Reply to the Review by William Collins (Referee)

Thank you very much for the helpful comments. We revised the text and Figures significantly and took all comments into account. Please find in black the original comments from William Collins and in red our reply.

This study addresses how the impact of aircraft NO_x emissions on ozone and methane vary according to the meteorological conditions. The concept is useful, but a lot more work is needed to present a coherent analysis.

Thank you very much for your review. Your input helped to improve the manuscript significantly. In order to provide a higher consistency within this work, we adapted major parts of the manuscript. Please find below an overview on the changes, based on the referees' comments, which will be discussed in more detail further below:

1. Abstract: The abstract was rephrased to better represent the work and results of this work
2. The introduction was extended to cover the chemical processes and an elaborated discussion why the O_3 maximum (w.r.t. time and magnitude) is selected for the analysis
3. The model description now includes a description of the lagrangain model used
4. Section 3 was eliminated since the chemical system is now covered in the introduction. A subsection was added to the results, discussing the variability of the O_3 maximum and how typical temporal developments of early, mid and late O_3 maxima look like.
5. The altitude analysis was changed to now focus on the mean altitude and not the altitude difference
6. The influence of temperature, NO_x and HO_x in summer and winter are now moved to a single section discussing the O_3 production efficiency
7. In order to account for the rapid cycling between OH and HO_2, we now analyze the OH recycling probability when discussing the CH_4 depletion
8. The discussion now includes a section on how the lagrangain modelling approach influences the results
9. All figures were updated to show the relation for winter and summer and make them visually more appealing

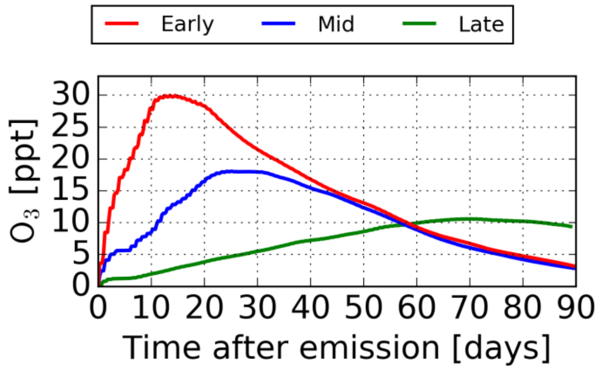
The key variable used seems to be the time of the ozone maximum, but the explanation of why this is chosen is hard to discover. The most obvious variables to use would be the integrated ozone perturbation (i.e. the area under the curve in figure 1). Or the integrated radiative forcing (i.e. integral of O_3 scaled by a radiative efficiency as a function of altitude and latitude). For instance figure 2 shows a correlation between the maximum O_3 concentration and time of the O_3 maximum, but it is not at all clear that one is actually driving the other. Presumably both these are simply identifying regions of high O_3 production efficiency.

In general we agree that the most obvious variable for addressing the climate impact is the integrated ozone perturbation. And this is the starting point of our investigation. The integrated values differ for emission locations. The aim of our paper is to understand these differences in more detail. Hence, identifying the resulting climate impact is not the target of this paper. In this work we are mainly interested in how transport processes affect the resulting ozone and methane change. By using our dataset, we identify that two characteristics (time and magnitude of the ozone maximum) differ most under varying weather conditions. We added an additional figure (Figure 1 in the revised version), a table (Table 1 in the revised version) and an additional paragraph to the introduction clarifying the purpose of this manuscript. In the figure we show that two emission regions next to each other lead to different ozone perturbations, characterized by a different time and magnitude. Since, the emission occurs under different weather conditions (in and west of a high pressure ridge), we open the question if the differences with regard to the ozone perturbations can be explained by the different weather conditions experienced by the air parcels.

