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

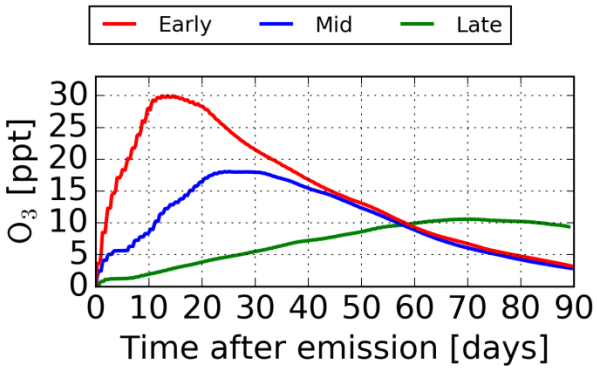
driving the other. Presumably both these are simply identifying regions of high O_3 production efficiency.

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.

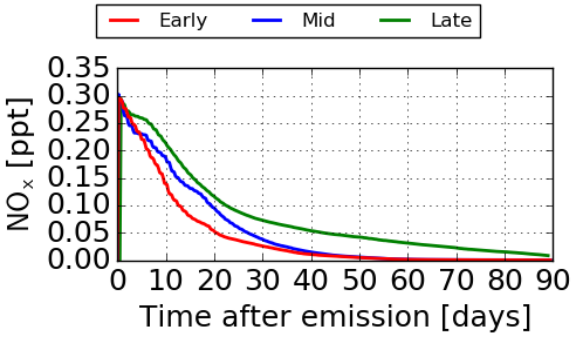
With the revised Figure 2 we want to demonstrate that only if the ozone maximum occurs early a high ozone magnitude is possible but that also early maxima might have only a low maximum. Within our manuscript we demonstrate that early maxima are only possible if the air parcel is transported towards lower 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:

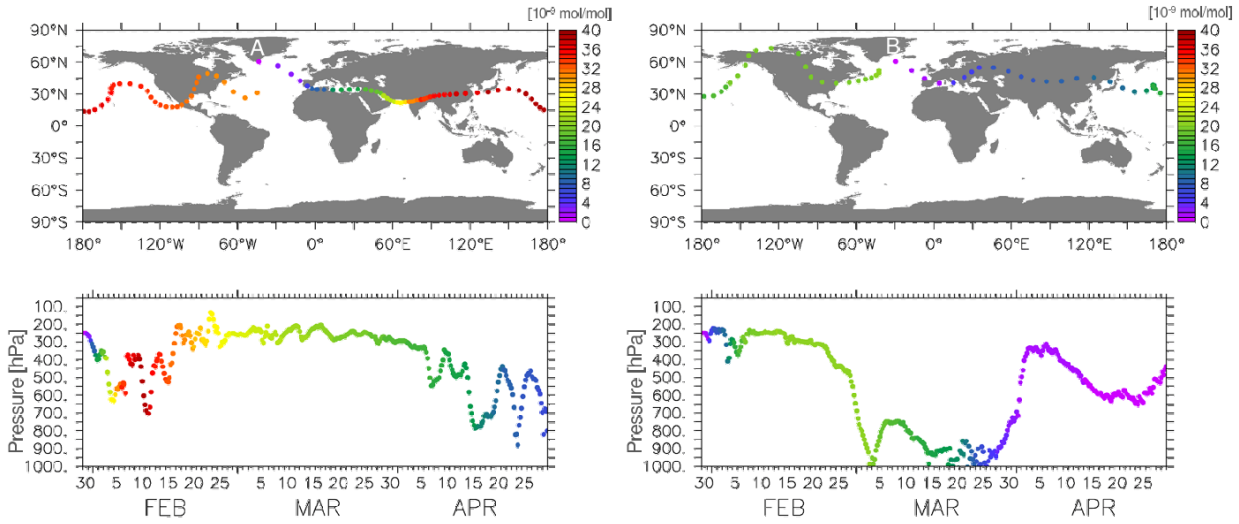


In the case of the late O_3 maximum (green line) NO_x is first reduced mainly due to the formation of HNO_3 (due to high background NO_x concentrations). After 20 days the air parcel is transported towards high latitudes with a largely reduced solar radiation (close to polar night). Due to the reduced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

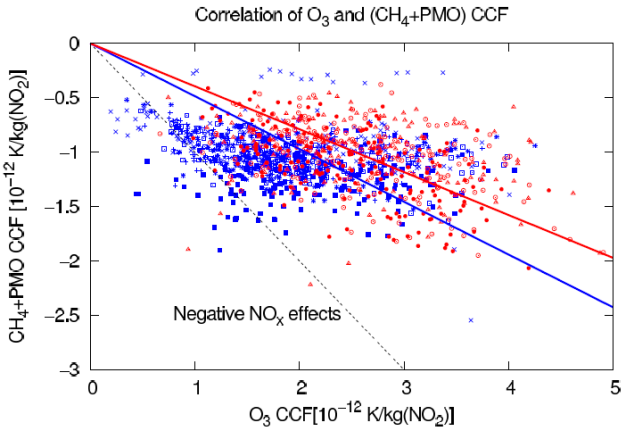
Nearly all the analysis is done for the winter (figs 2, 3, 4, 6, 7) when the effects will be far smaller than in the summer. The magnitude of the winter and summer impacts need to be compared. There is no need to consider the winter at all if it turns out to be unimportant, and certainly the analysis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

Reply to the review by the Anonymous Referee #2

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 reviewer #2 and in red our reply.

This work aims to provide insight into the effect that aviation NO_x has on the climate through its contribution to tropospheric ozone and depletion of atmospheric methane. In particular, the authors aim to show how the specific weather pattern into which the emission occurs could affect the long-term chemical impact. They evaluate this using a trajectory-tracking approach with simplified tropospheric chemistry, embedded within a global chemistry-climate simulation. Using the time and magnitude of the maximum change in ozone as proxies for the total ozone-related climate impact, they find the overall radiative forcing is likely to change significantly depending on the season and local meteorological conditions.

The central question of the paper is both interesting and important. If robust, this work could be a significant advance in the field of understanding the interactions between aviation and the atmosphere. It could also provide valuable, actionable information on how to reduce aviation's climate impacts. However, I have concerns regarding the methodological approach, and regarding the metric used to quantify "climate impact".

As such, I believe that major revisions are needed before this paper is ready for publication in ACP. I list major concerns below first, followed by minor issues.

Thank you very much for your review and seeing the value of our work to the community. 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

Major comments

While innovative, it is not clear to me that the chemistry and physics simulated within the "tracked" air parcel will provide an estimate of aviation's impacts on the atmosphere which are accurate or consistent with the "parent" EMAC model.

1. Is it validated against any estimates using more conventional techniques? The authors cite Grewe et al (2017c) Section 4.3, but this does not appear to have an explicit comparison of the Lagrangian model's result for aviation impacts compared too ther studies. A direct comparison showing (e.g.) the ozone and methane response in EMAC when performing a conventional simulation of an increment in aviation NOx would be very helpful. Alternatively, if such a comparison is already present in (eg) Grewe et al (2017c), a quantitative evaluation (e.g. "agreement to within X% when simulating aviation emissions") would be helpful. If not already present in Grewe et al2017c I recommend that such an analysis be added to Section 5.

Thank you for pointing this out. The concept used in this study is designed to gain more insights in aviation effects, which conventionally can otherwise not be obtained, limiting the comparability to conventional approaches. However, the following four indicators support the consistency of the modelling approach:

1. The transport scheme is reasonably well established (Brinkop and Jöckel, 2019).
2. The chemical response to a local emission agrees well with earlier findings of Stevenson et al. (2004), who simulated the monthly mean response of ozone and methane to a NO_x pulse and which are very similar to the results from this approach (Grewe et al., 2014, their Fig. 9).
3. The use of a trajectory analysis to interpret either observational data or modelling data is well established.
4. A first verification of the resulting global pattern of the atmospheric sensitivity to a local NO_x emission by comparing to sparsely available literature data was promising (Yin et al., 2018).

We added an elaborated discussion on this matter to the discussion section of our manuscript.

Brinkop, S. and Jöckel, P.: ATTILA 4.0: Lagrangian advective and convective transport of passive tracers within the ECHAM5/MESSy (2.53.0) chemistry–climate model, Geosci. Model Dev., 12, 1991–2008, https://doi.org/10.5194/gmd-12-1991-2019, 2019.

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.

