Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-863-RC1, 2016

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global sensitivity analysis of the GEOS-Chem chemical transport model: Ozone and hydrogen oxides during ARCTAS (2008)" *by* Kenneth E. Christian et al.

Anonymous Referee #1

Received and published: 13 December 2016

This paper describes a global sensitivity analysis of Arctic tropospheric oxidants (OH, HO2, ozone) in an ensemble of GEOS-Chem global chemical transport model simulations to uncertainties in emissions, kinetic parameters, and meteorological parameters. Analysis of model response to 50 different parameter uncertainties is presented, and the chief drivers of model uncertainty in ozone and HOx concentrations sampled along DC-8 flight tracks from the ARCTAS 2008 experiments are discussed. The paper identifies a handful of key emission sources, gas-phase kinetic rates, and heterogeneous processes responsible for driving the majority of model ensemble variance. In particular, uncertainty in the HO2 aerosol uptake coefficient is identified as a dominant driver of model uncertainty in HOx, particularly in spring. The paper is well written,

well structured, and has clear figures. The material is a suitable topic for ACP, and will be of wide general interest to those concerned with Arctic atmospheric chemistry and climate, as well as those interested in drivers of model uncertainty in tropospheric chemistry. While the conclusions are interesting and novel, I would recommend that a number of points are addressed before considering accepting the article for publication in ACP.

General comments

1) It would be useful to discuss how model-dependent the large ozone sensitivity to uncertainty in the NO2 + OH reaction rate may be. Previous studies comparing GEOS-Chem with ARCTAS observations have shown that the model displays a large over-prediction of HNO3 and a large under-prediction of PAN in the Arctic troposphere (Figs. 18 & 16 Emmons et al., (2015); Figs. 3 and 4 Arnold et al., 2015). To what extent is the sensitivity to the HNO3 production rate a reflection of the propensity for GEOS-Chem to produce large amounts of HNO3? i.e. is the sink for NO2 through formation of HNO3 (and therefore sensitivity to uncertainty in its rate) realistic? Does this version of the model include the NOy chemistry updates from Fischer et al., (2014) which greatly improved the simulation of NOy chemistry in GEOS-Chem? The authors should include some reference to these past studies comparing GEOS-Chem with ARCTAS data and other models in the discussion, and comment on how the model Arctic NOy budget compares with observations and implications for the inferred sensitivity to the kinetic uncertainties.

2) The large response to soil NOx emissions is a surprising and novel result, and also warrants further discussion. Given the high vertical stability of the Arctic troposphere, there is strong isolation of the free mid / upper troposphere from emissions and processes in high latitude / Arctic boundary layer, and air tends to be transported into the mid/upper troposphere from lower latitudes (e.g. Stohl, 2006, Wespes et al., 2012). Therefore ozone sensitivity at altitudes in the mid and upper troposphere is presumably driven by response to uncertainty in soil NOx emissions from lower latitudes, and

its impacts on ozone chemistry during uplift and long-range transport into the Arctic? It would be useful to expand on this in the manuscript, such that the reader has a better idea of what drives this sensitivity. A factor 3 uncertainty is assumed for these emissions based on Jaegle et al., (2005). Is this the most appropriate and recent reference for framing this uncertainty? Given the importance of this uncertainty for ozone in the N American Arctic, it would be helpful to discuss more widely estimates of the reliability (uncertainty in) the soil emissions if other studies are available and how robust the factor 3 estimate may be.

3) To what extent is the large HOx response to gamma_HO2 a reflection of the large uncertainty range implemented (factor 3)? It would be useful to show what actual range of gamma values this corresponds to. The authors show that the ensemble members with lower gamma values best match profile observations of HO2. How do these gamma_HO2 values compare with those used in previous GEOS-Chem studies? What are the implications for model comparisons with high latitude CO values, which in previous studies have been improved by implementing different formulations of aerosol uptake of HO2 (e.g. Mao et al., 2013)? How does the choice of product (H2O2 or H2O) affect comparisons with CO and ozone? It would be useful to discuss this, since underestimation of CO at high latitudes in CTMs is a persistent problem (e.g. Emmons et al., 2015).

4) It should be made clear in the abstract and the methodology that this analysis only provides information on drivers of model response to uncertainties in air masses sampled during ARCTAS. It cannot be assumed that this is representative of the whole Arctic unless this can be shown explicitly. Figure 2 shows a good spread of aircraft observations across altitudes, but the flights still only sample the N American Arctic on specific days, when there are certain specific air mass origins.

Specific / minor comments

Page 1, Line 1: “oxidation capability” change to “oxidation capacity”

Page 1, Line 19/20: “Increasing oil and gas exploration and extraction, coupled with summertime shipping lanes through the region will make air pollution worse”. This statement needs a reference.

Page 2, line 5: “.. model shortcomings are usually attributed to errors in the chemical reaction rates, emissions, or meteorology (e.g., Wild and Prather, 2006)”. The cited study is specifically about effects of model resolution? Please cite examples to back up the specific reasons you list.

Page 2, Line 10: Omit semi-colon.

Page 2, line 13: “two more input factors” should be “two or more input factors”?

Page 4, line 3: Better phrased as: “We note in the following section exceptions to this. . .”

Page 4, line 10: The Jaegle et al., (2005) reference is cited for estimating uncertainty in biomass burning emissions. The GFED 3 emissions are used, so is there a more recent and appropriate estimate of uncertainty specifically for these emissions? I am not suggesting re-running the ensemble, but again (as with soil NOx - see point above) framing the choice of factor 3 uncertainty against any other estimates would be helpful.

Page 8, Line 11: OH interferences being negligible in Arctic free troposphere. Probably correct in general, but what about in biomass plumes during ARCTAS-B?

Page 8, Sec. 2.4: The detail on the specific GEOS-Chem code for aircraft flight track interpolation seems unnecessary. Instead just describe what this does.

Page 8, line 26: I am not sure you can claim that the flights give a “representative view of the Arctic troposphere”. See my general point (4) above.

Page 8, line 27: You shouldn’t refer to Fig. 6 before you have referred to Figs. 3,4,5. Consider re-ordering / re-numbering the figures.

Page 11, line 17: Should be “are shown in Figure 7”.

Page 13, line 7-9: Mischaracterisation of advection from mid-latitudes effect on ozone. Has this been discussed in the main paper text? Previous multi-model studies have also shown low profile springtime ozone in the Arctic in GEOS-Chem, but no similar underestimation of ozone in other models driven by GEOS-5 meteorological data (e.g. Emmons et al., 2015, Figs. 16 & 17). It therefore seems unlikely to be related to advection errors. Please expand this discussion in light of this past work.

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