

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.

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We thank the referee for their thorough review and helpful comments. Below are our responses to the referee's comments (*italics*).

1. *It would be useful to discuss how model-dependent the large ozone sensitivity to uncertainty in the $\text{NO}_2 + \text{OH}$ reaction rate may be. Previous studies comparing GEOSChem with ARCTAS observations have shown that the model displays a large overprediction of HNO_3 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 HNO_3 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 NO_y chemistry updates from Fischer et al., (2014) which greatly improved the simulation of NO_y 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 NO_y budget compares with observations and implications for the inferred sensitivity to the kinetic uncertainties.

Response: In our model runs we likewise see similar over-prediction of HNO₃ and under-prediction of PAN in our domain. As noted, this isn't a novel result with GEOS-Chem but should be mentioned for those readers unfamiliar with the model. We've edited the manuscript to make note of this (P10 L10-15). Even with this HNO₃ overprediction, I'm hesitant to see it as GEOS-Chem specific result with other implementations of this method to box models in other regions finding similar sensitivity (Chen et al. 2012).

The model version used in this study (v9-02) implements many of the Fischer et al. updates such as the implementation of the Paulot isoprene oxidation scheme, updating various rate coefficients, and increasing the deposition flux of PAN. Not all of the updates suggested by Fischer et al. have been included in the standard code as of yet but are slated to be included in v11-2 http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_v11-02.

2) The large response to soil NO_x 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

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al., 2012). Therefore ozone sensitivity at altitudes in the mid and upper troposphere is presumably driven by response to uncertainty in soil NO_x 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.

Response: Over much of our Arctic domain in the summer, soils along with biomass burning are the primary emissions sources of NO_x in the model because of the lack of major anthropogenic sources. The stability of the Arctic atmosphere brought up in the Stohl and Wespes et al. papers is more of an issue for the winter and spring periods in which the thermal inversion is stronger. In the case of the Stohl paper, the greatest summertime sensitivity to midlatitude transport was further north than almost all the flights in ARCTAS-B. Also, in our results the sensitivity to soil NO_x was most pronounced in the summertime, not the spring when this higher altitude transportation from the mid-latitudes is more important over the Arctic domain. You are correct that advection from the midlatitudes into the mid-high troposphere is an important consideration in this domain, especially for the springtime. This point was made noting the sensitivity to Asian and USA emissions P10 L20-24. Bringing up specifically the dynamic reasons for this sensitivity is a good idea and is now made more explicitly (P10 L 24).

As far as the chosen uncertainty range, you are correct in there being some uncertainty to our chosen uncertainties. In the case of soil NO_x, there has been some more recent efforts made with satellite data such as Vinken et al. (2014) to reduce this un-

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certainty. However, as they note in citing Schumann and Huntrieser (2007), there is still a large variability in these estimates (4-15 TgN yr⁻¹). This large range of estimates carries over to biomass burning emissions as well (6-12 TgN yr⁻¹) (also Schumann and Huntrieser, 2007 as cited by Vinken et al., 2014). With this, a factor of 3 uncertainty may be slightly on the high side, but not unreasonable in our opinion. In tests we also varied the uncertainty of all the factors to $\sigma/2$ and 2σ in addition to the 1σ analyzed in this study and found almost exactly the same qualitative results (quantitatively the sensitivity indices values varied a few percent) giving us confidence in these results for a variety of different uncertainty ranges.

Changes: Added reference to the Vinken et al. and Schumann and Huntrieser papers P4 L10-13.

3) To what extent is the large HO_x response to gamma HO_2 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 HO_2 . How do these gamma HO_2 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 HO_2 (e.g. Mao et al., 2013)? How does the choice of product (H_2O_2 or H_2O) 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).

Response: Certainly the high uncertainty in Gamma HO_2 contributes to the high sensitivity. This high uncertainty is both evident in the JPL evaluation and in the wide range of treatment and values historically used in GEOS-Chem (P5 L10-17). Also, as we noted in the response to the previous point (# 2), in tests varying the uncertainty

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ranges, we found very similar results.

We described on Page 7 how we constructed the distributions in Section 2.2.1 (“Uncertainties”). As the perturbations followed a lognormal distribution, listing a range of values may not be most useful to the readers as the high and low values would be in the tails of the distribution and not indicative of the vast majority values used in the study. Excluding the upper and lower 5% of the distribution, the values roughly range from 0.04 to 1 which is within the range of values historically used in GEOS-Chem. We touched on the range of gamma values in (P12 L26, P13 L26) describing what values of gamma HO₂ provided the closest match to observed summertime HO₂ profiles.

As for CO, when the modeled HO₂ uptake produces H₂O₂ instead of H₂O, we find CO mixing ratios to be decreased throughout the vertical profile on the order of 10ppb for both spring and summer. Thus, this change exasperates the underprediction of modeled CO with the uptake product of gamma HO₂ being H₂O₂ rather than H₂O. As you note, models tend to underestimate CO in the high latitudes. While this is the case for the Arctic spring, in the summer we found the model to over-estimate CO by around a factor of 2 in the lowest 2km of the troposphere before shifting to under-prediction above 4km (Figs S1 & S2). As for ozone, we found very modest differences between these two scenarios as evidenced by the blue dashed lines in (Figs 2 & 6).

Changes: For readers interested in CO profiles and how the aerosol uptake product of HO₂ affects CO profiles we’ve created figures for both spring and summer in a new supplement (S1, S2).

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

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specific days, when there are certain specific air mass origins.

Response: This is a good point. While when writing the paper we thought readers would understand the geographic limitation of the study area, but it is probably best to make it clearer as suggested.

Changes: In the abstract instead of “period”, “flight tracks” is substituted (P1 L6). Also P8 L30-31 changed to “...providing a fairly representative view of the Arctic troposphere over this domain for the times corresponding to these flights.”

Specific / minor comments

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

Changed as suggested

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.

Changes: Added a citation to Granier et al. 2006.

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.

Changes: The Wild and Prather paper made this point (section 3, paragraph 13). In lieu of this general point, we've added citations to some papers dealing with each of these three specifically (meteorology, emissions, chemistry) Kinnison et al. 2007 for meteorology, Fischer et al 2014/Jaegle et al. for emissions, Chen et al., 1997 for chemical reaction rates.

Page 2, Line 10: Omit semi-colon.

Changed as suggested

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

Correct. Changed as suggested.

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

Changed as suggested

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 NO_x - see point above) framing the choice of factor 3 uncertainty against any other estimates would be helpful.

See comments for general point 2

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?

When excluding OH measurements taken within smoke plumes ($\text{HCN} > 1000$ ppt), the mixing ratios differ less than 10% in nearly all the vertical bins. This is similar to what was noted in the paper with HO_2 where there is a similarly small effect. This difference doesn't change the conclusions of the paper.

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.

Scaled back a bit P8 L18-25 and removed the last sentence in that section.

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.

Response to general point 4 should cover this.

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.

The order of the figures seems to be in a good, logical order as currently ordered so the reference to these figures has been removed here. The new sentence was already edited for general point 4.

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

Correct. Changed as suggested

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.

Response: Thank you for bringing this recent literature to our attention. After considering some of the NO_x profiles, the ozone conclusions have been refocused in a different direction and mention the POLMIP results.

Changes: Moved discussion of Alvarado paper and its comparison to the POLMIP results into Section 3.2.2 (P11, L10-14)

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