Stevenson, D. S., Doherty, R. M., Sanderson, M. G., Collins, W. J., Johnson, C. E., and Derwent, R. G.: Radiative forcing from aircraft NOx emissions: Mechanisms and seasonal dependence, Journal of Geophysical Research: Atmospheres, 109, https://doi.org/10.1029/2004JD004759, 2004.

Yin, F., Grewe, V., Frömming, C., & Yamashita, H.: Impact on flight trajectory characteristics when avoiding the formation of persistent contrails for transatlantic flights, Transportation Research Part D: Transport and Environment, 65, 466 – 484, doi: https://doi.org/10.1016/j.trd.2018.09.017, 2018

2. More detail on the Lagrangian model would be helpful. For example, what is the total air mass of the well-mixed box? How is diffusion treated? This is important because of the role of non-linear chemistry (see e.g. Kraabøl et al 2002), which could result in suppressed ozone production when concentrations are very high (i.e. early in the plume's development).

We agree that more information on the Lagrangian model is needed such that the reader understands how the transport processes (identified to be important for the resulting ozone and methane change) are simulated. Therefore, a detailed description of the submodel ATTILA was added to the base model description. Note also that the Lagrangian transport scheme has been evaluated in detail by Brinkop and Jöckel (2019).

Brinkop, S. and Jöckel, P.: ATTILA 4.0: Lagrangian advective and convective transport of passive tracers within the ECHAM5/MESSy (2.53.0) chemistry–climate model, Geosci. Model Dev., 12, 1991–2008, https://doi.org/10.5194/gmd-12-1991-2019, 2019.

The approach used appears to quantify the direct effect that aviation NO_x and H_2O could have on the climate through increases in short-term ozone and decreases in methane, on the basis that both are greenhouse gases. However, one of the major effects of aviation NO_x is a long-term reduction in tropospheric ozone, driven by the methane loss (see e.g. Holmes et al 2001). Is this accounted for here? If I have understood the trajectory-tracking mechanism correctly it does not include feedbacks, so I would not expect it to be accounted

for. If so, it would be useful to include an estimate of the total magnitude of the missing long-term ozone loss.

This effect is known as primary model ozone (PMO). Within REACT4C, PMO was not explicitly simulated, as in most other studies, since this change in ozone occurs far beyond the 90 days simulated. Simulating it explicitly would be too computationally expensive and unfeasible for this modelling approach. Instead a constant scaling factor was applied to the climate impact due to changes in methane, to account for PMO. Due to this simplified approach, PMO cannot be taken into account in this study. We added a paragraph to section 2.3 to explain these circumstances.

The analysis is predicated on the idea that the time and magnitude of the peak change in ozone is a significant indicator of overall climate impacts. However, it is not clear to me that this is the case, and I could not find a quantitative justification or citation to this effect in the paper. The closest I found was the assertion that, in REACT4C, a higher ozone concentration change "generally" leads to a larger RF (p7, line 12). Why use these metrics instead of (e.g.) the total integrated ozone perturbation over the time of the simulation?

In general, we agree that the most obvious variable for addressing the climate impact is the integrated ozone perturbation or the resulting RF. We have realized that our introduction guided both reviewers into a wrong direction on the objective of this paper. Apologies for that. Identifying the resulting climate impact is not the objective of this paper. In this work, we are mainly interested in how transport processes effect 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. To improve the introduction and to avoid such a misinterpretation, 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 these differences can be explained by the different weather conditions experienced by the air parcels.

At the end of section 2.1, it is stated that "50 trajectories [are initialized] at 6, 12 and18 UTC", but this is then immediately followed by "in the present study, only 12 UTC is considered". I'm confused – why have the first statement? Also, what is the error which we can expect from only including one time point? Won't this result in same geographical biases?

6, 12 and 18 UTC was only simulated for WP1 within REACT4C. Grewe et al. 2014 demonstrated that the results are more sensitive to the horizontal than the temporal representation. It was thus decided to only simulate 12 UTC for all other weather patterns, due to limited computational resources. We therefore can only include 12 UTC within this study. We updated this paragraph to include this additional information.

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.

Many of the conclusions seem to treat correlations as causal links (e.g. section 4.3).Some conclusions – such as that "during summer the O_3 formation is limited by the background NO_x concentration, whereas in winter low HO_2 concentrations limit the total O_3 gained" – do not seem to be sufficiently supported by the data. It would be helpful to see a more explicit justification for why this must be the mechanism.

In order to approach this, we performed major parts of the analysis again. In addition, parts of Section 4.1, 4.2 and 4.3 were merged into a single section (now Section 3.3). In this section we now discuss which factors influence the efficient production of O3. In the case of the O_3-NO_x-HO_x relation, NO_x and HO_x are analyzed in parallel. The mechanistic why NO_x and HO_x limit the production of O_3 in summer and winter was added.

More generally, much of the analysis is not very quantitative – such as page 14, lines 3-5which states that a visual inspection of a correlation makes it "evident" that winter pat-tern 5 looks "almost the same" as for summer pattern 3. I strongly recommend that the authors redo this analysis in a more quantitative fashion and remove conclusions which cannot be both quantified and mechanistically explained in a way which is supported by data.

Thank you very much for pointing this out. In general, we agree with the comment. Some of the analysis is not obvious to the reader, based on the figures provided. We therefore expanded most of the analysis performed in this study and added results for summer and winter. In this particular case, the distribution of HO_x is now presented for WP1, WP5 and SP3 in an additional figure. The reason why HO_x is not the limiting factor for WP5 is now addressed by using this figure and mean and median values.

Some of the analysis used for chemistry is difficult to interpret, and seems to ignore the cycling nature of certain key species. For example, on page 13, it is stated that "35%and 42% of background OH is produced by HO_2 reacting with O_3 and NO, respectively". However, OH and HO_2 are expected to be cycling rapidly. As such, "production" of OH in this fashion is hard to interpret, since it usually matters more to consider what the sources of HO_x are. I am therefore skeptical of the claim that H_2O is only a minor source of OH. I recommend that the authors rephrase this discussion to be about the OH/HO_2 ratio and the production and loss of HO_x than to talk separately about OH and HO_2.

This is a good point and an important fact missing in our analysis so far. Two measures were performed to account for the cycling between OH and HO_2. In Section 3.3 (earlier Section 4.3) the analysis was performed with respect to HO_x instead of only HO_2.

Additionally, Section 3.4 (earlier Section 4.4) was completely redone. In order to account for the cycling between both species, we now analyze the OH recycling probability.

Minor issues

P2 l22: I went back and checked the claim that Gilmore et al (2013) showed that "during summer the climate impact is up to 1.5 times higher and only half in winter, when compared to the annual meaning". I do not think this is true. They did show that the ozone production efficiency was 50% higher than the average in summer, but this is largely compensated by changes in ozone lifetime. The overall change in ozone production rate is only about 10% above the annual mean in summer, and correspondingly about 10% below the mean in winter (see Figure 1 of said paper).

This is indeed correct. Thank you for pointing this out. We intended to write about the O_3 production efficiency which is about 50% higher in summer. We changed this section in the manuscript to represent their findings appropriately.

The claim that "the standard deviation of background concentrations [of ozone and NO_x] are generally considered to be higher than changes induced by aviation . . . making them hardly detectable" (p16 l30, and paraphrased on page 14, lines 17-19) is based on a single, outdated study. Wauben et al 1997 uses a 1995 aircraft NO_x inventory, now 20-25 years old. Since total aviation NO_x emissions have likely more than doubled since then (e.g. Wasiuk et al 2016), I think this claim either needs a more recent and robust backing or it should be removed.

We could not find a more recent peer-reviewed study addressing this problem. Based on their findings we still think that even a doubling of the NO_x emissions will not be detectable on a global scale and that their statement is still valid. We therefore keep this part within the discussion.

The manuscript has several spelling and grammar mistakes. For example, in several locations (e.g. p2, l34) the authors use the word "adopt" when I suspect "adapt" is intended, and on p12 line 19 the word "exited" should be "excited". I would recommend another sweep through the manuscript to fix these and other typos.

The errors pointed out are changed in the manuscript. In addition, the manuscript was checked for further errors of this type.

What happened to WP2 in Table 1?

WP2 was not taken into account in our analysis due to technical problems. These problems lead to an incomplete dataset. We added this information to section 2.3.