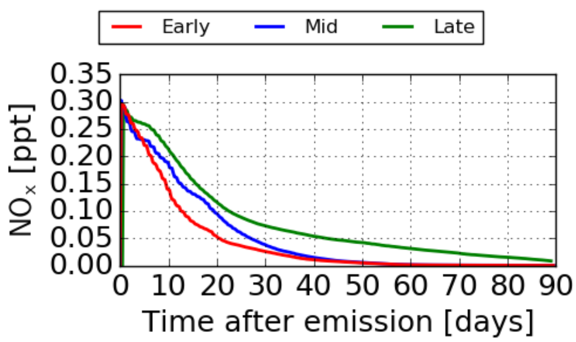
With the revised Figure 2 we want to demonstrate that only if the ozone maximum occurs early a high ozone magnitude is possible but that also early maxima might have only a low maximum. Within our manuscript we demonstrate that early maxima are only possible if the air parcel is transported towards lower altitudes and therefore into regions with a higher chemical activity. The magnitude of the ozone maximum still depends if the air parcel is transported into regions favoring high ozone production efficiency.

It is not at all clear even what the time of the O_3 maximum means at longer timescales. Presumably in these cases the O_3 time series doesn't look like a stretched version of figure 1, but rather a varying timeline that happens to bump up at 40 or 50 days for some meteorological reason. Any NO_x signal will have long dissipated after 20 days(figure 1) so it is not obvious that there is any physical meaning to the later ozone maxima. Example time series for "early" and "late" maxima need to be shown.

In the case of late O_3 maxima the temporal development looks like a stretched version of early maxima. The following figure illustrates the typical development of an early, a mid and a late O_3 maximum:



The corresponding NOx concentration looks as follows:

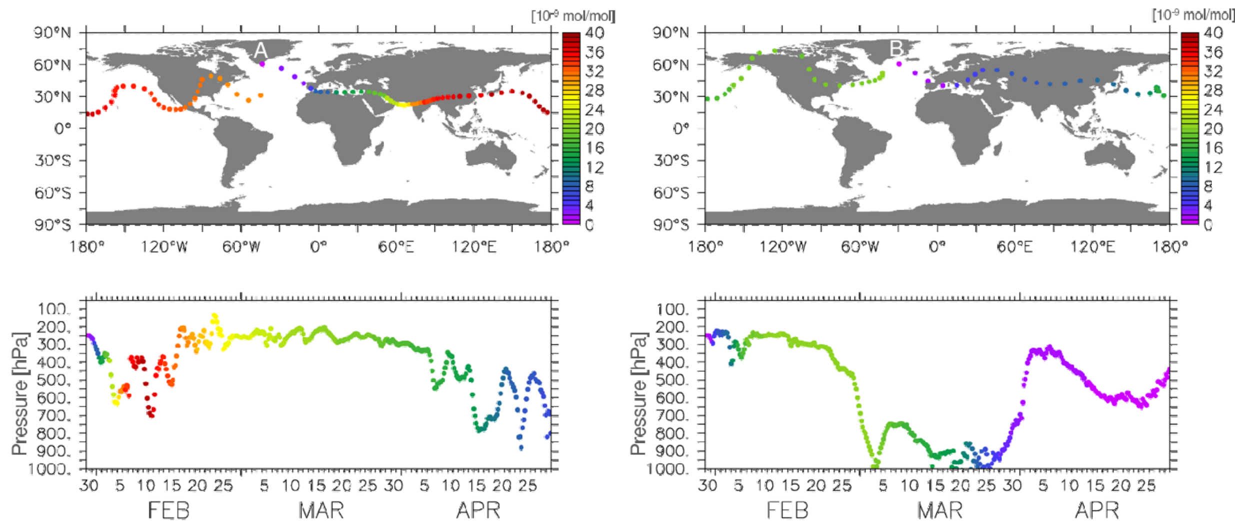


In the case of the late O_3 maximum (green line) NO_x is first reduced mainly due to the formation of HNO_3 (due to high background NO_x concentrations). After 20 days the air parcel is transported towards high latitudes with a largely reduced solar radiation (close to polar night). Due to the reduced solar radiation, only little O_3 is formed over time, leading to a low and late O_3 maximum.

