Response to anonymous referee #2

We thank reviewer #2 for valuable comments which helped improve the manuscript. All the reviewer's points have been carefully considered, and our manuscript has been modified accordingly.

Below follow answers to the general and specific comments.

General comments

This study investigates the impact of non-land based traffic emissions on Ozone, OH and RF, for the B1 and B1 ACARE scenarios (improved NOx technology) for 2000, 2025 and 2050. The authors used previously established methods to analyze model results. The paper seems to be lengthy, but could profit from investigating in more detail why for some changes occur (as outlined below).

Answer: The cases outlined below have been investigated in more detail (see answers to specific comments).

The paper is well written and the figures are to the most part readable. The paper is well organized. It can be technically improved by pointing the reader more often to the figures and lines in the plot that are referred to in the text.

Answer: The revised manuscript has been technically improved by pointing the reader more often to the figures and lines in the plot.

This paper is a model study only. However, some important questions should be addressed: How well do models compare to observations? How well do models reproduce chemical species in the year 2000? Is the difference in the RF between the models a result of a different representation of atmospheric species?

Answer: The authors agree and have included in the appendix of the revised manuscript a comparison between the model results and multi-year ozonesonde observations from Logan (1999). The beginning of Section 3 has been modified slightly and, based on the comments of referee #1, we have also included a brief synthesis of the validation study by Schnadt et al. (2010).

Specific comments

Abstract: Line 14: what does '(scaled to 100%)' mean? Not clear in this context. Answer: This has been made clear in the revised manuscript. "Scaled to 100%" means that the response of the 5% perturbation has been multiplied by a factor 20.

Introduction: Earlier study: Sovde 2007: 10ppbv increase of ozone in the UTLS due to aircraft emissions in 2050. What scenario was used in IPCC terms?

Answer: Søvde et al. (2007) used future emission projections from Airbus and these were not based on IPCC scenarios, but the aircraft NO_X emissions of 2.18 TgN yr⁻¹ in their study are comparable to the QUANTIFY emissions for 2050 A1B, which are 2.28 TgN yr⁻¹.

Emissions and simulation setup: Page 16809, line 15: what do you mean with: 'most models update the surface mixing ratios with values from IPCC future simulation'. Are there models that do not change CH4 surface mixing rations for future runs? **Answer:**

There were some differences in the setup of the CH_4 mixing ratios of the different models. Two of the models, p-TOMCAT and MOCAGE, kept the 2000 values also for the future runs, while the other four models (OsloCTM2, UCI CTM, TM4 and LMDz-INCA) updated the mixing ratios based on IPCC (2001). The global CH_4 abundances from IPCC (2001) were 1760 ppbv, 1909 ppbv and 1881 ppbv for 2000, 2025B1 and 2050B1, respectively. The models applied a hemispheric scaling with approximately 5% higher abundances in the northern hemisphere than in the southern hemisphere.

In order to investigate the effect of using different CH_4 surface mixing ratios, sensitivity simulations have been performed with the OsloCTM2 model. Reference (BASE) and perturbation simulations (AIR and SHIP) were carried out for the 2025B1 scenario with the CH_4 abundance fixed to year 2000 (1760 ppbv). The results were then compared to the original OsloCTM2 simulation using 2025B1 CH₄ abundance (1909 ppbv). The 2025B1 scenario was chosen because the difference from the year 2000 CH₄ surface mixing ratio is larger than for the 2050B1 scenario, hence this sensitivity test should provide an upper estimate of how the different CH₄ mixing ratios may affect our results.

Table 1 shows that the relative methane lifetime changes due to aircraft and ship emissions are only slightly affected (up to 1-2%) by using CH_4 abundances for 2000 instead of the 2025B1 value from IPCC (2001). Global distributions of aircraft- and ship-induced ozone and OH have also been investigated, and the impacts of changing the CH_4 mixing ratio were found to be insignificant.

Based also on a comment by referee #1, the manuscript has been updated clarifying which models kept the 2000 CH4 values in the future simulations, and what the CH_4 abundances were in 2000, 2025B1 and 2050B1. A sentence stating that there is only a small effect on the aircraft- and ship-induced perturbations when using 2000 surface methane in the 2025B1 simulations has also been added.

Table 1. Relative changes (%) in methane lifetimes (integrated up to 50 hPa) due to a 5% decrease in aircraft emissions and due to a 5% decrease in ship emissions, as calculated by the OsloCTM2 model for two different CH_4 surface mixing ratios. Values are given relative to the BASE case, and are scaled to 100% by multiplying with 20. Note that this table does not include the feedback effect of methane changes on its own lifetime.

	AIRCRAFT		SHIPPING	
CH_4 :	2000 (1760 ppb)	2025B1 (1909 ppb)	2000 (1760 ppb)	2025B1 (1909 ppb)
2025B1	1.110	1.113	4.013	3.957

Line 20: Meteorological data from the year 2003 were used. This year was a strong El Nino year. Do you have a feeling about the variability of your results if using a different year or a 10-year model average for example?