The impact of weather pattern and related transport processes on aviation's contribution to ozone and methane concentrations from NO_x emissions

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Abstract. Aviation attributed climate impact depends on a combination of composition changes in trace gases due to emissions of carbon dioxide (CO_2) and non-CO_2 species. Nitrogen ~~oxide~~ oxides (NO_x = NO + NO_2) emissions ~~lead to~~ induce an increase in ozone (O_3) and a depletion of methane (CH_4) ~~, whereas water vapour () can additionally lead to the formation of persistent contrails~~leading to a climate warming and a cooling, respectively. In comparison to CO_2, non-CO_2 contributions to the atmospheric composition are short lived and are thus characterised by a high spatial and temporal variability. In this study, we investigate the influence of weather ~~pattern~~ patterns and their related transport processes on composition changes caused by aviation attributed NO_x emissions~~,~~. This is achieved by using the atmospheric chemistry model EMAC (ECHAM/MESSy). Representative weather situations ~~are~~ were simulated in which unit NO_x emissions are initialised in specific air parcels at typical flight altitudes over the North Atlantic flight sector. By explicitly calculating ~~composition changes~~ contributions to the O_3 and CH_4 concentrations induced by these emissions, interactions between trace gas composition changes and weather conditions along the trajectory of each air parcel are investigated.

~~The resulting climate impact from~~ Previous studies showed a clear correlation between the prevailing weather situation at the time when the NO_x ~~via changes of~~ emission occurs and the climate impact of that NO_x emission. Here, we show that the aviation NO_x contribution to ozone is characterised by the time and magnitude of its maximum and demonstrate that a high O_3 ~~mainly depends on the magnitude of the maximum induced composition change. In general, the earlier the maximum change occurs the larger the total change and therefore the resulting climate impact. In this study we show that~~ maximum is only possible if the maximum occurs early after the emission. Early maxima occur only if the air parcel, in which the NO_x emission occurred, is transported to lower altitudes, where the chemical activity is high. This downward transport is caused by subsidence in high pressure systems~~leads to an earlier maximum and that the maximum change is~~ . A high ozone magnitude only occurs if the air parcel is transported downward into a region in which the ozone production is efficient. This efficiency is limited by atmospheric NO_x and HO_x concentrations during summer and winter, respectively. ~~The resulting climate impact due to composition changes of depends only on the magnitude of the induced depletion of , where a larger depletion of leads to a larger cooling effect.~~ We show that a large CH_4 depletion is only possible if a strong formation of O_3 occurs ~~and if large~~

due to the NO_x emission and if high atmospheric H_2O concentrations are present ~~.~~along the air parcel's trajectory. Only air parcels which are transported into tropical areas, due to high pressure systems, experience high concentrations of H_2O and thus a large CH_4 depletion.

~~Re-routing flight trajectories based on the experimental setup used in this study is currently too computationally expensive. This work demonstrates that transport processes are of most interest when identifying~~ Thus, the controlling factor to identify the climate impact from aviation NO_x emissions ~~. The insights gained~~ are transport processes. Avoiding climate sensitive areas by re-routing aircraft flight tracks is currently computationally not feasible due to the long chemical simulations needed. The findings in this study ~~suggest an approach to re-route flightsin the future, by performing less computationally expensive purely dynamic simulations.~~ comprises a step towards a climate impact assessment of individual flights, here with the contribution of aviation NO_x emissions to climate change, ultimately enabling routings with a lower climate impact by avoiding climate-sensitive regions.

1 Introduction

The importance of anthropogenic climate change has been well established since years (Shine et al., 1990) and it is well known that air traffic contributes substantially to the total anthropogenic climate change (Lee et al., 2009; Brasseur et al., 2016; Grewe et al., 2017a). A major fraction of its contribution comes from non-CO_2 emissions which lead to changes in greenhouse gas concentrations as well as contrail and contrail-cirrus formation in the atmosphere (Kärcher, 2018). The climate impact of CO_2 is mainly characterised by the emissions strength, due to its long lifetime. However, non-CO_2 effects are known to be characterised by a high spatial and temporal variability. This implies that the total contribution to concentrations of non-CO_2 emissions is not only influenced by the emissions strength but also by the time and location of the emission itself.

~~The emission of nitrogen~~ Nitrogen oxides (NO_x = NO + NO_2)~~leads to changes in ozone and methane concentrations.~~ , emitted in the upper troposphere, lead to a formation of ozone (O_3) following a catalytic reaction. NO reacts with HO_2 forming NO_2. Via photodissociation, NO_2 forms $O(^3P)$ leading to the formation of O_3.

$$HO_2 + NO \rightarrow OH + NO_2 \quad \text{(R1)}$$

$$NO_2 + h\nu(\lambda \leq 410nm) \rightarrow NO + O(^3P) \quad \text{(R2)}$$

$$O(^3P) + O_2 + M \rightarrow O_3 + M \quad \text{(R3)}$$

The additionally formed OH leads to an oxidation of CH_4:

$$CH_4 + OH \rightarrow CH_3 + H_2O \quad \text{(R4)}$$

Additional concentration changes in ozone are introduced by changes to the precursor methane, known as primary mode ozone (PMO) (Wild et al., 2001). Earlier studies already identified that the climate impact resulting from aviation attributed NO_x emissions varies strongly within the atmosphere. In general, the increase in O_3 has a warming effect whereas the depletion of CH_4 leads to cooling effect. The warming caused by O_3 out weights the cooling via CH_4 leading to an overall warming due to

Table 1. The integrated O_3 and CH_4 contribution to atmospheric concentration for both locations given in Fig. 1. Additionally, the resulting climate impact for both locations is given. For further details on the climate impact see Frömming et al. (2020). Integrals are given in kg days and Climate-Cost-Functions (CCF) are given in K $kg(N)^{-1}$.

-	45°N 45°W	45°N 30 °W
Integral O_3	469.6×10^7	600.3×10^7
Integral CH_4	-99.9×10^7	-158.5×10^7
CCF O_3	1.65×10^{-12}	2.31×10^{-12}
CCF CH_4	-9.19×10^{-13}	-9.56×10^{-13}
CCF PMO	-2.67×10^{-13}	-2.78×10^{-13}

aviation attributed NO_x emissions (Lee et al., 2009; Grewe et al., 2019). Köhler et al. (2013) showed that the climate impact is larger for emissions occurring in lower than in higher latitudes. A larger climate impact also occurs in regions with low aviation activity (such as in low altitudes and latitudes) for the same amount of NO_x. Both can be explained by higher incoming solar radiation and lower background NO_x concentrations in those regions compared to higher latitudes. A similar impact was identified by Stevenson and Derwent (2009). The general lower background NO_x concentration in the Southern Hemisphere (SH) compared to the Northern Hemisphere (NH) also explains the inter-hemispheric discrepancy of the resulting climate impact from NO_x emissions. In the SH the climate impact from the same amount of NO_x is generally larger. Köhler et al. (2008) identified that the emission altitude strongly influences the resulting climate impact~~and~~ , which is generally larger for emissions at high altitudes. Frömming et al. (2012) demonstrated that the overall climate impact can be reduced by ~~adopting~~ adapting flight altitudes, suggesting a possible mitigation strategy. The season in which the emission occurs influences the resulting climate impact in addition to the emission location. Gilmore et al. (2013) identified that during summer the ~~climate impact is up to 1.5 times higher and only half~~ production of O_3 is about 50 % higher in summer and 40 % lower in winter, when compared to the annual mean. Changes in CH_4 lifetime compensate parts of this higher O_3 production, leading only to a 10 % higher climate impact. In winter, the resulting climate impact is about 10 % lower. Grewe et al. (2017a) and Frömming et al. (2020) demonstrated that the total change in ozone is larger if the NO_x emission occurred within a high pressure ridge compared to emissions occurring west of this blocking condition. ~~This shows that the impact of aviation emissions depends on weather situation at the time of emission~~

Figure 1 shows the "typical" temporal development of O_3 and ~~along the trajectory.Further investigations are necessary to improve the understanding of those emissions to provide possible mitigation strategies in~~ CH_4 due to an aviation attributed NO_x emission for two emission locations next to each other (45°N 45°W and 45°N 30°W), representative for the examples presented in Grewe et al. (2017a) and Frömming et al. (2020). In exact, one emission region is within and the ~~future.~~

~~In~~ other one is west of a high pressure ridge (see top panel in Fig. 1). NO_x is reduced almost exponentially and washed out after about a month. While the emitted NO_x decreases, the O_3 concentration increases due to the described production

