

Interactive comment on “Ozone Impacts of Gas-Aerosol Uptake in Global Chemistry Transport Models” by Scarlet Stadtler et al.

Anonymous Referee #1

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The manuscript "Ozone Impacts of Gas-Aerosol Uptake in Global Chemistry Transport Models" by Stadtler et al. deals with the importance of heterogeneous chemical uptake of several inorganic compounds for the tropospheric chemical budget of ozone. To investigate the relevance of the uptake, parameterised reaction rates producing a loss term to the budget of N_2O_5 , NO_3 , NO_2 , O_3 , HNO_3 and HO_2 are implemented in two chemistry models, i.e. the EMEP chemical transport model and the ECHAM-HAMMOZ CCM. The analysis of the results is based on sensitivity tests, each neglecting one of the reactions. The dominating heterogeneous reaction for the tropospheric O_3 burden is the (well known) heterogeneous uptake of N_2O_5 , whereas the other reactions show a minor relevance. Given the large temporal variability of the ozone measurements at the given stations, even the heterogeneous N_2O_5 uptake is hardly significant, whereas

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for the other reactions a statistical significance cannot be stated. For some reactions the two models even give results of the opposite sign.

The paper is well written and the study is reasonable and should be published in ACP after addressing the comments below. However, as the main finding is the importance of the N_2O_5 uptake, which is well known since more than 20 years, the new findings from this manuscript are relatively limited.

Major comments:

1) The neglect of direct nitrate formation in HAMMOZ might not only lead to a shift between chloride release and nitrate formation, but might affect also the size distribution and therefore the surface area of the aerosol. To which degree does the SAD change in the EMEP simulations in case of the neglect of N_2O_5 or HNO_3 uptake? Can you quantify the total loss of these compounds in the EMEP simulations versus the HAMMOZ runs?

2) To which degree do you think that reactive nitrogen chemistry as in "Reactive nitrogen chemistry in aerosol water as a source of sulfate during haze events in China": Yafang Cheng, Guangjie Zheng, Chao Wei, Qing Mu, Bo Zheng, Zhibin Wang, Meng Gao, Qiang Zhang, Kebin He, Gregory Carmichael, Ulrich Pöschl, Hang Su Science Advances, 21 Dec. 2016; DOI: 10.1126/sciadv.1601530' is relevant on the global scale in your simulations?

3) The interaction of the individual uptake reactions might lead to a masking of the direct effects of the reactions by chemical interactions. Why did you not choose to analyse individual reactions, but always the sum of reactions neglecting only one of them. Would a method such as a Factor Separation Method (see Stein, U. and P. Alpert, 1993: Factor Separation in Numerical Simulations. J. Atmos. Sci., 50, 2107–2115, https://doi.org/10.1175/1520-0469(1993)050<2107:FSINS>2.0.CO;2) not be more suitable to address your scientific question (given the extra computation time is available).

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4) To which degree might your results depend on the selected year. Would you expect substantial inter-annual variability based on the variability in both the aerosol surface and the constituents?

5) How sensitive are your results to the selected γ values for the individual compounds? Are the values important or is the uptake anyhow mostly determined by the available aerosol surface and the availability of the constituents?

6) Why did you only consider O_3 uptake on dust and not on liquid aerosol particles, where it can contribute to "in-aerosol" sulphate formation by oxidation of dissolved SO_2 . Even though the solubility of O_3 is quite low, the effective uptake is determined by the reaction rate.

7) What is the tropospheric CH_4 lifetime in your simulations? Due to HO_2 uptake, the oxidation capacity might be reduced, but due to HONO formation and subsequent photolysis otherwise affected. Is this similar in both models? Is there a substantial impact on the oxidation capacity which is not that obvious in the well buffered compound O_3 ?

Technical comments:

1) Fig. 6 to 8: The grey shaded area is very difficult to see. Please think about a better visualisation of the observations.

2) page 7, last line: citation is missing/wrong (ibid)

3) page 13, line 7 (wrong line number): citation is missing/wrong (ibid) "transport model, GEOS-Chem (ibid),....."

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