We added the first figure to the revised manuscript (Figure 3) including a discussion on these characteristics.

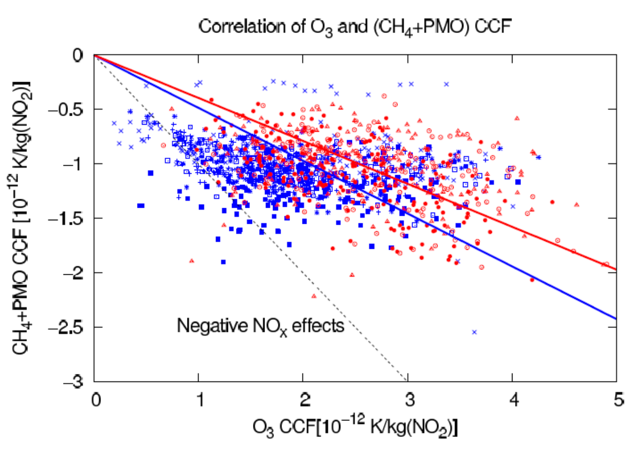
Nearly all the analysis is done for the winter (figs 2, 3, 4, 6, 7) when the effects will be far smaller than in the summer. The magnitude of the winter and summer impacts need to be compared. There is no need to consider the winter at all if it turns out to be unimportant, and certainly the analysis should focus on the summer.

In a companion paper (submitted to ACPD, acp-2020-529) we present the resulting climate impact of all weather situations. In general, the climate impact is indeed higher during summer, however not negligible in winter. In the following figure (a similar figure can be found in the companion paper (their Figure 8)), we show the trajectories of two air parcels released at higher latitudes during winter (marked A and B):



Both air parcels are released close to each other, but only air parcel A is transported towards lower altitudes and into the tropics, leading to a higher climate impact. Obviously the air parcels are released at different location within a weather situation leading to different transport pathways. Hence even if the chemistry is slow at the location of the emission, the transport of the emitted species to tropical regions leads to a significant chemical processing. Therefore there is a clear difference between the seasons. Thus, the ozone contribution from NO_x emissions in winter is not negligible.

The following figure from the companion paper (their Figure 13) gives the climate impact due to changes in ozone and methane for winter (blue) and summer (red):



It becomes evident that the resulting climate impact during winter has a similar variability and therefore similar or even higher re-routing possibilities. Thus, winter can be considered equally interesting for our purpose.

We still see the necessity to show also results for summer and thus modified all figures to show the results and correlations for both seasons.

The variability of the ozone response with respect to the emission location and season has been shown in Frömming et al, the companion paper. Here we are more interested in understanding the differences.

Page 1

The following five comments are all related to the abstract. After receiving your review, it became obvious that the abstract does not represent the intention and the findings of this manuscript adequately. Thanks a lot for the helpful comments! We therefore decided to rephrase major parts of the abstract.

Line 12: It is not at all obvious that the time of maximum should be the controlling factor, rather than the magnitude of the maximum.

You are indeed correct, since the time of the maximum is not the controlling factor. However, only early maxima allow for a high total ozone change. We rephrased this part and also included what we identify as the two major characteristics of the ozone perturbation (time and magnitude of the ozone maximum). See also the discussion above.

Line 13: It is more likely that the subsidence leads to greater ozone production efficiency, and that the earlier ozone maximum is a consequence of this, rather than a cause of it.

In this study we find that the subsidence leads to transport into regions of higher chemical activity. This higher chemical activity then leads to the earlier maximum. We rephrased this section to explain this relation and to better represent the findings of this work.

Line 15: This seems to be stating the obvious – the size of the CH_4 decrease depends only on the size of the CH_4 decrease.

True. We removed this section.

