

***Interactive comment on “Aviation 2006  
NO<sub>x</sub>-induced effects on atmospheric ozone and  
HO<sub>x</sub> in Community Earth System Model (CESM)”  
by A. Khodayari et al.***

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The manuscript by Khodayari et al. examines changes in ozone, HO<sub>x</sub>, methane lifetime, and ozone radiative forcing due to aviation NO<sub>x</sub> emissions, based on simulations using the CAM4 and CAM5 models. The findings, though not surprising, are clear and interesting, and will be a useful addition to the discussion on aviation effects on atmospheric composition and climate. The manuscript is well written and the topic is certainly suitable for Atmospheric Chemistry and Physics. Therefore, I recommend publication following some revisions described below.

GENERAL COMMENT:

The manuscript is based on results from two versions of the CAM model. This certainly adds value to this work, as it explores (to some extent) the robustness of the results. However, the manuscript does not provide substantial insight as to why the two models respond in significantly different ways when it comes to oxidant responses to aviation NO<sub>x</sub> emissions. The fact that the authors stress from an early stage (Abstract) that the main difference between the models is that they handle aerosols differently, and the fact that in Section 2 there is a fairly thorough description of the aerosol component of the models, makes the reader expect that the results will be discussed in the context of those differences in the model set-up between the two sets of experiments. More specifically, one expects the answer to the question: why does a different handling of aerosols in the models lead to a) differences in oxidants, and b) different responses to aviation NO<sub>x</sub>?

I would suggest that the authors at least add some discussion to address such questions.

*We thank the reviewer for the comments and agree that some more discussion has to be added on the reason for differences in oxidants and response to aviation NO<sub>x</sub>, please see specific comments below.*

*It is also noted that minor differences between the model simulations in the original manuscript are due to differences in the amount of lightning NO<sub>x</sub> in the model. In order to remove this difference, we adjusted the CAM5 simulation to produce a similar amount of lightning NO<sub>x</sub>, as produced by CAM4. As such, we revised the manuscript and updated the numbers with our new simulations.*

SPECIFIC COMMENTS:

Page 6164, Lines 25-27: It is important to clarify here whether the radiative forcing estimate includes effects of NO<sub>x</sub> on ozone through methane. Clearly the study does not address such an effect, but it should be clear in the abstract.

*Sure. The sentence at page 6164 lines 25 was modified to “Aviation NO<sub>x</sub> emissions are associated with an instantaneous change in global mean short-term O<sub>3</sub> radiative forcing (RF) of 40.3 and 36.5 mWm<sup>-2</sup> in CAM5 and CAM4, respectively.” to reflect the fact that calculated changes in ozone are related to short-term ozone and not changes in ozone through methane.*

Page 6165, Lines 23-25: I am not sure whether this statement is correct: aerosols would cause more cirrus nucleation, leading to more longwave radiation being trapped, and thus leading to heating.

*This sentence at page 6165 line 23 was modified to the following sentence to avoid confusion. This is based on the Gettelman and Chen, 2013 work which concluded that overall, the indirect effect of aviation aerosols results in negative forcing.*

*“The indirect effect of sulfate aerosols may, on the other hand, result in a negative radiative forcing via liquid clouds which dominates the warming caused from contrails and black carbon (BC) emissions (Gettelman et al., 2012).”*

Page 6166, Lines 8-11: Please add a few sentences to summarise the findings of these “several studies”.

The following paragraph has been added to page 6166 line 9 to summarize the findings of these “several studies.

“Derwent et al. (2001) analyzed the changes in methane and tropospheric ozone after emitting pulses of NO<sub>x</sub> at the surface and upper troposphere in both the northern and southern hemispheres and found that while the changes in methane radiative forcing were dominated by methane emissions, changes in tropospheric ozone radiative forcing were dominated by changes in ozone precursor gases, notably NO<sub>x</sub> emissions. Stevenson et al. (2004) looked at the effects of an extra pulse of aviation induced NO<sub>x</sub> at four months representing the seasonal cycle. Their results showed a seasonal dependence in the O<sub>3</sub> radiative forcing with a long term net radiative forcing of approximately zero.”