Answer:

Additional simulations have been carried out with the OsloCTM2 model in order to study the effect of using a different meteorological year:

- 1. The simulations with 2000 emissions have been continued from 2003 and through 2004 for both the reference simulation (BASE) and the perturbation simulations (AIR, SHIP).
- 2. The year 2002 spin-up simulation with 2000 emissions have also been used in the comparison below. However, results from the perturbation simulations for the beginning of 2002 had to be excluded because the amount of spin-up time was not sufficient (the emission perturbation was started on 1 January 2002).

First, the results of the BASE simulations for 2002, 2003 and 2004 have been compared with the Logan sonde observations in Figure 1 (similar to Figure A1 in the revised manuscript). The comparison shows that there are only small differences between the meteorological years, and there is no evidence of an El Niño effect from these plots.

When looking at the total O_3 column in Figure 2, 2003 sticks out in the southern hemisphere compared to 2002 and 2004. Around the equator and south of 60°S, total ozone columns are up to 40 DU lower in 2003, while they are 10-20 DU higher between 10°S and 60°S. Hsu and Prather (2009) found the same signal when calculating total O_3 columns with the UCI CTM using the same meteorological input data from ECMWF-IFS (their Figure 4d). However, they show that the anomalies in 2003 can be largely explained by the quasi-biennial oscillation (QBO). When comparing with a five-year climatology (2001-2005), year 2003 shows similar anomalies as the years with the same QBO pattern (2001 and 2005) except for a stronger positive anomaly between 20°S and 50°S in 2003.

The focus of this paper is on the impacts of non-land based traffic sectors on atmospheric chemistry, hence it is interesting to see if the choice of meteorological year changes the perturbation signal. Figure 3 shows the zonal mean ozone impact of year 2000 emissions from AIR and SHIP (comparable to Figure 5 and 7, respectively, in the manuscript) for different meteorological years (2002-2004). The figure shows that the impact of changing the meteorological year is for the most part minor, and the same signal is seen for the ozone column perturbations (not shown). One exception is the ozone impact of aircraft emissions during northern summer when the difference in the fully scaled perturbation between 2002 and 2004 exceeds 1 ppbv (Figure 3, bottom right). The 2003 results fall in between 2002 and 2004 for this particular case.

Relative changes in methane lifetimes due to emissions from AIR and SHIP have also been investigated in the new simulations with 2004 meteorological data. The original results with year 2003 meteorology showed relative changes of 0.852 % for AIR and 3.77 % for SHIP (Table B1 and B2, respectively, in the revised manuscript). The new

results for 2004 are almost the same with corresponding numbers of 0.851 and 3.73 %, respectively.

To conclude this analysis, the choice of meteorological year has an impact on the total O_3 column, but the impact on the non-land based traffic induced O_3 and OH perturbations was relatively small. Despite the strong El Niño effect, there is no reason to believe that the choice of 2003 meteorology had a significant impact on our results compared to what would be the case if another year from the 2000s was chosen. A 10-year average would provide a more robust result than 1-year time slices, but based on this simple analysis it does not seem to be worth the tremendous effort required (this only applies when studying effects of emission changes using offline CTMs, and not when studying e.g. effects of changes in climate).



Figure 1. Comparison of the monthly mean ozone observations (black line with dots) from Logan (1999) with the OsloCTM2 model results for 2002, 2003 and 2004 (see legend for color codes) at different latitude bands (from left to right: 90°S-30°S, 30°S-EQ, EQ-30°N, 30°N-90°N) and different pressure levels (from top to bottom: 250 hPa, 500 hPa, 750 hPa).



Figure 2. Yearly averaged total ozone columns (DU) calculated by OsloCTM2 for 2003 (top), and difference in total ozone columns for 2003 – 2002 (bottom left) and 2003 – 2004 (bottom right).

Figure 3. Zonal mean perturbations of ozone (Δ ppbv) in the upper troposphere (300 – 200 hPa) due to a 5% perturbation of aircraft emissions (top) and in the lower troposphere (> 800 hPa) due to a 5% perturbation of ship emissions (bottom) during January (left) and July (right) for the simulations with year 2000 emissions. The left y-axis shows the unscaled impact of the 5% perturbation of aircraft emissions, while the right y-axis is scaled up by a factor 20 from 5% to 100%. Results for January 2002 are not included because they are considered as spin-up.

Page 16810: Line 13: 'have been scaled to 100 %' which regard to what? **Answer:** "Unscaled" means the result due to a 5% emission perturbation (i.e. the direct difference between BASE-AIR and BASE-SHIP), while "scaled to 100%" means that the result due to a 5% emission perturbation has been multiplied by 20 (i.e. 20×(BASE-AIR) and 20×(BASE-SHIP)). This has been made clearer in the revised manuscript.

Model descriptions: Line 22: name the model that was nudge to the meteorological data. Did all the models use 6h meteorological fields? Further, did all models run 2002 for spin-up and 2003 to be analyzed, as noted for TM4? Just describe the common setup in the paragraph ahead and do not repeat this information for the specific models. **Answer:** This has been done. The common model properties are described at the beginning of Section 3 and the individual model descriptions have been harmonized. A systematic comparison of model properties is given in Table 2 in the manuscript.

