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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Sensitivity of a global model to the uptake of $N_2O_5$ by tropospheric aerosol" *by* H. L. Macintyre and M. J. Evans

**Anonymous Referee #2**

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This short paper presents a conceptual overview of the effect of revisions in the value for heterogeneous uptake coefficients for N2O5 on global models of NOx, O3 and OH. Several previous modeling papers have considered the effect of switching this chemical reaction on or off for fixed values of the heterogeneous uptake efficiency, and a few have tried to incorporate aerosol composition-dependent uptake coefficients in regional and global models. This paper is a unique contribution since it explores variation in the uptake coefficient over its entire plausible range, from very small to very high values. Although it clearly simplifies the picture considerably by fixing the value for each global simulation and neglecting any regional or seasonal variability, it does demonstrate the range over which global NOx, O3 and OH are sensitive to this reaction. The conclusion that the range of uptake coefficients determined from more recent laboratory and field studies is exactly the range over which these species are

most sensitive is compelling. For this reason I recommend publication in ACP.

A few comments that the authors should consider:

The paper considers competition between daytime photochemical conversion of NOx to HNO3 via OH and nighttime oxidation of NOx in O3 to produce N2O5. While this competition is certainly relevant – and likely an important metric in a global scale model – it neglects competition between nighttime heterogeneous reactions of N2O5 and those of the nitrate radical, NO3. In fact, no details are actually given about how NO3 chemistry is handled here. One obvious impact, for example, of changing N2O5 heterogeneous uptake would be the change in formation of other NOx species, such as organic nitrates, from NO3 oxidation reactions. Such chemistry would be less important in winter, when the largest effects due to N2O5 heterogeneous reaction are inferred. The authors should include some comment about variability in NO3 chemistry and how this is handled in their model.

A related issue is the branching between HNO3 and ClNO2 formation in N2O5 hydrolysis. The latter is a relatively recent discovery – at least in terms of its potential importance in the troposphere. It is clear that this branching has not been considered in this paper. The authors should explicitly state this assumption.

Lastly, the effect of model resolution is not considered in this global-scale model study. Anthropogenic NOx emissions tend to be highly localized, and the chemistry that leads to N2O5 formation is quadratic in NO2 (i.e., two NO2 required to form each N2O5). Therefore, inputs of average NOx into a model grid cell may not represent how NOx and N2O5 actually evolve overnight in the near source regions where this chemistry is likely to have the greatest impact. Such fine-scale interactions are clearly not the goal of this paper, which is intended to simulate global scale effects. However, some comment on the potential for these kinds of interactions to affect the larger scale conclusions would be appropriate.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 13557, 2010.