Line 29: Presumably the aim of this study is to identify those meteorological conditions that are conducive to ozone formation so that the computationally expensive chemical trajectories are not needed?

Ultimately yes. This paper tries to be a step towards an improved understanding of the relation between the actual weather situation at the time of emission and the ozone response. We rephrased parts of the abstract and the introduction, to make this more obvious for the reader.

Line 30: It is not explicitly stated that the aim is to avoid producing ozone, but to enhance the destruction of methane.

The re-routing approach presented in Grewe et al. 2014a and Grewe et al. 2014b performs the optimization based the total climate impact from NO_x. Regions with a high overall climate impact are avoided and regions with a low climate impact are favored, independent if the lower climate impact is caused by less ozone being produced or more methane being depleted. You are raising an important discussion, which has been addressed in another paper: Grewe et al. 2017:

“In our approach, the routes which reduce the climate impact avoid regions where warming contrails are formed or the ozone impact is large. However, routes are also favored, where contrails contribute to cooling or the emission of NO_x leads to a methane reduction which cools more than the increase in ozone warms. This raises the question, to what extent should additional contrail formation be allowed, which—over a chosen time span—cools the global climate more than the additional CO_2 emitted by climate optimized routing warms. These questions have to be considered carefully for any climate-optimized routing.”

We think this paper is not the right forum to further discuss this, here we are focusing on the understanding of the atmospheric processes, whereas any application of climate-optimized routing must address these issues. We do not think that there is a clear scientific answer to the question, it might be more a decision by policy makers and society guided by science.

Grewe, V., Frömming, C., Matthes, S., Brinkop, S., Ponater, M., Dietmüller, S., Jöckel, P., Garny, H., Tsati, E., Dahlmann, K., Søvde, O. A., Fuglestvedt, J., Berntsen, T. K., Shine, K. P., Irvine, E. A., Champougny, T., and Hullah, P.: Aircraft routing with minimal climate impact: the REACT4C climate cost function modelling approach (V1.0), Geosci. Model Dev., 7, 175–201, https://doi.org/10.5194/gmd-7-175-2014, 2014a.

Grewe, V.; Champougny, T.; Matthes, S.; Frömming, C.; Brinkop, S.; Sø vde, A.; Irvine, E.; Halscheidt, L.Reduction of the air traffic’s contribution to climate change: A REACT4C case study. Atmos. Environ. 94, 616–625, 2014b.

Grewe, V., Matthes, S., Frömming, C., Brinkop, S., Jöckel, P., Gierens, K., Champougny, T., Fuglestvedt, J., Haslerud, A., Irvine, E., Shine, K., Climate-optimized air traffic routing for trans-Atlantic flights. Environm. Res. Lett. 12(3), 034003, DOI: 10.1088/1748-9326/aa5ba0, 2017.

Page 2

Line 12: It needs to be made clear in these sentences whether the climate impact is warming or cooling. It would help to contrast the effects on ozone and methane.

These are crucial information for the reader. We added two sentences covering this information.

Page 4

Line 4: The method for calculating the trajectories needs to be described. How do they account for sub-grid scale vertical motion?

We added an elaborate explanation of the submodel (ATTILA) which is used to model the transport of each air parcel to the base model description. The sub-grid scale vertical motion in ATTILA is calculated in three steps. First, mapping the ATTILA tracer concentrations from the air parcels to the EMAC grid. Second, calculating the convective mass fluxes similarly as for standard EMAC tracers. Third, mapping the calculated tendencies back to the air parcels.

Page 6

Figure 1: Is this figure a change in the global burden? It would be useful to show changes along a trajectory since that is what is used in all subsequent figures.

In this figure the change in the global burden was shown. However, this figure has been removed in favor of the new figure added in the introduction (discussed earlier). This figure covers the same information for different positions. To be able to compare the results to the resulting climate impact the change in the global burden needs to be shown. However, figure 3 shows an early, a mid and a late O_3 maximum along the trajectories.