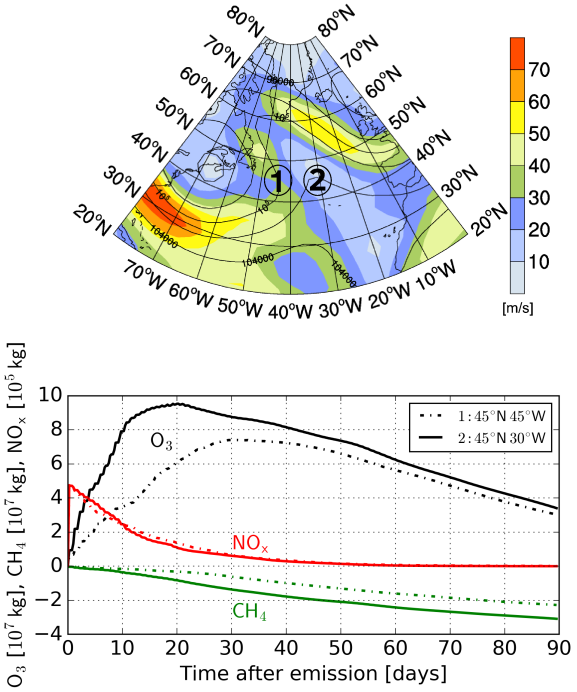

Figure 1. Weather conditions at time of emission (top) represented by geopotential height (black contours in gpm) and wind velocities (see colorbar, in $\mathrm{m\,s^{-1}}$) and the composition change in O_3 and CH_4 induced by the emitted NO_x at two emission locations (bottom).

processes (Reaction R1 - R3). When the NO_x mixing ratio is below a certain level, only little O_3 is produced and loss terms dominate the O_3 chemistry leading to a continuous decrease in the formed O_3. At the same time, the additional O_3 and NO_x form OH leading to a depletion of CH_4 (Reaction R4). After all NO_x and O_3 is lost, the negative CH_4 anomaly starts to decay and will later reach its original values (not seen in Figure 1). For these two emission regions, the resulting O_3 gain differs, with the one emitted in the high pressure system having an earlier O_3 maximum with a higher magnitude. In contrast, the CH_4 depletion is only characterised by a varying magnitude. This large variability in the NO_x-O_3-CH_4 relation, induced by the same NO_x emission, is also presented in Fig. 9 of Grewe et al. (2014). The question remains if the different resulting characteristics for both emission locations given in Fig. 1 can be explained by the the different weather conditions experienced by each air parcel.

Within the present study, we investigate the impact of weather situations on changes in ozone and methane concentrations induced by NO_x emissions in the upper troposphere, emitted over the North Atlantic flight sector. ~~Here, special focus~~ In general, one tends to analyse the integrated O_3 change or the integrated radiative forcing due to these changes in O_3. Table 1 gives the integrated O_3 and CH_4 for both regions (see Fig. 1). Additionally, the so called Climate Change Functions (CCFs), a measure on how the Earth surface temperature will change due to a locally restricted NO_x emission (for more details see Grewe et al. (2014) and Frömming et al. (2020)), is given. It becomes obvious that an earlier and larger O_3 change

leads to a higher integrated O_3 and a higher climate impact. Analysing the integrated O_3 and CH_4 is not feasible when analysing the influence of weather conditions on the induced composition changes due to aviation attributed NO_x emissions. Comparing varying weather conditions to a single data point (e.g. the integrated O_3) is difficult, due to the chaotic nature of weather conditions. Typical characteristics of the temporal development of O_3 and CH_4 are more suitable for this analysis since it is expected that they are directly influenced by varying weather conditions. Therefore, special focus in this study is on how weather conditions influence the ~~length of the time period in which~~ time when the O_3 ~~is mainly produced~~maximum occurs, the total O_3 gained as well as the total CH_4 depleted. Our findings are additionally analysed with respect to inter-seasonal variability. This is achieved by using the results of simulations performed in the European project REACT4C (Reducing Emissions from Aviation by Changing Trajectories for the benefit of Climate, https://www.react4c.eu/ (Matthes, 2011)). ~~REACT4C was intended to elaborate the feasibility of adopting flight altitudes and routes to minimise the climate impact of aviation and estimated the global effect of such an air traffic management measure in the North Atlantic flight sector (Grewe et al., 2014, 2017b). Within this analysis, interactions between weather conditions and trace gas concentration changes along the trajectory of each air parcel in which the emission occurs are investigated.~~ The modelling approach of REACT4C as well as the methodology used in this study is elaborated in Section 2. ~~Characteristics of changes in ozone and methane concentrations induced by emissions will be analysed in Section 3.1.~~ Afterwards all findings of this study will be presented (Section 3). In Section 4, uncertainties and findings ~~in~~ of this study will be discussed. Possible implementation strategies are presented in the conclusion (Section 5).

2 Methodology

Our analysis of the general concept of REACT4C as well as the modelling approach used will be elaborated first, to understand how the impacts of NO_x emissions on O_3 and CH_4 were simulated. The idea of the project is presented by Matthes (2011) and Matthes et al. (2012). A complete description of the modelling approach used is given by Grewe et al. (2014). Afterwards, a detailed description of the steps taken within ~~our analysis~~ the analysis of this work is presented.

2.1 REACT4C

REACT4C investigated the feasibility of ~~adopting~~ adapting flight routes and flight altitudes to minimise the climate impact of aviation and estimate the global effect of such air traffic management (ATM) measures (Grewe et al., 2014). In this particular study, this mitigation option was tested over the North Atlantic region. The general steps in this modelling approach were as follows: (1) select representative weather ~~pattern~~patterns, (2) define time-regions, (3) model atmospheric ~~contribution~~ contributions for additional emissions in these time-regions, (4) calculate the adjusted radiative forcing (RF), (5) calculate the climate change function (CCF) for each emission species and induced cloudiness, (6) optimize aircraft trajectories, based on the CCF results, by using an air traffic simulation system (System for traffic Assignment and Analysis at a Macroscopic level, SAAM) which is coupled to an emission tool (Advanced Emission Model, AEM), and (7) calculate the resulting operation

costs and the resulting climate impact reduction. For the present study, only step one to ~~five~~ three are important and will be further elaborated.

Irvine et al. (2013) identified, that by simulating frequently occurring weather situations within a season, the global seasonal impact can be estimated. They analysed meteorological reanalysis data for 21 years ~~of~~ for summer and winter. This reanalysis leads to three distinct summer (SP1-3) and five winter patterns (WP1-5). The different weather patterns mainly vary in their location, orientation and strength of the jet stream and the phase of the North Atlantic Oscillation and the Arctic Oscillation, two distinct teleconnection ~~pattern~~patterns. A graphical representation of each defined weather pattern is given by ~~Irvine et al. (2013, Figure 7 and Figure 8 for winter and summer, respectively)~~ Irvine et al. (2013, Fig. 7 and Fig. 8 for winter and summer and the actual weather ~~situation~~ situations simulated in REACT4C are presented in Frömming et al. (2020). Due to the lower variability of the jet stream in summer, only three distinct weather situations were determined. ~~Analogously, REACT4C simulated eight distinct model days, each representing one of these weather patterns.~~ The summer pattern occur 19 (SP1), 55 (SP2) and 18 (SP3) and the winter pattern 17 (WP1 & WP2), 15 (WP3 & WP4) and 26 (WP5) times per season in the reanalysis data (Irvine et al., 2013). Analogously, REACT4C simulated eight distinct model days, each representing one of these weather patterns.

To calculate the climate change functions, a time-region grid was defined in the North Atlantic region for seven latitudes (between 30°N to 80°N) and six longitudes (between 80°W to 0°W) over 4 different pressure levels (200, 250, 300 and 400 hPa) to account for different flight levels. At each time-region grid point, unit emissions of CO_2, NO_x and H_2O are initialised on 50 trajectories at 6, 12 and 18 UTC. However, ~~in the present study~~Grewe et al. (2014) found that the results show only minor sensitivity with respect to the temporal resolution. Therefore, only 12 UTC is considered in this study. The 50 trajectories are randomly located in the respective model grid box in which the specific time-region grid point is located. At each time-region grid point, 5×10^5 kg of NO (equals 2.33×10^5 kg(N)) are emitted, which is then equally distributed onto the trajectories (Grewe et al., 2014).

2.2 Base model description

The ECHAM/MESSy Atmospheric Chemistry (EMAC) model is a numerical chemistry and climate simulation system that includes sub-models describing tropospheric and middle atmosphere processes and their interaction with oceans, land and human influences (Jöckel et al., 2010). It uses the second version of the Modular Earth Submodel System (MESSy2) to link multi-institutional computer codes. The core atmospheric model is the 5th generation European Centre Hamburg general circulation model (ECHAM5, Roeckner et al. (2003)). For the present study we applied EMAC (ECHAM5 version 5.3.02, MESSy version 2.52.0) in the T42L41-resolution, i.e. with a spherical truncation of T42 (corresponding to a quadratic Gaussian grid of approximately 2.8 by 2.8 degrees in latitude and longitude) with 41 vertical hybrid pressure levels up to 5 hPa.

