen et al.

Anonymous Referee #2

Received and published: 19 May 2017

This paper describes a new parametrization for the N2O5 hydrolysis which is dependent on T, RH and aerosol composition. The development of such a parameterization for N2O5 hydrolysis for a model framework that does not explicitly track aerosol surface area could be of interest to the community. However, the paper presented here has several serious shortcomings and errors, detailed below, so that I cannot recommend it for publication.

Major concerns:

1. The paper completely misrepresents the parameterization from Riemer et al. (2003): It is stated by the authors that the particle surface area in Riemer et al. (2003) was set to a constant value of 600 μm2 cm-3. However, this constant value was used only

for box model runs. In all other simulations (KAMM/DRAIS and EURAD) a constant value was not used, instead the mass, the number and thus the particle surface area were calculated with the modal aerosol model MADE and were highly variable. In Figures 8a and 11a of Riemer et al. (2003), examples of these variable horizontal distributions of the aerosol surface area density are depicted. It is clearly stated in Riemer et al. (2003) that even the corrected formula of Chang et al. (1987) shows a big difference in comparison to the more-complete parameterization that takes into account the dependence on aerosol surface area concentration (Sec. 4.2 in Riemer et al., 2003). The comparison with the so-called Riemer03 parametrization and an assumption of a reaction probability of 0.1 (Dentener and Crutzen, 1993) is not very helpful because numerous papers (e.g. Davis et al. (2008)) show that 0.1 is seen as an upper limit of gamma .

2. Chang et al. (1987) calculated the rate constant by the following equation: Eq. 17, Chang et al. (1987).

Whereas in this paper: Eq. 2 and 3, this study is written.

It is not clear whether this is an error in the paper, or also in the parameterization itself. It is not clear which formulation was the basis for the presented simulations. The authors need to check this because using the equation written in the paper gives values that are orders of magnitude different.

3. In equation 5, there is no explanation as to why the expression for gammaN2O5 is divided by a factor of 0.1. This leaves me with the impression that the factors are introduced to yield the best fit with the nitrate observations, which limits the general applicability of the parameterization to other domains and conditions. Similarly, there is a division by 600 in equation 4 which is also not explained. Furthermore, the units of fs are unclear. Based on the units stated in the text below equation 4, fs appears to have units of m-1 , but the factor should be unitless.

4. The reference to Chang et al. (2016) is missing. They also combined the Davis et

al. (2008) parametrization with the coating parameterization of Riemer et al. (2009).

Chang, W. L., S.S. Brown, J. Stutz, A.M. Middlebrook, R. Bahreini, N.L. Wagner, W.P. Dubé, I.B. Pollack, T. B. Ryerson, and N. Riemer (2016), Evaluating N2O5 heterogeneous hydrolysis parameterizations for CalNex 2010, J. Geophys. Res. Atmos., 121, 5051–5070,doi:10.1002/2015JD024737.