

Interactive comment on “Chemistry of hydrogen oxide radicals (HO_x) in the Arctic troposphere in spring” by J. Mao et al.

Anonymous Referee #2

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General comments: Mao et al. investigate HO_x chemistry in the Arctic troposphere by comparing the HO_x concentrations observed by an LIF instrument aboard DC-8 and those simulated by a chemical transport model GEOS-Chem for the ARCTAS field campaign. Their major finding was that the HO₂ (and OH) concentrations were significantly overpredicted by the model. They attributed this to the heterogeneous loss of HO₂ on the aerosol surfaces. The oxidizing capacity in the Arctic troposphere has been less explored than other regions and therefore this manuscript provides new knowledge, within the scope of Atmospheric Chemistry and Physics. I agree that the heterogeneous loss process is one of the likely causes for the HO_x concentrations lower than predicted. However, more information should be provided to convince the readers. For example, averaged vertical profiles of temperature, relative humidity, and aerosol surface density (modeled one) should be presented. Any information about the phase of

C2363

the particles (liquid or solid), which is critical for the conclusion, should be sought and provided. It is also important to see that the J values are well simulated by the model even when the cloud effect is included. Overall, I suggest that the paper should be published after responding to the comments above and taking into account the following minor comments.

Specific comments:

1. Abstract. As a loss process, HO₂ conversion to HSO₅- is too much speculative to be mentioned in the Abstract.
2. page 6958, line 12. radicals
3. page 6958, line 20. Is the sentence "HO_x radicals originate from water vapor" appropriate? In (R1) and (R2), 50% of oxygen atoms of OH radicals originate from O₃, not from H₂O.
4. page 6959, line 2. By the radical-radical reactions that return water vapor, do the authors mean a single reaction HO₂+OH reaction? Or any other? In relation, at line 11, which process do the authors indicate by "against conversion to water vapor?"
5. page 6962, lines 24. How do the authors assume the vertical distributions of the total ozone columns? How well did the calculated J values reproduce the observed ones (including influence from clouds)?
6. Page 6963, from line 23. With respect to wet deposition, was there any significant bias between observed and modeled rainfall distributions/amounts?
7. Page 6965. What is the expected uncertainty range for HO₂ in the GEOS-Chem model caused by the fact that the J values, H₂O, O₃, and HCHO concentrations were not constrained by the observations? Is the disagreement in the HO₂ concentrations by a factor of ~2 under discussion beyond this uncertainty?
8. Page 6968. Is there any observational evidence suggesting that the aerosol parti-

C2364

cles are wet droplets, under the cold temperature conditions? Such as particle shape analysis, humidogram, or H₂O signal from the Aerodyne Aerosol Mass Spectrometer?

9. Page 6969, line 6. ranging

10. Page 6969, from line 19. Can these successive processes provide a first-order loss for HO₂ where an uptake coefficient can be theoretically defined? Is a reaction step (R7) necessary? Even without (R7), HO₂ can be lost via conversion to SO₅- (by R6), then to HSO₅- (oxidation by O₂-, HCOO-, and HSO₃- as written in text) and finally to sulfate by R8?

11. Page 6972, line 12. Is ozone "production" expected in the arctic troposphere? If not (namely ozone loss regime), it is inappropriate to mention "NO_x-limited production" here.

12. page 6972, lines 25-26. Is the lower troposphere defined with an altitude range 0-2km?

13. Figures 5 and 6. Is the radical loss via OH + NMHCs reactions giving organic nitrates negligible?

14. page 6974 and Figure 7. Is it better to provide two separate diagrams for lower troposphere and above region, because the importance of the heterogeneous loss is different?

15. When the HO₂ loss is governed by the heterogeneous loss of it whose rate is linear with [HO₂] (not by the self reaction whose rate is quadratic with [HO₂]), HO₂ concentration should have more linear dependency against the HO_x production rate. Can the authors show this to test the conclusion?

16. Figure 1. Can the authors add averaged vertical profiles of temperature, relative humidity, aerosol surface density (modeled one), the applied gamma value, and J values (observed and modeled)?

C2365

17. Can the loss of HO_x onto the cloud droplets be also important, in addition to the loss on the aerosol particles?

18. Can the introduction of the heterogeneous loss process for HO₂ into the global chemical transport model damage any past comparisons of the observed/modeled HO_x radicals and thus will we need reanalysis of them? Or is the heterogeneous loss important only in the Arctic troposphere?

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 6955, 2010.

C2366