The applied model setup comprised multiple MESSy submodules important for the performed simulations. Each of the tracers (i.e. NO_x and H_2O) is emitted in an air parcel by the submodel TREXP (Tracer Release EXperiments from Point sources~~(TREXP)~~). The air parcel is then advected by the submodel ATTILA (Atmospheric Tracer Transport In a LAgrangian model~~(ATTILA) (Reithmeier and Sausen, 2002) .~~) (Reithmeier and Sausen, 2002) using the wind field from EMAC. In addition

to the 50 air parcels with tracer loading starting at each time-region, empty background air parcels are modelled in the northern hemisphere to allow for additional mixing, which in total yields about 169000 air parcels. The air parcels have a constant mass and the mixing ratio of each species is defined on the parcels centroid. The centroid is assumed to be representative for the whole air parcel and the Lagrangian cells are considered isolated air parcels. While ATTILA is per se non-diffusive, inter-parcel mixing is parameterized by bringing the mass mixing ratio in a parcel closer to the average background mixing ratio, which is the average mixing ratio of all parcels within a grid box. The vertical transport due to subgrid-scale convection 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. While a gain of tracer mass is distributed evenly among the air parcels in a grid cell, a reduction of tracer mass is calculated according to the mass available. Further details are given in Reithmeier and Sausen (2002).

For each trajectory the contribution of the emission (i.e. NO_x and H_2O) to the atmospheric concentration of CH_4, O_3, HNO_3, H_2O and OH is calculated over a time period of 90 days by using the submodel AIRTRAC (version 1.0, Frömming et al. (2013); see Supplement of Grewe et al. (2014)). The tagging approach used by AIRTRAC was first described by Grewe et al. (2010). In this approach each important chemical reaction is doubled. The first reaction applies to the whole atmosphere (from here onwards referred to as background) and the second one only to the additionally emitted tracer (from here onwards referred to as foreground). The submodel MECCA (Module Efficiently Calculating the Chemistry of the Atmosphere) is used to model the background chemical processes in the troposphere and stratosphere. The chemical mechanism used by MECCA can be grouped into sulfur, non-CH_4 hydrocarbon, basic O_3, CH_4, HO_x and NO_x and halogen chemistry (Sander et al., 2005). AIRTRAC on the other hand calculates the resulting changes due to the additional emitted NO_x in the foreground. AIRTRAC assumes that each concentration change of O_3 due to aviation is attributed to the emitted NO_x, which is consistent with Brasseur et al. (1998). Concentration changes due to additionally emitted NO_x are calculated based on the concentration of all chemical species involved in the general chemical system and the concentrations due to the extra emitted NO_x. The actual concentration change is then calculated based on the background reaction rate and the fraction of foreground and background concentrations of all reactants (Grewe et al., 2010). In detail, the foreground loss of O_3 ($L^f_{O_3}$) via Reaction R5 is based on the foreground and background concentrations of NO_2 and O_3 (NO_2^f, O_3^f and NO_2^b, O_3^b for foreground and background, respectively) and the background loss of O_3 ($L^b_{O_3}$), as given in Equation 1.

$$NO_2 + O_3 \rightarrow NO + 2O_2 \quad \text{(R5)}$$

$$L^f_{O_3} = L^b_{O_3} \times \frac{1}{2}\left(\frac{NO_2^f}{NO_2^b} + \frac{O_3^f}{O_3^b}\right) \quad (1)$$

~~In total~~In total, AIRTRAC calculates the mass development of NO_x, O_3, HNO_3, OH, HO_2 and H_2O by tracking 14 reactions and reaction groups. One group for the production and one for the destruction of O_3 as well as one for the formation of HNO_3~~is tracked~~. Three and five reactions are tracked for the OH production and destruction, respectively. In addition, three reaction

groups for the production and destruction of HO_2 are tracked. Further loss processes like wash-out and deposition are taken into account (Grewe et al., 2014). The results of this mechanism agrees well with earlier studies with respect to the regionally different chemical regimes and the overall effect of aviation emissions (Grewe et al., 2017c, Section 4.3, therein). The tagging mechanism also enables the quantification of methane losses due to the two major reaction pathways, the change in HO_x partitioning towards OH due to a NO_x emission and the production of OH due to an enhanced ozone concentration (Grewe et al., 2017c, Figure 8). Section 4 includes an elaborate discussion on the modelling approach used.

2.3 Analysis performed in this study

Within this study we used the simulation output created by the REACT4C project. As some output variables were not available for all emission locations and weather patterns, not all time regions and weather pattern could be included in the present study. Some of the raw data of WP2 were subject to data loss and not all analyses could be performed with this weather pattern. Therefore, WP2 has been excluded from this analysis. Emissions occurred in a time region grid of 7 latitudes, 6 longitudes and 4 pressure levels at 12 UTC. From originally 1344, 1115 emission locations are analysed. At each emission location all 50 air parcels are taken into account, resulting in 55750 trajectories being analysed. An output resolution of six hours was used by REACT4C over 90 days.

The variables taken into account in this analysis can be categorised into three different groups: (1) background and foreground chemical concentrations, (2) background and foreground chemical reaction rates and (3) general weather information. All foreground variables are present on the tracer grid (trajectory id and time), whereas background data are stored on the original EMAC grid. To simplify our analysis, background data were re-gridded onto the tracer grid. Here it was assumed that all background data within a grid box are valid for each air parcel within this specific EMAC grid box.

Due to the general complexity of the atmospheric chemistry, many variables can potentially influence changes in O_3 and CH_4 concentrations induced by NO_x emissions. Therefore, correlation matrices were used to identify interacting parameters. For these matrices the three most common statistical measures to identify correlations were used (Pearson, Kendall, and Spearman's rank correlation coefficient). Statistical significance is ensured by using t-, one-way analysis of variance (ANOVA) and Tukey's honest significant difference (HSD) tests.

3 ~~Characteristics of the temporal development of and due to~~

~~In the upper troposphere is mainly produced by a catalytic reaction including . Emitted reacts with forming . Via photodissociation, forms which leads to the production of (Reaction R1 - R3).~~

~~$HO_2 + NO \rightarrow OH + NO_2 NO_2 + h\nu (\lambda \leq 410nm) \rightarrow NO + O(^3P)\, O(^3P) + O_2 + M \rightarrow O_3 + M$~~

~~The additionally formed leads to an oxidation of~~ The long term reduction of ozone due to the induced CH_4 ~~:~~ loss (i.e. PMO) occurs far beyond the 90 days simulated by REACT4C. Simulating the effect of PMO explicitly is computationally too expensive for the modelling approach used. In REACT4C, PMO was thus not modelled explicitly. Instead a constant scaling

factor of 0.29, based on Dahlmann (2012), was applied to the resulting climate impact of methane (Grewe et al., 2014). The effect of PMO is therefore not considered within in this study and focus will be on the total CH_4 depletion, only.

~~$CH_4 + OH \rightarrow CH_3 + H_2O$~~

~~Changes in and concentrations introduced by an emitted unit emission of (emission strength: 5×10^5 kg of NO).~~

~~Figure 1 shows the "typical" temporal development of~~

3 Results

Within this section, the results of this study are described. First, a short analysis of the characteristics of the variability in the O_3 ~~and due to an aviation attributed emission. is reduced almost exponentially and washed out after about a month. While the emitted decreases, the~~ maximum is presented. The influence of transport processes on the time of the O_3 ~~concentration increases due to the described production processes (Reaction R1 -R3). When the mixing ratio is below a certain level, no further~~ maximum is analysed in Section 3.1. The mechanisms controlling total O_3 ~~is produced and loss terms dominate the chemistry. The additionally formed continuously decreases and reaches zero after about three months. At the same time the additional and form OH leading to a depletion of~~ gained are investigated in Sec. 3.2. The influence of tropospheric water vapour on total CH_4 ~~(Reaction R4). After all and is lost, the negative anomaly starts to decay and will later reach its original values (not seen in Figure 1). There is a large variability in this --relation, which is presented in Figure 9 of Grewe et al. (2014). In the following we analyse, which parameters lead to this large variability~~ depletion is discussed in Section 3.3. These findings are presented for summer and winter. An inter-seasonal variability analysis is performed in Section 3.4.

