

Interactive comment on “Evaluation of updated nitric acid chemistry on ozone precursors and radiative effects” by K. M. Seltzer et al.

Anonymous Referee #1

Received and published: 2 March 2015

I like the experiment performed by the authors. They have evaluated the results of changing the $\text{NO}_2 + \text{HO}^* \rightarrow \text{HNO}_3$ kinetics in a Global Chemical Transport Model. They have evaluated both the changes in chemical concentration, as well as computing subsequent radiative effects with an offline radiative transfer model.

This paper requires significant clarification of several parts of their experiment. In addition some conclusions, discussion, and hypothesis are interspersed in the results rather than being in the discussion.

This paper is not ready for publication in its current state. After a rewrite of the discussion, the paper should be reevaluated.

I look forward to seeing the rewrite.

C487

Specific Comments:

A page number and line number precede each of my comments.

3220, 21: Instead of speaking of "trade off", simply state "In comparison to regional models, GCTM's have decreased sensitivity to boundary conditions and increased sensitivity to emissions, transport, and chemistry". A reference to some paper showing this comparison would be useful.

Section 2: In the methods section, the authors need to clarify how they compute an annual forcing, when they only seem to run the GEOS-chem model for the INTEX-A periods.

3225, 14: Is there a reference for P. Wennberg's data? If not, this is fine.

3227, 13: Could the authors be specific as to which techniques are precluded? In addition, is there a reason that a simple r^2 regression test would not be valid?

3229, 5: "The affect of temper ... altitude." This sentence is misleading. There may be other ways to see the effects. Perhaps the authors choose to evaluate this sensitivity in this manner?

Section 2.5: Surface radiative forcing is confounded discussion. The authors need to clarify if the forcing is "instantaneous radiative forcing" or "radiative forcing". "Radiative forcing" was defined as the change in flux (at the top of atmosphere or tropopause) including a stratospheric temperature adjustment under the assumption of fixed dynamical heating. If the authors have computed "instantaneous" forcing, then the surface forcing makes sense, otherwise they need to address the extent of atmospheric and surface process adjustments.

3230, 13: "result in variances in " -> "affect"

3230, 15: While previous papers by Henderson, et. al., have focused on the 8-10 km region, readers of this paper will be caught off guard by this sentence. Perhaps a note

C488

in the introduction, or something clarifying the reason for this focus at this point in the paper would be useful.

Section 3.2 Please either list the computed radiative effects of each component or remove the discussion of having evaluated each of them and focus on the ozone and sulfate radiative effects.

In addition, the authors need to clarify whether the radiative forcing is computed as an instantaneous effect, or with the stratospherically adjusted temperature due to fixed dynamical heating. If Strat. Adjust. was not used, then the 4 month equilibrium is a red herring.

One more clarification would be to state that the "change in flux" is a net increase in net downward solar and terrestrial flux due to the change in mechanism. (both "net"s are necessary as well as the "downward") You could, instead, simply refer to net trapped energy.

Several times, the authors refer to troposphere when they mean tropopause.

3235, 3: "Due to the increase... " "The increased ozone leads to a net increase in trapped energy beneath the top of the atmosphere of ... and beneath the atmosphere of...â Please also clarify that for the sulfate aerosol, the increase the albedo of the earth system, reflecting additional solar radiation to space.

3235, 10-20: I am uncertain what the authors are trying to say. This paragraph needs to be rewritten. Perhaps they are trying to say that while the methods and altitude at which radiative forcing are computed are different from those used in the IPCC, the relative magnitude of the correction indicated that the change to the kinetics could be important to understanding processes relevant to policy? If so, this paragraph may belong in the conclusion rather than results.

3235, 26: "Also, a larger magnitude of forcing.... surface. The net atmos..... precipita-tionâ Perhaps the authors mean "The net absorption of energy by the atmosphere

C489

as seen in the third panel of figure 4 will affect convective and transport processes." While the reference to Shindell's analysis is nice, does the total of the ozone effect and the aerosol effect lead to a clear effect on precipitation that is explicitly confirmed by the results in this paper, or should this also be in the discussion?

3236, 5: Do the authors mean "indirect effects" or "atmospheric responses and feed-backs"?

