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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 #1

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This paper presents a focused study of the sensitivity of GEOS-Chem predicted quantities to the uptake of N2O5 to aerosol. GEOS-Chem is a state of the art global chemical transport model that is widely used. Moreover, N2O5-aerosol chemistry remains highly uncertain and poorly constrained. Thus, the results of this study are likely to be useful guides for development and improvement of other models, and to help focus experimentalists to further constrain this chemistry in regions and time periods it is most likely important. The paper is well written, and the conclusions are strong and supported by the simulations. In particular, the authors do a commendable job highlighting the implications of high sensitivity, or lack thereof, to specific processes or problems of current interest. While the paper is a little on the lean side, I do recommend publication in ACP for the above reasons. I have a few comments and suggestions that the authors should address.

Larger Issues:

I accept the need to simplify the situation by spanning a range in gamma values that are constant across aerosol types. However, the authors should discuss how such a choice impacts their interpretations of the regional sensitivities where possible, as certain regions might be more heavily affected by one of the aerosol types for which the large range in gammas may not be appropriate.

It should be noted that much of the justification for the large range in gammas (especially the very low values) used in the study comes partly from some field observations limited exclusively to NH summer time. That is, very little observational constraints exist during the period and region for which the model is most sensitive to the value of gamma (NH winter). Laboratory studies at lower temperatures on relevant aerosol compositions are even fewer. There are some constraints from isotopic studies in the field that did not get mentioned here, e.g. Alexander, et al ACP 2010.

The role of model resolution in biasing the conclusions should be discussed. The simulations were conducted at 4x5 degree resolution, which is rather coarse. N2O5 formation (NO2+ O3), thermal and photochemical stability via the N2O5/NO3 ratio depend strongly on the NOx concentration. Moreover, as noted, the N2O5 loss rate depends as much on aerosol surface area as on the gamma – NOx and aerosol plumes will not be well mixed on the 4x5 degree scale. To what extent would or could the conclusions drawn about the sensitivity to gamma change if the model had been run at higher resolution? Perhaps this question is more appropriate for regional air quality models, but global models running simulations of past and future conditions are even moving to higher resolution.

The importance of NO3 was mentioned only to note that N2O5 chemistry was confined to nighttime. However, NO3-VOC reactions at night likely influence the sensitivity of the model to the value of gamma, especially in summer but also year round in the tropics. This issue should be briefly discussed in the context of the completeness of the NO3

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mechanism in GEOS-Chem.

Smaller Issues:

Abstract: Line 23, This sentence is a little awkward. I came to understand it only after having read the paper and inspected Figure 3. Perhaps just note the main conclusion of this section which was that for studies of coupled aerosol and chemistry changes over time, the results may be sensitive to the choice of gamma.

Pg 13560, Reaction R2 – in light of recent findings the descriptions of products from this reaction should be amended to include nitryl chloride (ClNO2), though it is apparent that the modeling studies presented herein do not include it. Presumably its effects could be inferred as the yield of ClNO2 is to first order a lowering of the gamma to form 2HNO3.

Pg 13560 line 16 – To my knowledge, Mozurkewich and Calvert, while the first aerosol flow tube measurement, did not specifically focused on cold sulfuric acid aerosol. Fried, et al 1994 is probably a better reference for that statement. That said Mozurkewich and Calvert should still be referenced as it is relevant to the overall argument.

Pg 13564 line 21 – Please clarify that the curves shown in Figure 3 are derived from error function fits to the model output shown in Figure 2, and not from additional model runs.

Pg 13564 last line going onto next page – I agree with this statement for global O3 and OH, but the sensitivity of global tropospheric NOx seems change at most by a factor of 2 or so from gamma = 0.1 to 0.001 (in fact it changes very little between those two end points).

Figure 3 caption: change to, “Impact of a 10% reduction in the product gamma_N2O5*A, where A is aerosol surface area density."

References

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