Interactive comment on "Aviation 2006 NO_x-induced effects on atmospheric ozone and HO_x in Community Earth System Model (CESM)" *by* A. Khodayari et al.

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The manuscript by Khodayari et al. reports the results of calculation of the impact of aviation NOx emissions using two versions of the NCAR CAM model using identical chemical mechanisms. The model is based on 2006 aviation emissions. Results are presented for the calculated ozone perturbation, HOx changes, and associated radiative forcings.

General Comments:

This a fairly pedestrian paper comparing results of two similar models. Results from the two models are discussed but the reasons for the differences do not seem to be well thought out. The differences in the two models are presented as due to the different aerosol treatments but the authors do not build a strong case that the other differences between CAM4 and CAM5 are inconsequential for the aviation NOx impact. In the discussion of the detailed chemistry, the authors only discuss the gas phase reactions. There is no detailed discussion of the aerosol processes which might support their thesis. If the purpose of the study is to evaluate the reasons for the differences between the two very similar models, then the authors need additional analyses and perhaps additional model runs.

The description of the comparison of the modeled output for a number of the chemical species with observation at "within the central 50% and 90% of available observations" seems very clumsy. The authors seem to believe this is good agreement. This reviewer is skeptical of that conclusion and not quite sure of what to make of that statement. The authors should consider better ways to present quantitative comparisons with observations. It is generally accepted that all models have some weaknesses and the authors should not be afraid to show the challenges as well as the successes.

We thank the reviewer for the comments and agree that some more discussion has to be added on the reason for differences between the models. 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 NOx in the model. In order to remove this difference, we adjusted the CAM5 simulation to produce a similar amount of lighting NOx, as produced by CAM4. As such, we revised the manuscript and updated the numbers with our new simulations.

Also the description of the comparison of the modeled output with observation has been revised to address the reviewer comment.

Specific Comments:

Page 6166 lines 27- page 6167 lines 1-3. The authors state that the major difference between the CAM4 and CAM5 models was the treatment of aerosol chemistry. This is misleading since CAM5 also used different treatments of convection, dynamics, and radiation. Later in the paper, the authors fail to quantify the importance of the aerosol chemistry for this problem. If CAM5 can be run with the bulk aerosol treatment used in CAM4 and shows a similar result to that of CAM4, their claim would be much more persuasive.

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) 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 NOx in the model. In order to remove this difference, we adjusted the CAM5 simulation to produce a similar amount of lighting NOx, 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.

We expect that running with a bulk aerosol treatment in CAM5 would indeed result in similar responses of the models to aviation NOx. However, the architecture of CAM5, which includes the coupling between MAM and cloud properties, does not allow a simple replacement of the two aerosol schemes, which does not make it possible to perform the suggested model simulation.

The following lines at Page 6166 lines 27 were modified to be clearer about the differences.

"While the calculated effects in CAM4 and CAM5 provide a new reference for the aviation NOx-induced effects in comprehensive climate-chemistry models, they also provide a measure for the effects of different oxidative capacity in the models, due to differences in description of the physical processes in the model, and especially due to the different treatment of aerosol processes (see model description).:.

Also this has been addressed more clearly in the last paragraph of the aviation NOx emissions and simulation setup section (Section 3) as follow:

"Since both models were run with same emissions, same total lighting NOx 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 NOx-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."

Page 6167 lines 13-15. The NCAR site shows that there are 5 official releases of CAM5 but the authors seem to be using some beta version. The authors should identify which of those releases is closest to the beta version used in this paper and justify why they are not using one of the official releases.

The following lines were added to Page 6167 line 15 to clarify this.

"Since the coupling of aerosols and chemistry in CAM5 has not been released at the time model runs were performed, a development version of close to CESM1_2_0 release version (cesm1_2_beta08_chem) was used for both CAM4 and CAM5 simulations, which includes this coupling."

Page 6169, lines 3-5. The authors should explain why they are still using zonal mean clouds from ISCCP in their radiative forcing calculations when cloud fields are available that would be self consistent with the meteorology in CESM.

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. A 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. A 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, lines 10-11. The authors describe the total number of vertical levels but should also state the vertical resolution near the tropopause, which is important for this

problem.

The sentences on paper 6169 line 7 were modified to the following sentence to reflect the resolution near the tropopause. "Both models were run at a horizontal resolution of 1.9° latitude x 2.5° longitude and were configured with 56 vertical levels covering from the surface up to ~2 hPa with near tropopause resolution of about 1.3 km."

Page 6170, lines 3-18. The authors describe their evaluation of modeled ozone versus measurements but do not address a number of critical tests for studying NOx perturbations in the upper troposphere – How good is the background NOx and NOy? Is the NOy partitioning accurate or are some species (e.g., PAN) wrong? How does the modeled HOx precursors in the UT compare with measurements? Just evaluating the background ozone is not enough.