3236, 5: Do the authors mean "simulation" or "offline computation"?

3236, 7: Are the "localized adjustments" an increase or decrease in oxidation of the SO₂ -> SO₄? And is there data to back up this assertion?

3236, 16: "It is hypothesizedâ This sentence belongs in the discussion.

3236, 23: both HNO₃ and NO_x have an inverse relationship with what? Perhaps with each other? Would a scatter plot make that inverse relationship clear?

3236, 25: Why are these counterintuitive? Is there a reason for these to be opposite our intuition? It would be useful to have a reason why these results are the opposite of the direct effect of the kinetics.

3237, 10: "The previous hypothesis"... I do not know to which hypothesis the authors refer. Perhaps the discussion of sulfate distribution belongs in another paragraph stating that the sulfate concentrations are more localized to the surface and to more polluted areas.

3237, 19: I do not know what is meant by "Literature updates". Perhaps "Updates to the NO₂+HO reaction rate provided by () have been implemented in GEOS-Chem. The resulting changes in chemistry composition more closely match the INTEX observations. In particular we find..."

3238, 1 I don't know what is meant by "was decreases". Perhaps the authors mean to say, "Decreases in No_x lead to a near global increase in ozone. The resulting increase

C490

in oxidation potential leads to an increase in sulfate. Additional work needs to be done to understand the surface layer concentrations of HNO₃ and NO_x, as they are contrary to the direct implication of the decreased reaction rate coefficient."

3238, 18: The authors need to clarify what they mean by "performance".

3238, 28: "change in ozone sensitivity". Sensitivity to what?

Paragraph starting at 3238, 28: This paragraph needs help. I don't know what is meant by "modest", or how a "model uses NO_x". Do the authors mean "sensitivity of predicted O₃" or "change in O₃ concentrations"? I am having a hard time understand the specific meaning of these sentences.

Paragraph starting at 3239, 9: The first two lines of this paragraph could be rewritten to say, "The radiative effects of the change in ozone and sulfate distributions was evaluated with an offline radiative transfer code". Please refer to previous discussion of how to be precise about forcing numbers. (Yes, I know this is a bother. Thanks for being precise.) Do you mean variance or change?

3239, 25: To which policy implications do the authors refer? I do not understand the second sentence of this paragraph. Do the authors mean "robust" or "very similar"? Why do the updates need lab confirmation? What additional evidence, in particular, would be helpful?

For all figures: Are these annual averages, or only average during the INTEX period.

Technical Corrections:

Spelling should be checked and corrected throughout.

3222, 15: "trace gas composition.... atmosphere" -> "concentration in the troposphere and subsequent changes to radiative fluxes"

3222, 25: Split the line starting with "The GEOS-Chem..." into 2 sentences: one with what is simulated and one describing the inputs.

C491

3223: "concentration fields" -> "chemical concentrations" or "concentrations of chemicals"

3223, 8: "will cover" -> "covers"

3225, 1: "extreme" -> "extreme events, or events on a horizontal scale smaller than model resolution"

3227, 3: "which can reduce the va...." -> "reducing the variance of chemical concentrations"

3231, 23: "and nitrogen partitioning" -> "and partitioning of NO_y species"

3232, 29: "udpated" -> "updated", "high-bias" -> "high bias in O₃". In addition, is there a reference for the fact that the baseline model has a high bias?

3233, 8: "profie" -> "profile"

3234, 8: "each were" -> "all were".

3234, 10: "complete difference associated with " -> " combined effect of"

3234, 13: "localize" -> "localized"

3234, 14: remove "variables"

3234, 15: "changing atmospheric.... enables" -> "change in atmospheric oxidation potential."

3235, 24: "it is observed" -> "figure 4 shows"

3236, 10: "only above land" -> "predominantly over land"

3236, 14 "associated"

Section 3.3 Stating that "The change in kinetics leads to a change in the spatial distribution of... could combine the first two sentences

C492

3236, 23: Remove the phrase "which displays...simulations".

3237, 1: "inversely proportional" or "inversely related"?

3237, 6: Perhaps the authors mean "Figures 8 and 9 show a nearly global extent of increases in ozone with particular increases in the....âĀĲ

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 3219, 2015.