Page 7

Figure 2: How well is the “Time of the O_3 maximum” defined? It might be that after 20 days there is no well-defined peak, but rather fluctuations of greater or lesser magnitude.

We defined the O_3 maximum such that it is the highest O_3 concentration with no further increase of O_3 afterwards. Information about this was added to the manuscript.

Line 5: Why is it assumed that the early maximum is the cause? It could just as easily be written that an early O_3 maximum is only possible if the concentration change is high.

You are right that the earlier O_3 maximum is only possible if the O_3 production is high. In this study we do not assume that the earlier maximum is the cause for the higher O3 maximum. Instead we just state that high O_3 maxima are possible if the O_3 maximum is reached early. This is of course only possible if the production efficiency is high (see Section 3.3).

Line 7: The processes involved here need to be understood. It could be that higher altitude emissions don’t produce much ozone, so that any fluctuations in ozone appear as spurious “late” maxima. The time series for these late maxima need to be shown.

Please see our earlier answer on this matter. A graphic showing representative examples for early, mid and late maxima was added to the manuscript.

Line 9-13: The RF or CCF is not mentioned again in this study. It appears they come from other work with the REACT4C project. Unless these can be related to the case studies analysed here it is not helpful to discuss them. For example the comments on Lacis et al. (1990) refer to a higher radiative efficiency at altitude in contrast to the lower ozone production efficiency found in this study. Why do the time and magnitude of the O_3 maximum influence the climate impact? Instead it seems it should be the integral of the ozone perturbation with a radiative efficiency factor for latitude and altitude. What is CCF and how is it determined?

The RF and CCF are indeed from other works within REACT4C. An elaborate explanation on how the CCFs are calculated, including a validation/verification, is presented in Grewe et al. (2014) and a more detailed analysis of the CCFs is given in more detail in our companion manuscript (acp-2020-529). Within this study, we are concentrating on a specific aspect of the CCFs and we found that early and high O_3 maxima relate well to high RF and CCF values. The introduction was extended, to increase the understanding of this relation.

Grewe, V., Frömming, C., Matthes, S., Brinkop, S., Ponater, M., Dietmüller, S., Jöckel, P., Garny, H., Tsati, E., Dahlmann, K., Søvde, O. A., Fuglestvedt, J., Berntsen, T. K., Shine, K. P., Irvine, E. A., Champougny, T., and Hullah, P.: Aircraft routing with minimal climate impact: the REACT4C climate cost function modelling approach (V1.0), Geosci. Model Dev., 7, 175–201, https://doi.org/10.5194/gmd-7-175-2014, 2014.

Page 8

Figure 3: The caption says the analysis is based on the first seven days after emission, but the figures show values out to 90 days.

The Figure 3 bottom shows the mean latitude of the air parcel for the first seven days after emission in relation to the time when the ozone maximum occur. We rephrased this caption to make clearer that the seven days are only related to the mean latitude and not the time of the ozone maximum. We used seven days for the latitudinal mean since the first ozone maxima occur after seven days. Using only seven days reduces the potential bias of air parcels with a late maximum.

Page 9

Line 2-3: It is not obvious why the altitude difference is the crucial variable, rather than the absolute altitude. The discussion makes plausible arguments about increased ozone production at lower altitudes, therefore it would seems more logical to plot the altitudes where the trajectory ends up (maybe the mean altitude in the first 20 days),whereas there doesn’t seem to be any argument that it is the amount of descent that is important. Except obviously that if the emissions all occur at similar flight levels then greater descent will give lower trajectories.

We agree that analyzing the absolute altitude makes more sense in the scope of this analysis. Therefore, the analysis was changed accordingly. However, instead of 20 days we only use the first seven days for calculating the mean altitude (see earlier explanation).

