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Atmospheric Chemistry and Physics Discussions

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 HOx chemistry in the Arctic troposphere by comparing the HOx 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 HOx 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

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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 "HOx 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 $\sim$2 under discussion beyond this uncertainty?

8. Page 6968. Is there any observational evidence suggesting that the aerosol parti-

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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 "NOx-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 HOx 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)?

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17. Can the loss of HOx 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 HOx 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.