Page 6166, Lines 27-28: But the paper does not really discuss how different aerosol handling can modulate such effects. Can you suggest some possible mechanisms?

We agree with the reviewer and added the following paragraph on the main differences between CAM4 and CAM5 in the aviation NO<sub>x</sub> emissions and simulation setup section (section 3).

“Since both models were run with same emissions, same total lightning NO<sub>x</sub> values, and with identical meteorological fields with 100% nudging, the differences in the description of aerosols very likely have the largest impact on the chemistry of aviation NO<sub>x</sub>-induced effects, while differences in clouds may also contribute to some degree. In particular, differences in the aerosol burden, but especially in the surface area density that are caused by differences in the aerosol size distribution (effective radius) and mass, have an influence on the heterogeneous chemistry and therefore influence the oxidative capacity of the atmosphere and therefore the chemical composition, as further discussed in Section 4.4. The impact of differences in dynamics is expected to be small, since in both models the horizontal winds, surface fluxes and temperatures were prescribed with GEOS meteorological analysis fields.”

Also, we added the following discussion on the impact of aerosols on chemistry to Section 4.4.

“As shown in Table 1, The ratio of NO<sub>x</sub> : NO<sub>y</sub> is about 7% higher in CAM5 perturbed run than in CAM4 perturbed run implying a smaller shift of the NO<sub>x</sub> : NO<sub>y</sub> relationship to NO<sub>y</sub> in CAM5. The smaller shift of the NO<sub>x</sub> : NO<sub>y</sub> relationship to NO<sub>y</sub> in CAM5 is tied to heterogeneous reactions and related to less aerosol surface area density in CAM5 compared to CAM4. Under lower aerosol surface area density, heterogeneous reaction can be less effective in moving NO<sub>x</sub> to NO<sub>y</sub> and this results in more OH, and shorter CH<sub>4</sub> lifetime (as seen in Table 2). Heterogeneous reactions that are included in CAM chemical mechanism are listed in Eq 1-3.



As such, due to less efficient transfer of NO<sub>x</sub> to NO<sub>y</sub> in CAM5 compared to CAM4 there is more nitrogen available in its reactive form (NO<sub>x</sub>) to trigger the ozone formation reactions in CAM5, resulting in higher aviation NO<sub>x</sub>-induced ozone perturbation.”

Page 6167, Lines 20-23: So, it is not just the aerosols that differ, it seems. If so, you would need to amend previous parts of the text (e.g. lines 27-28 on page 6166 and lines 9-11 on Page 6168, Line 16: What is a “triangular distribution” in this context? Please explain.

We agree with the reviewer that certainly not only the aerosol treatment is different but also the underlying physical processes between CAM4 and CAM5. We updated the model description (Section 2) and also Section 3 to point this out more clearly. Differences in the description of aerosols have very likely the largest impact on chemistry, while differences in clouds may also contribute to some degree. In particular, differences in the aerosol burden, but especially in the surface area density that are caused by differences in the aerosol size distribution (effective radius) and mass, have an influence on the heterogeneous chemistry and therefore also on the chemical composition.

Besides this, minor differences between the model simulations in the original manuscript are due to differences in the amount of lightning NO<sub>x</sub> in the model. In order to remove this difference, we adjusted the CAM5 simulation to produce a similar amount of lightning NO<sub>x</sub>, as produced by CAM4. On the other hand, the impact of differences in underlying physical processes (dynamics) is expected to be small,

*since horizontal winds, surface fluxes and temperatures were prescribed with GEOS5 meteorological analysis fields.*

*Triangular distribution has its own typical definition here, a distribution with a lower limit, an upper limit and a mode.*

Section 2: As stated earlier, this section is dominated by the description of the differing aerosol handlings, whereas there is not much discussion on how these matter later on. On the other hand, not much is mentioned for other crucial aspects of the models, e.g. how is photolysis handled?