3.1 Characteristics of the temporal development of O_3

Figure 2 shows the maximum O_3 ~~concentration~~ mixing ratio in relation to the time after emission when the maximum occurs. ~~A high concentration change is~~ During winter and summer, high concentration changes are only possible if the O_3 maximum occurs early. In this scope, the O_3 maximum is defined as the maximum mixing ratio after which no further increase of O_3 occurs. Figure 3 shows the "typical" temporal development of O_3 for an early, a mid and a late O_3 maximum. The early maximum is characterised by a high production of O_3 in the first days after emission. The mid and late maximum are dominated by a slower O_3 production. In the case of the late maxima, the extreme slow O_3 production leads to a stretched version of temporal development of the air parcel with the early and mid maximum. As stated in Sec. 1, only the early maximum is characterised by a high O_3 maximum and the magnitude decreases by 1/3 and 2/3 for the mid and late maxima, respectively. The top panel of Fig. 2 gives the frequency of when the maximum occurs for both seasons. About 47.5 % and 72 % of all air parcels reach their O_3 maximum ~~within~~ during the first 21 days ~~during~~ in winter and summer, respectively. Only a small number of air parcels reach their maximum ~~at~~ towards the end of the simulation (winter: 2.5~~%~~ %, summer: < 1~~%~~ %). All air parcels with a ~~late maximum~~ maximum at the end of the simulation are emitted at higher altitudes (200 or 250 hPa) ~~. It is known that the concentration change of~~ and high latitudes. During winter, these air parcels stay at high latitudes and do not experience any solar radiation (i.e. solar night). The missing solar radiation dampens the O_3 ~~is not the only factor that influences~~

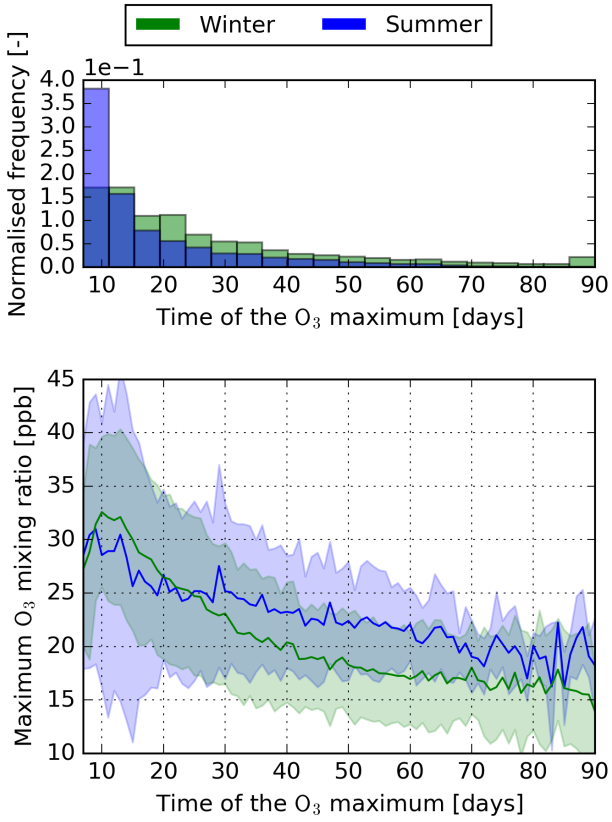

Figure 2. ~~Maximum~~ Top: Normalised histogram of when the O_3 ~~concentration~~ maximum is reached after emission. Results are normalised to the total number of air parcel in summer and winter, respectively. Bottom: Mean (solid line) and standard deviation (shaded area) of the maximum O_3 mixing ratio in relation to the time when the O_3 maximum is reached. ~~Data are taken from the first winter pattern. The Spearman rank correlation coefficient ρ is provided as a statistical measure.~~

~~the resulting radiative forcing.Lacis et al. (1990) demonstrated that also the altitude at which the concentration change occurs plays a significant role on the radiative impact. The climate impact increases from the surface towards the tropopause, then decreases again resulting in a cooling effect in the lower stratosphere above 30 km. Within the REACT4C experiment a higher~~ formation, leading to no distinct O_3 ~~concentration change generally leads to a larger RF (Spearman rank coefficient of 0.74). Even though the RF is only an approximation of the CCF, the time and magnitude of the~~ maximum ~~influence its potential climate impact. Both characteristics are thus analysed within this study. In contrast, the RF due to a depletion of is considered to be proportional to the total loss. This is due to the long tropospheric lifetime of . Therefore, focus will be on the total depletion, only.~~ within the 90 simulated days.

4 ~~Results~~

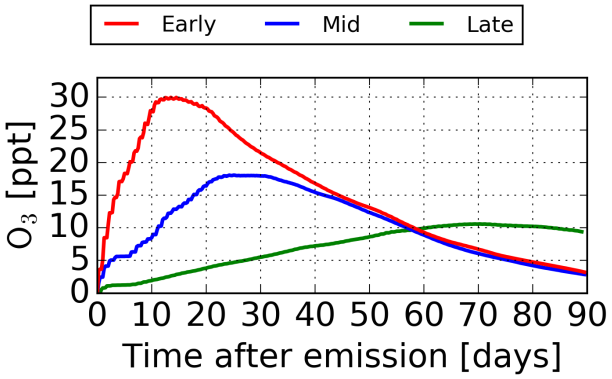

Figure 3. "Typical" temporal development of an early (red), a mid (blue) and a late (green) O_3 maximum.

~~Within this section, the results of this study are described. The influence of transport processes on the time of the maximum is analysed in Section 3.1. The mechanisms controlling total change are investigated in Section ?? and ?? for summer and winter, respectively. The influence of tropospheric water vapour on total depletion is discussed in Section 3.3. These findings are presented for representative weather patterns. An inter-seasonal variability analysis is performed in Section 3.4.~~

3.1 Importance of transport processes on the time of the O_3 maxima

~~Tropospheric chemistry is controlled by multiple factors like concentrations of reactants or~~ It is well established that the ozone production efficiency depends on the general chemical activity, controlled by weather conditions (i.e. temperature), and the concentration of each reactant. These weather conditions and reactant concentrations differ significantly across the troposphere~~. Our results show~~ , such that certain regions have a higher ozone production efficiency. Therefore, the transport into these regions controls the ozone gained. Our analysis shows that air parcels with an early maximum are characterised by a strong downward wind component, whereas late maxima have a weak downward or even an upward vertical wind component (not shown). Therefore, air parcels with an early O_3 maximum are transported to lower altitudes (~~Figure 4a~~top panel in Fig. 4) and lower latitudes (~~Figure 4b~~bottom of Fig. 4). Air parcels with a late maximum mostly stay at the emission altitude and latitude or are transported to higher altitudes and latitudes. For all winter patterns most maxima occur in a region spanning from 15°N to 35°N at pressure altitudes between 900 to 600 hPa. The maximum region is slightly shifted to higher altitudes for all summer patterns~~, which can be related to generally earlier maxima in summer, leaving less time for downward transport.~~ . No maximum occurs at high latitudes during winter due to the absence of solar radiation in the polar region during this time period. ~~In total, this indicates that~~ This indicates that a significant O_3 production, leading to an early O_3 maximum~~only occurs~~ , is only possible if an air parcel is transported to lower altitudes and latitudes.