Line 5-6: The use of the time of maximum as the controlling variable is not obvious. For instance the claim that the earlier maxima in summer give less time for downward transport is much more likely to be due to the enhanced photochemistry in the summer giving more ozone production at higher altitudes, hence for a descending trajectory the maximum will occur earlier.

Agreed, the time of maximum is not the controlling factor, but an important piece of information in understanding the variations in the ozone response to a NO_x emission (see also discussion above). Thank you very much for pointing this out. We checked the data and we find the same relation and added a section covering this relation to the analysis.

Line 8: Rather than focussing on the early ozone maximum, it would be more scientifically rigorous to state that significant ozone production only occurs if an air parcel is transported to lower altitudes and latitudes.

We agree on this. The text has been changed accordingly. In addition, we restructured the original Section 4.2 and 4.3 and merged it into a single Section 3.3 focusing on the O_3 production efficiency.

Page 10

Line 2: To what extent is sub-grid scale convection included in the trajectory calculations?

We addressed this in an earlier answer above.

Lines 14-24: In section 3 the argument was that earlier O_3 maxima lead to greater ozone production. But here the opposite argument is being made – that increased ozone destruction leads to earlier maxima. In which case the early O_3 maxima should be associated with less ozone not more. This is another example of why the time of the O_3 maxima should not be used as a controlling variable. Note also that while reactions R2 to R4 might have negative temperature dependencies the origin of the HO_2 andRO_2 has strong positive temperature dependence, so higher temperatures do lead to more ozone production.

Thank you for pointing this out. The temperature relations are of course correct as you described them. With the new structure of Section 3.3 (focusing on O_3 production efficiency instead of temperature, NO_x and HO_x separately) this part is no longer necessary and was thus removed. In this study, the time of the O_3 maximum is not the controlling factor of the resulting climate impact but rather a typical characteristic of the temporal development of O_3. In general, the production phase is more interesting for this study, since the ultimate goal is to have short simulations to predict the climate impact from aviation and allow for efficient re-routing. Using the 90 days integral is thus not feasible. An elaborate discussion on this manner was added to the introduction.

Page 12

Line 2: This sentence doesn't seem correct. Do you mean to correlate NO_x with ozone maxima?

We understand this confusion - apologies. In this sentence we intended to refer to the correlation between the mean NO_x concentration and the magnitude of the ozone maximum. We updated the description of the relation in the new section accordingly.

Page 15

Lines 10-14: This study uses prescribed emissions of NO_x (5x10ˆ5 kg) so there doesn't seem any value in comparing this to NO_x concentrations in an aircraft study. I suggest removing this paragraph.

A direct comparison of our results to other studies is complicated, since our approach was not used by any other modelling study yet. However, in Søvde et al. 2014 EMAC (our base model used) was compared to other models focusing on the impact of aviation emissions. In this part of the discussion, we focus on how well EMAC compares to other models and find that it compares reasonably well. We think that this is information is important and thus decided to keep it within our discussion.

Søvde, O. A., Matthes, S., Skowron, A., Iachetti, D., Lim, L., Owen, B., Øivind Hodnebrog, Genova, G. D., Pitari, G., Lee, D. S., Myhre,G., and Isaksen, I. S.: Aircraft emission mitigation by changing route altitude: A multi-model estimate of aircraftNOxemission impactonO3photochemistry, Atmospheric Environment, 95, 468 – 479, https://doi.org/https://doi.org/10.1016/j.atmosenv.2014.06.049, 2014

Line 32: Stevenson and Derwent only analysed summer as ozone production and methane depletion are not important in winter.

This is correct. However, analyzing the importance of weather conditions in winter is crucial, due to the high variability in the resulting climate impact and the possibility that air parcel are transported to equatorial regions during winter (see earlier discussion).

Page 16:

Line 14: There has been no calculation of "resulting climate impact" in this study, therefore it is not clear how this can be a conclusion from this work.

That is indeed correct. We changed the manuscript accordingly.