*Please see previous comment.*

Section 2, last paragraph: Please briefly state why the radiative forcing calculations were not performed online, to avoid inconsistencies with model (e.g. having to use ISCCP etc).

*The clouds were used from ISCCP since we wanted to use consistent cloud fields for both CAM4 and CAM5 simulations to make sure the differences we get in radiative forcing is due to the differences in chemistry and not due to the differences in the cloud fields. Previous study by Conley et al. 2013 shows that using different cloud fields in an offline radiative transfer model makes very little to no difference on the calculated change of radiative forcing for radiative active species. The following lines were added to Page 6169 line 5 to clarify this:*

*“The use of the same cloud fields for both CAM4 and CAM5 simulations in the offline radiative forcing calculations ensures that the differences in the calculated change in radiative forcing are due to the differences in chemistry and not due to the differences in cloud fields. Previous study by Conley et al. 2013 shows that using different cloud fields in an offline radiative transfer model makes very little to no difference in the calculated change of radiative forcing for radiative active species.”*

Page 6169, Line 26: Is it certain (e.g. from previous studies, or from the authors' tests) that the atmosphere would need 6 years to reach steady-state, even with fixed methane? It might have been worth discarding less years and using more years to assess statistical significance post spin-up. Please discuss.

*Yes, it is...we did look at the year-to-year variation of results to figure out the number of years of simulations needed to reach steady-state; this is also consistent with other studies such as Olsen et al, 2013.*

Fig. 1: Please state in caption that this is for the control runs.

*Done. The sentence was modified to “Taylor diagram of modeled background ozone from the control runs against ozonesonde climatology for four pressure levels and three latitudinal regions.”*

Pages 6170-6171: It would be interesting to discuss in this section whether adding aviation NO<sub>x</sub> emissions improves model performance in general (in CAM4 and in CAM5).

*We added a new comparison between model simulations for both control and perturbed case and aircraft observations (please see the new Figure 2). Additional aviation NO<sub>x</sub> does not improve the model performance in comparison to available observations; it rather results in a slightly larger overestimation of ozone between 2-7 km. The model performance may be improved by reducing the amount of lighting NO<sub>x</sub>, however, this is beyond the scope of this paper.*

Pages 6174, Lines 2-3: Do not see ozone decreases below 450hPa in Fig. 5. Ozone seems to be increasing everywhere. The authors possibly refer to the net ozone production.

*Correct. In pages 6174 Lines 2-3, we are referring to the changes in the net rate of ozone production. So, even though the net rate of aviation NO<sub>x</sub>-induced ozone production is decreasing below 450 hPa (as seen in Figure 5 (old Figure 4)), the changes in the aviation NO<sub>x</sub>-induced ozone could be positive (as seen in Figure 6 (old Figure 5)). For clarity, the sentence at page 6174 line 2 was modified to “The impact of aviation induced NO<sub>x</sub> on ozone results in a net increase in the rate of ozone production with a maximum around 250hPa, and a net decrease in the rate of ozone production below 450 hPa.”*

Pages 6175, Lines 6-7: Differences are arguably not “small”, compared to intermodal variability, but perhaps “smaller”.

*Agreed. The sentence was modified to “While such differences seem to be smaller compared to ...”.*

Pages 6176, Line 11: Any ideas why it is more distributed towards the surface in CAM5.

*It is likely due to the differences in aerosols surface area density.*

Pages 6176, Lines 26-28: Which feature? The previous sentences just state that the high-altitude perturbation extends into the surface for both seasons/models.

*The feature refers to the extension of the high-altitude perturbation extending to the surface for both seasons/models.*

Pages 6177, Line 27: But why is photolysis increasing in the mid-troposphere?

*It is not that the photolysis is increasing in the mid-troposphere, the increase in the HO<sub>x</sub> formation is the results of the two factors, the O<sub>3</sub> photolysis and the presence of water vapor. The aviation-NO<sub>x</sub> induced O<sub>3</sub> photolysis is greater in the upper-troposphere, the water vapor is higher at lower altitudes and overall the outcome of these two trends is that we see increased production of HO<sub>x</sub> in the mid-troposphere. The sentence at page 6177 and 27 was modified to the following sentence for clarity:*

*“This is due to the increased production of HO<sub>x</sub> in the mid-troposphere triggered by O<sub>3</sub> photolysis and the presence of water vapor.”*

Pages 6178, Lines 21-22: Please change “concentrations” to “perturbations” or “concentration changes”.