Tropospheric vertical transport processes have many causes, e.g. temperature differences, incoming solar radiation, as well as latent and sensible heat. Vertical transport occurs in deep convection and conveyor belt events and causes an exchange of trace gases between the upper and the lower troposphere. Figure 5 ~~a~~ (top) shows the mean layer thickness anomaly of the 850

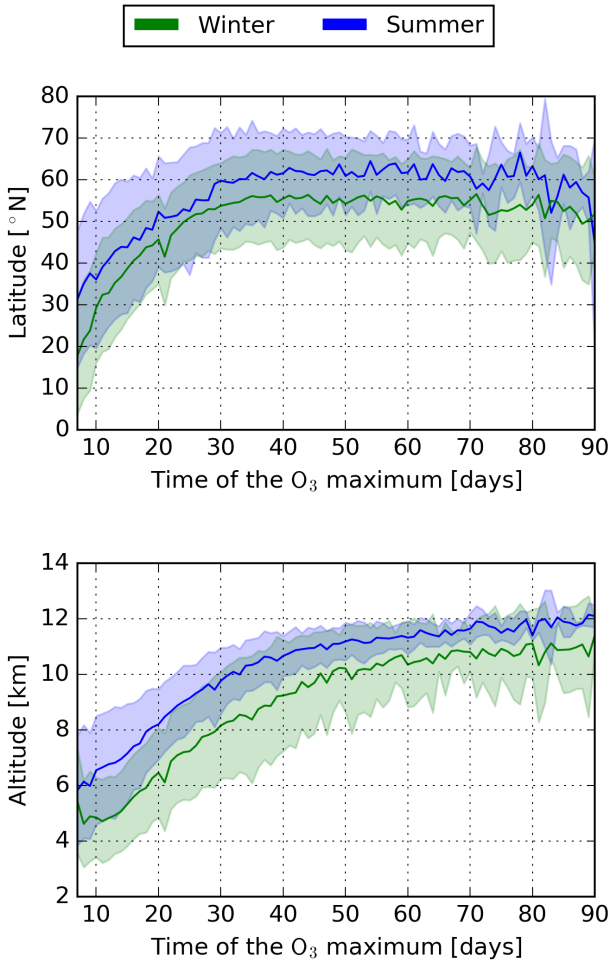

Figure 4. Mean (asolid line) ~~Mean altitude of air parcels at the time of the maximum relative to the emission altitude.~~ and standard deviation (~~b~~shaded area) ~~Mean latitude~~ of ~~air parcels at~~ the ~~time~~ location of each air parcel within the ~~maximum. The analysis is based on the~~ first seven ~~day~~ days after emission in relation to when the maximum O_3 mixing ratio is reached. ~~Data are taken from WP1~~Top: Latitude. ~~The Spearman rank correlation coefficient ρ is provided as a statistical measure~~Bottom: Altitude.

to 250 hPa layer. The layer thickness is proportional to the mean virtual temperature of the layer: a higher layer thickness indicates a higher temperature and moisture content. Here, the anomaly is presented for a better comparison of summer and winter, since higher mean virtual temperatures during summer lead to generally higher layer thickness values. The anomaly is obtained by deducting the seasonal mean. Air parcels with an early maximum have a higher mean layer thickness, whereas late maxima are associated with low layer thicknesses. In classical weather analysis, the layer thickness is used to identify synoptic weather systems. This indicates that in our study early maxima only occur if an air parcel originates or is transported into and stays within a high pressure system. No correlation exists between the time of the maximum and the layer thickness

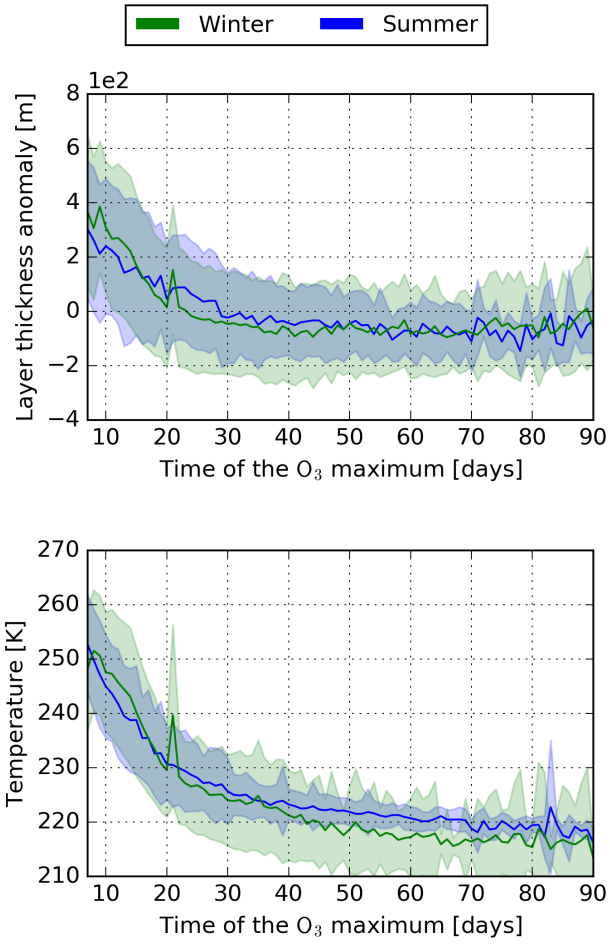

Figure 5. ~~(a)~~ Top: Mean thickness of the 850 - 250 hPa layer. ~~(b)~~ Bottom: Mean dry air temperature. ~~Both are given in~~ In both cases the relation to the time when the O_3 maximum is reached is given. The seasonal mean ~~value~~ is ~~based on~~ represented by the ~~time span from emission until~~ solid line and the ~~maximum is reached. Data are taken from WP1. The Spearman rank correlation coefficient ρ is provided~~ standard deviation as ~~a statistical measure~~shaded area.

at emission (Spearman rank coefficient of -0.2). Still, air parcels originating within in the core of a high pressure system have generally earlier maxima compared to air parcel which are transported into high pressure systems after emission (not shown). It is well known that subsidence is dominating vertical transport processes within high pressure systems, explaining the strong downward motion of air parcels, characterised by early maxima. Air parcels with late maxima stay within low pressure systems in which upward motion dominates.

~~Even though vertical transport is identified to be the main cause of earlier~~

3.2 Weather conditions controlling the O_3 production efficiency

Even though early O_3 maxima are characterised by strong vertical downward transport, transport processes ~~themself~~ do not directly influence chemical processes in the atmosphere. ~~Figure 5 b~~ Temperature is known to be a major factor controlling chemical processes in the atmosphere and is generally higher at lower altitudes and latitudes. The bottom panel of Fig. 5 shows the mean dry air temperature along the air parcel trajectory until the O_3 maximum is reached. The mean dry air temperature is higher for air parcels with early O_3 maxima, which is due to the downward and southward transport (leading to higher temperatures) within high pressure systems. ~~Temperature is known to be a major factor controlling chemical processes in the atmosphere.~~ These higher temperatures lead to higher background ~~reaction rates~~ chemical activity (higher background reaction rates) and therefore accelerate foreground chemistry. ~~The reaction rates for the production and loss of due to differ in their respective temperature dependency. Temperature has a low negative influence on the production rate coefficient but a strong positive impact on the destruction of . Temperature generally decreases with increasing altitude (in the troposphere) and decreases towards higher latitudes. If an air parcel is transported to lower altitudes with higher temperatures, the chemical loss dominates the production of leading to a subsequent decrease of total and thus an earlier maximum. Higher temperatures~~ Higher temperatures and enhanced photochemical activity at higher altitudes during NH summer~~also~~ , explain the tendency of earlier maxima in this season.

3.3 ~~Influence of background on total change in summer~~

~~A high mean air temperature causes an early maximum but only weakly correlates with the maximum concentration (Spearman rank coefficient about 0.5), suggesting that additional factors control the maximum gain.~~ From classical chemistry ~~, the production rate further depends on the~~ the efficient production of O_3 does not only depend on the higher chemical activity, due to higher temperatures, but also on the concentrations of the reactants involved. In ~~case of~~ the case of the formation of O_3 due to NO_x, these are NO and HO_2 (Reaction R1). Figure 6 ~~shows the dependency of~~ and 7 show how the mixing ratios of NO_x and HO_x ($=OH + HO_2$) relate to the maximum O_3 ~~concentration in relation to the mean~~ mixing ratio. Here, NO_x ~~concentration. Only~~ and HO_x are used to account for the rapid cycling of the species within each radical group. For both seasons, only low NO_x concentration will lead to high ~~changes in~~ O_3 ~~concentrations~~contributions. The production of O_3 via Reaction R1 to R3 dominates at low background NO_x concentrations, whereas at high background concentrations NO_2 is eliminated by reacting with OH and HO_2 forming nitric acid (HNO_3) and peroxynitric acid (HNO_4), respectively. From Fig. 7 it becomes evident that a high increase in O_3 is only possible at high HO_x concentrations, since at low HO_2 no O_3 will be formed via Reaction R1.