Ozone: Page 16814, Line 6: briefly describe the approach of Greve 2010. **Answer:** The sentence has been rephrased to clarify how the scaling was done, and one of the findings of Grewe et al. (2010) has been briefly described in the revised manuscript at the end of Section 2, where it is more appropriate.

Effect on aircraft emissions: Page 16815: Line 3ff: it would be helpful if the reader is pointed to the line the authors refer to in the text, for instance: 'the results indicate an increase in ozone impact from aircraft between 2000 (gray solid line) and 2025 (black dotted line).' and so on. Also explain in the text what the red curves represent, to make it easier for the reader (black minus blue)*20, if that is what you show. Answer: The manuscript has been updated with pointers to figure lines where appropriate, and an explanation of what the red curves represent.

Figure 5 etc.: please indicate that the red axis on the right, does not correspond to the red lines in particular but to all lines, if that is what you meant. **Answer:** The figure captions have been modified to clarify that the red axis refer to all lines, but the red lines refer only to the red axis.

Line 10: move 'it depends strongly on the model' before 'whether or not' **Answer:** This has been corrected.

Line 12: Do you refer to the maximum at 25 degrees S. Also, do you mean increased emissions in the tropics based on Figure 2?

Answer: The manuscript has been corrected with maximum at 25 deg S (instead of 30S) and a reference to Figure 3, which clearly shows the aircraft emission increase in the tropics, has been added.

Line 23. Where does this peak in the tropics come from? It seems to exist in 4 of the 6 models (Figure A1). Is this due to transport processes, related to the OH increase in 30N in the upper troposphere? It might help to show and describe the NOx distribution with latitude and altitude.

Answer:

The January peak in the tropics (~20-30N) exists in all 6 models, but does not show up for LMDz-INCA and MOCAGE in Figure A1 because the peak is less pronounced for these two models (and the contour line interval is too large).

The peak in the tropics during winter arise because aircraft-induced NO_X and O₃ are transported from the northern mid-latitude UTLS to the low-latitude middle troposphere where the background levels of NO_X and O₃ are lower (Fig. 4). During summer, the peak does not occur in this region because the vertical exchange is less efficient (illustrated by Hoor et al. (2009), their Fig. 6) and aircraft-induced NO_X and O₃ are then more efficiently transported towards the pole.

An explanation of the peak in the tropics has been given in the revised manuscript. However, in order to limit the number of figures we have chosen not to include the NO_X distribution.

Figure 4. Zonal mean distributions (ppbv) of NO_X (top) and ozone (bottom) as induced from a 5% perturbation of aircraft emissions (left) and in the background (right) during January for the 2050 B1 scenario. In the left figures, solid contour lines show the change relative to the BASE simulation while the dashed line indicates the tropopause.

Page 16816: Line 11, add 'red lines' to 'Fig.5'. Line 25: as mentioned before it might help to add NOx distribution (altitude vs. latitude) to explain this. Further, how is it that higher background levels of CH4 result in more ozone production? Explain.

Answer:

"Red lines" has been added to "Fig. 5". We prefer not to include the NO_X distribution (Fig. 4, top right) in order to maintain brevity of the paper and since we think that the original explanation (including the reference to Fig. 3 in the manuscript) is sufficient. It is well-known that CH_4 is a catalyst for O_3 production due to the formation of RO_2 peroxy radicals, and higher background methane levels could therefore result in more ozone production. However, this explanation has been removed as we have shown above (in the answer to the question on future CH_4 mixing ratios) that changing the background methane levels only had a minor impact on the ozone perturbations.

Page 16817: Line 4: Do you observe the impact of changes in road traffic emissions here?

Answer: The impact of changes in road traffic emissions on NO_X levels in the UTLS is clearly shown in Fig. 5 with a strong decrease from 2000 to 2050 B1. However, as this study does not deal with impacts of road traffic emissions in particular, we choose not to include Fig. 5 in the manuscript.

Figure 5. Zonal mean perturbations (Δ ppbv) of NO_X due to a 5% perturbation of road traffic emissions shown for July 2000 (left) and 2050 B1 (right).

Methane lifetime: Page 16820: Line 17: What is the method described in Hoor 2009? **Answer:** The method used in Hoor et al. (2009) was described in the subsequent sentence, but this was poorly expressed in the original manuscript. The sentence has now been rephrased to avoid confusion.

Page 16821: Line 16: Why speculating about the reason? Do you see a decrease in background NOx?

Answer: Yes, Fig. 6 shows that there is a strong decrease in background NO_X , particularly in Europe and the US. This has been made clearer in the revised manuscript.

Figure 6. Difference in NO_X distributions (Δ ppbv) in the lower troposphere (> 800 hPa) in January (left) and July (right) from 2025 to 2050 for the B1 scenario.

Line 20: remove 'scenarios' (I think you mean the evolution of emissions, and not the evolution of emission scenarios) Answer: This has been corrected in the revised manuscript.

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