*Done.*

Pages 6178, Line 24: I would rephrase to “thus increasing OH, and subsequently HO<sub>2</sub>”.

*Done.*

Pages 6179, Line 6: Please rephrase to “In both CAM5 and CAM4, the changes in methane loss are mostly confined...”

*Done.*

Table 3: It is stated in the caption that the models underestimate observation-based methane lifetime. I would suggest mentioning this in the text too. It is also worth mentioning that most current models feature such methane lifetime underestimates (e.g. see ACCMIP studies for most recent multi-model estimates: Voulgarakis et al., 2013 and Naik et al., 2013).

*The following sentence was added to page 6179 line 16:*

*“It is noted that same as most other models (Voulgarakis et al., 2013 and Naik et al., 2013) the calculated lifetimes here are shorter than the CH<sub>4</sub> lifetime derived based on Methyl chloroform analysis (Prather et al., 2012).”*

Pages 6179, Lines 18-19: It would be useful to give reference for the methane lifetime feedback concept, as well as for the feedback factor mentioned further down (line 23).

*Done. The following references were added to page 6179 line 19 and 23 for methane feedback and the feedback factor of 1.4:*

*“...feedback of changes in methane concentration on its own lifetime (e.g. Prather 1994; Fuglestedt et al. 1999; Wild et al. 2001 and IPCC 2007).”*

*“... decreases the lifetime by a factor of 1.4 (IPCC, 2001).”*

Section 4.5: It is worth highlighting that the overall highest RF changes are over Southern Europe and the Middle East.

*The sentence at page 6180 line 6 was modified to the following sentence to highlight the area of highest RF changes:*

*“Both models show the greatest RF in the NH between 30-60°N with highest RF changes over Southern Europe and the Middle East.”*

Pages 6180, Lines 14-15: How do these figures compare with RF from other sectors? It would be useful to put these estimates into context.

*Fuglestedt et al. 2008 does a detailed comparison of aviation contribution in changing the radiative forcing to the contribution from other transportation sectors in great detail. The following sentence was added to page 6180 line 17 to provide a reference for such comparison.*

*“It is noted that Fuglestedt et al. 2008 compares the aviation contribution in changing the radiative forcing to the contribution from other transportation sectors.”*

Pages 6180, Lines 16-17: Please explain more clearly what is meant by “short-term ozone”.

*The following sentence was added to page 6173 line 2:*

*“The NO<sub>x</sub>-induced changes in tropospheric ozone are complicated by two stages, a short-term increase in O<sub>3</sub> concentrations associated with a positive forcing, and a long-term reduction of O<sub>3</sub> concentrations tied to the aviation induced methane decrease. This long term-reduction is associated with negative forcing (Wild et al., 2001; Stevenson et al., 2004). Since our simulations were performed with fixed CH<sub>4</sub> mixing ratios at the boundary layer, the calculated changes in O<sub>3</sub> concentration are the short-term changes.”*

Pages 6181, Line 10: I believe the authors intended to write “longer” rather than “shorter” when referring to the winter photochemical lifetime of ozone.

*Fixed.*

Pages 6181, Line 25: I do not particularly like the phrase “various complicated aerosols processes”. Please either remove this or state which processes are implied.

*Removed.*

Pages 6181, Lines 25-28: I understand that the authors meant to state that the difference between CAM3 and CAM5 ozone responses is smaller than the present-day model uncertainty regarding these effects. If so, please rephrase.

*The sentence at page 6181 line 25 was modified to the following sentence:*

*“It is noted that while the simulated change in ozone is relatively different between the two models, the difference between CAM4 and CAM5 ozone responses is considerably smaller than the current estimates of the uncertainty in aviation effects on ozone.”*