Regarding NOx and NOy in the upper troposphere, it is challenging to evaluate these species, since aircraft data do not often reach as far up. An updated evaluation of O3, NOx, HNO3, PAN, and CO in comparison to available aircraft campaigns between 2-7km is presented in the revised version of the manuscript (shown in the new Figure 2). The following lines were added to Page 6172, line 3 to discuss the evaluation of O3, NOx, HNO3, PAN, and CO in comparison with observation:

"Comparisons of O3, NOx, HNO3, PAN, as well as CO to aircraft observations between 2-7 km (Emmons et al., 2000; Tilmes et al., in preparation), where the majority of the observations were taken is also shown in Figure 2, both the control and perturbed simulations.

In comparison to aircraft data, ozone is slightly overestimated in the tropics, especially for the perturbed simulations, in agreement with ozonesonde observations, while there is a reasonable agreement in mid- and high latitudes. Both model versions simulate the regional differences in NOx in comparison to available aircraft observations reasonably well, but NOx is slightly underestimated by all model simulations is summer in NH mid-latitudes. Both model versions overestimate PAN and HNO3 in tropics and mid-latitudes and high latitude in spring. Model differences between CAM4 and CAM5 are within the variability of the observations. CO is underestimated in both model versions, with much larger deviations from the observations for CAM5 than CAM4. This points to a significant overestimation of OH in CAM5, as also indicated by the smaller methane lifetime in CAM5 compared to CAM4. The increase in NOx due to aircraft emissions does not affect NOx, NOy, and CO very much in the altitude considered. However, ozone is slightly increased in the perturbed case for both CAM4 and CAM5." Also, the following sentence was added to page 6170, line 3 to note that previous studies by Weber, 2011 and Olsen et al., 2013 showed that CAM reasonably reproduces the effects of aviation NOx-induced emissions on distribution of tropospheric O3 and NOx."

Also, the following sentence was added to page 6170, line 3 to note that previous studies by Weber, 2011 and Olsen et al., 2013 showed that CAM reasonably reproduces the effects of aviation NOx-induced emissions on distribution of tropospheric O3 and NOx.

"Previous intercomparisons of multiple climate-chemistry models indicated that CAM reasonably simulates the effects of aviation NOx-induced emissions on distribution of tropospheric O3 and NOx (Weber, 2011 and Olsen et al., 2013)."

Page 6171, line 21 – the authors state that in some places the "estimated ozone is very accurate" while noting in other places that it is not very good. Can the authors be more quantitative?

We have revised the text regarding the comparison to ozonesonde observations and remove statements like "very accurate" to be more quantitative.

Page 6172, lines 7-16. The authors report that in the 4-8 km altitude range (i.e., below the altitudes where the aircraft mostly fly) the simulations fall in the range of 50-90% of the observations. That doesn't sound like particularly good agreement to this reviewer although the authors seem to imply that is is. The fact that the models agree with each other when they assume the same chemistry, the same emissions, and the same boundary values does not reveal anything about the accuracy of the model.

As stated above, most available aircraft observation from research aircraft fly in 2-7km and provide the largest coverage for evaluation. Instead of discussing single profiles, in the revised manuscript we sorted different aircraft campaigns regarding location and season and compared those to the model results (similar to what was

done in Lamarque et al., 2012). Differences between both model simulations are much smaller than in the observations.

Page 6173, lines 1-20. If the aerosol treatment is the key factor in explaining the differences between the two models, please add those processes to the mechanism discussion.

We agree with the reviewer and added the following paragraph on the main differences between CAM4 and CAM5 in the aviation NOx emissions and simulation setup section (Section 3).

"Since both models were run with same emissions, same total lighting NOx 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 NOx-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 NOx : NOy is about 7% higher in CAM5 perturbed run than in CAM4 perturbed run implying a smaller shift of the NOx : NOy relationship to NOy in CAM5. The smaller shift of the NOx : NOy relationship to NOy 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 NOx to NOy and this results in more OH, and shorter CH4 lifetime (as seen in Table 2). Heterogonous reactions that are included in CAM chemical mechanism are listed in Eq 1-3.

$$N_2O_5 \rightarrow 2HNO_3 \qquad \textit{(Eq 1)}$$

$$NO_3 \rightarrow HNO_3 \qquad \textit{(Eq 2)}$$

$$NO_2 \rightarrow 0.5 \times (OH + NO + HNO_3) \qquad \textit{(Eq 3)}$$

As such, due to less efficient transfer of NOx to NOy in CAM5 compared to CAM4 there is more nitrogen available in its reactive form (NOx) to trigger the ozone formation reactions in CAM5, resulting in higher aviation NOx-induced ozone perturbation."

"Page 6174, lines 13-26. Results are presented but not a discussion of the processes that explain the differences. For example, why is CAM5 more efficient at producing

ozone than CAM4? Is it a transport effect, a radiative effect, or ???

Please see the previous comments.

Page 6177, line 16. The authors should clarify that the change in mean ozone column ozone is an increase.

Done. The sentence on Page 6177 line 16 was modified to "Figure 7 shows the increase in aviation induced zonal mean annual OH perturbations."