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Interactive comment on "Global sensitivity analysis of GEOS-Chem modeled ozone and hydrogen oxides during the INTEX campaigns" *by* Kenneth E. Christian et al.

Anonymous Referee #2

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1   General comments

This manuscript presents a global uncertainty analysis of the concentration of oxidants (O_3, OH, and HO_2) at various altitudes through the troposphere, and in four geographical regions (central and northeastern U.S. and Canada, Gulf of Mexico, Pacific ocean near Honolulu, Hawaii, and Pacific ocean near the southern coast of Alaska). The authors use an ensemble of 512 GEOS-Chem simulations in which various inputs have been pseudo-randomly perturbed within prescribed uncertainty ranges, along with the high dimensional model representation (HDMR) technique to apportion the uncertainty in modeled oxidant concentrations to each of the perturbed inputs. The geographical

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regions studied in the manuscript feature various chemical and meteorological regimes and different local and upwind emissions profiles. Comparison of the results for these regions brings valuable insight into the model inputs that influence oxidant concentrations in these various conditions. The study is well conducted and the results are clearly presented and explained.

A concern for the publication of this manuscript is the similarity of this study with a previous study (Christian *et al.*, 2017a), mostly by the same authors. I do think that the proposed manuscript brings significant new contributions that warrant publication, but the authors should discuss more explicitly the insights that are novel and significant in this manuscript compared to the authors' previous work. The sections below describe in more detail the suggestions and comments that I would ask the authors to address prior to publication.

2   Specific comments

- As mentioned in the Overview section, the authors should discuss the novel insights that this study brings compared to the previous work of Christian *et al.* (2017a). One novel and insightful aspect of the proposed manuscript seems to be the comparison of the uncertainty apportionment between different regions, as well as the vertical resolution of the analysis.

- A similarly worded description of HDMR is already present in the previous work. Although it is useful for the proposed manuscript to summarize the principal concepts of this method, I suggest this description be re-worded further. The same comment applies to other parts of the "Methods" section.

- Page 4, Equation (1): shouldn't $f_i(x_j)$ be $f_i(x_i)$ instead?

- Page 2, Lines 16-22: "Instead, the sensitivity analyses of GEOS-Chem modeled

results has either used local methods in which the factor of interest is perturbed individually and compared to the model state without this perturbation, or the GEOS-Chem adjoint (Henze et al., 2007). [...] While useful in determining some individual sensitivities, these methods neither can nor were intentioned to provide a complete picture of model sensitivities in which many inputs have uncertainties."

The adjoint sensitivity technique can be used to efficiently calculate first-order sensitivities of a model metric or cost function to many model inputs (sensitivities of a given model metric or cost function to all model inputs can be efficiently calculated with a single "adjoint simulation"). Although the results from such an analysis provide "only" first-order local sensitivities, one can argue that they do provide a fairly comprehensive picture of model sensitivities for a given metric and a large number of model inputs that have uncertainties.

A strength of the HDMR method used in the proposed manuscript resides in the fact that model non-linearities are accounted for in the propagation of uncertainties, while other sensitivity approaches are often limited to first-order sensitivities. However, the HDMR approach does require a large number of model simulations (512 here). Additionally, the apportionment of the overall uncertainty with the HDRM method relies on *a priori* estimates of the uncertainties on relevant inputs. Sensitivity or uncertainty apportionments based on other sensitivity methods often do not depend on such *a priori* estimates.

Could the authors discuss these considerations in greater detail in the manuscript prior to publication?

- Page 3, Lines 18–20: "As uncertainties are not published for the meteorological models, we define our meteorological uncertainties as the average of the monthly standard deviations of the difference between GEOS-4 and GEOS-5 meteorological fields for 2005, a year of overlap between the models."

How different are these two models? If they are fairly similar, the uncertainties

on meteorological inputs may be significantly underestimated. Can the authors discuss the fairness of this assumption?

- Page 3, Lines 12–13: "In general, there were typically small differences between modeled results using either $4° \times 5°$ or $2° \times 2.5°$ resolutions but we illustrate in our results where this is not the case."

  The authors do discuss some of these differences in the Results section (for example: section 3.1.2), but it would be insightful to see more quantitative information describing the model-versus-observations agreement (for example: mean bias, standard deviation) with the lower resolution simulations on the one-hand, compared to the higher resolution simulations on the other hand.

- Page 5, Equations (3) and (4): I am unsure as to whether φ_r^i in equation (3) is the same as φ_p^i and φ_q^j in equation (4) if $r = p = q$ and $i = j$. Additionally, the use of superscripts as indices can also introduce confusion. Can the authors add text to clarify these concepts?

- Can the authors discuss the contributions of uncertainties associated with inputs interacting with one another (*i.e.* "missing" slices in Figures 6–9), and the significance of these missing slices for the interpretation of the results presented in the manuscript?

3 Technical corrections

- The authors repeatedly use the word "standard" to refer to notions such as "common practice" or "default value" or "default configuration". I suggest that the word "standard" be reserved for a more restrictive meaning of the word (*i.e.* a formalized norm or convention). Examples:

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- – Page 3, Line 8: "We use in this study the standard GEOS-Chem model"
- – Page 3, Line 21: "the model ensemble made use of the standard emissions inventories"
- – Page 4, Line 7: "the standard model treatment"
- – Page 14, Line 21: "as opposed to the standard 0.20"

In what follows, text that I suggest be removed is written inside curly braces and in red, and suggested replacement text is in blue.

- Page 2, Line 15: "save for {a} some recent work"

- Page 3, Line 8: "We use in this study the standard GEOS-Chem model (v9-02), a {popular}widely-used global chemical transport model"

- Page 3, Line 21: "{For much of the developed world}For many industrialized regions"

- Page 6, line 29: "During INTEX-A, the NASA DC-8 primarily sampled the eastern half of the United States and Canada {INTEX-A} during the summer of 2004"

- Page 9, Line 5–6: "aerosol uptake {to}of HO_2"

- Page 9, Lines 8–9: "In contrast, {uncertainty}uncertainties in both OH and HO_2 mixing ratios were considerable"

- Figures 6–9: the different colors used in these Figures translate to very similar shades of gray when converted to gray-scale. I suggest changing some of these colors so that the different categories of inputs (Emissions, Kinetics, Photolysis, Meteorology, Heterogeneous) can be more easily distinguished when these Figures are converted to gray-scale. I also suggest showing on these Figures the

numerical values corresponding to the slices (*i.e.* contribution of each input to total uncertainty, in %), at least for the largest slices.

- Acknowledgments: "University Maryland". Missing word "of"?

4 Reference(s)

Christian, K. E.; Brune, W. H.; Mao, J. 2017a. Global sensitivity analysis of the GEOS-Chem chemical transport model: ozone and hydrogen oxides during ARCTAS (2008). *Atmospheric Chemistry and Physics*, 17 (5) 3769–3784.

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