In the upper troposphere the background NO_x concentration is altitude dependent and generally increases towards the tropopause (not shown). On the other hand, HO_x is high at low altitudes and latitudes and decreases towards the tropopause. Air parcels with a fast downward transport, generally experience lower mean background NO_x concentrations and higher HO_x background concentrations, resulting in a higher O_3 gain. Air parcels which stay close to the tropopause or are even transported into the stratosphere experience a high background NO_x concentrations, leading to ~~less~~ a lower O_3 ~~being formed.~~

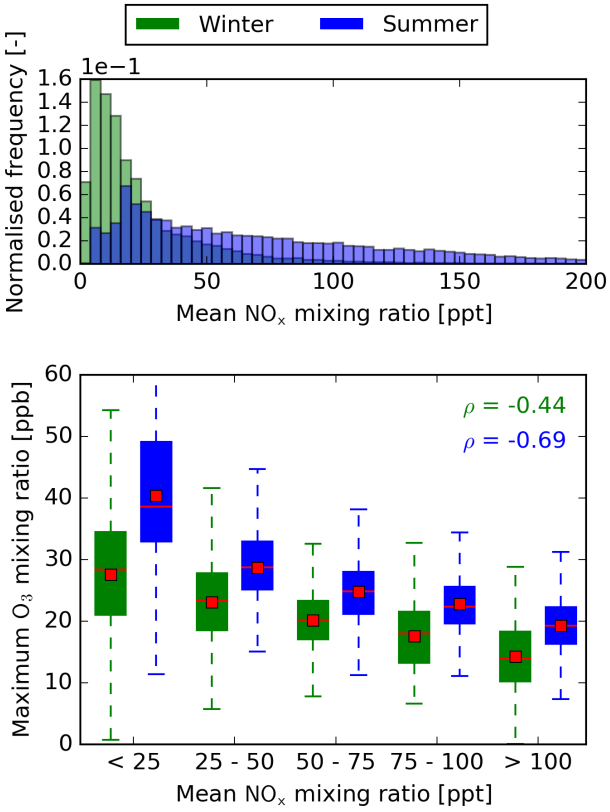

Figure 6. ~~Maximum concentration in relation to~~ Top: Normalised histogram of the mean ~~background~~ NO_x ~~concentration from emission~~ mixing ratio until the ~~maximum~~ O_3 ~~concentration~~ maximum is reached. ~~Data~~ Results are ~~taken from SP3~~normalised to the total number of air parcel in summer and winter, respectively. Bottom: Binned box plots showing the relation between the mean NO_x mixing ratio and the magnitude of the O_3 maximum. The median and mean for each box plot are represented by red lines and red boxes, respectively. Additionally, the Spearman rank ~~correlation~~ coefficient ρ is ~~provided as a statistical measure~~given for summer and winter.

3.3 ~~Influence of background on total change in winter~~

~~Maximum concentration in relation to the mean background concentration from emission until the maximum concentration is reached. Data are taken from WP1. The Spearman rank correlation coefficient ρ is provided as a statistical measure. The background concentration strongly correlates with emissions in summer but weakly with emissions in winter. The Spearman rank coefficient for most winter pattern is below 0.44. During winter the maximum concentration correlates strongly with the mean background concentration of (see Figure 7), the second reactant in Reaction R1. It becomes evident that only high background concentrations of lead to a high formation of . High background concentrations~~ formation, since most NO_x is eliminated by forming HNO_3 and HNO_4. In addition, high background HO_x concentrations dominate in high pressure systems (not shown) ~~. As for , has a high variability throughout the troposphere and is generally lower in the upper~~

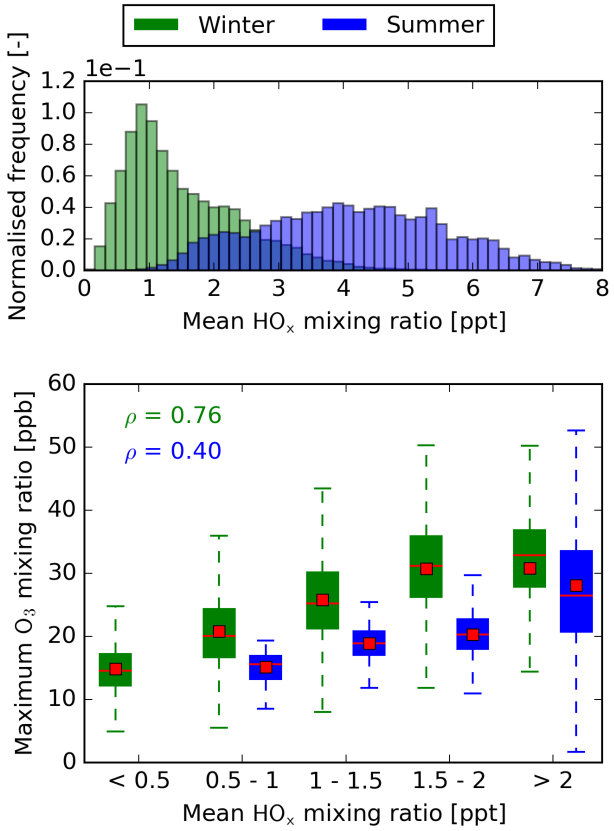

Figure 7. Top: Normalised histogram of the mean HO_x mixing ratio until the O_3 maximum is reached. Results are normalised to the total number of air parcel in summer and winter, respectively. Bottom: Binned box plots showing the relation between the mean HO_x mixing ratio and the magnitude of the O_3 maximum. Please note that during summer HO_x mixing ratios below 0.5 ppt show no statistical significance. Therefore, no box plot is provided in this case. The median and mean for each box plot are represented by red lines and red boxes, respectively. Additionally, the Spearman rank coefficient is given for summer and winter.

~~troposphere and increases towards lower altitudes and latitudes. This explains~~ explaining why air parcels in high pressure systems ~~experience higher background concentrations, since they are quickly transported to lower altitudes and thus into areas where high concentrations dominate~~have a generally higher O_3 formation.

~~The correlation of and maximum~~ Summer and winter significantly differ with regard to the correlation of NO_x and HO_x with the O_3 ~~during summer is rather weak (~~maximum. Summer has a high Spearman rank coefficient ~~of about~~ for NO_x (ρ = -0.69) but only correlates weakly with HO_x (ρ = 0.40). ~~Thus, it can be concluded that during summer the~~ Winter on the other hand correlates well with HO_x (ρ = 0.76) but weakly with NO_x (ρ = -0.44). This difference is explained by varying NO_x and HO_x concentrations in both seasons. The top panel of Fig. 6 and 7 gives the normalised frequency of NO_x and HO_x for both seasons. It becomes evident that winter is characterised by low NO_x and HO_x concentrations, where as summer is dominated

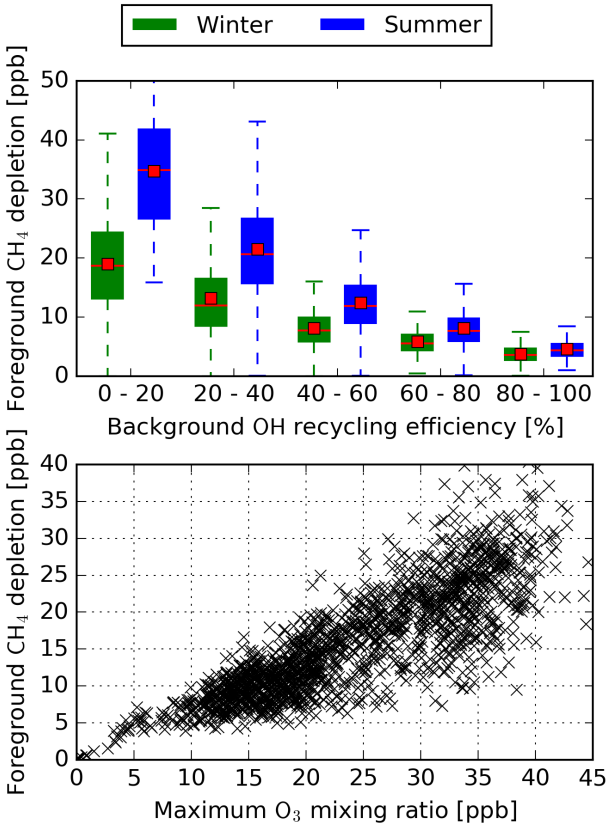

Figure 8. ~~Relative contribution~~ Top: Binned box plots of ~~all considered sources to~~ the ~~total background~~ OH ~~production~~recycling probability. ~~Air parcel with a total depletion of more than 5 ppb~~ The median and mean for each box plot are ~~considered to have a high total depletion~~represented by red lines and red boxes, respectively. ~~Results are valid~~ Bottom: Maximum O_3 mixing ratio vs. maximum CH_4 mixing ratio for ~~the first winter pattern~~recycling probabilities below 20 % for WP1.

by high NO_x and HO_x concentrations. For many air parcels during summer, enough HO_x is present to allow a high formation of O_3~~formation is limited by the background~~ , but a higher NO_x ~~concentration, whereas in winter low~~ concentration limits the formation of O_3 and leads to the formation of HNO_3 and HNO_4. During winter, the low NO_x concentrations theoretically allow for a high O_3 formation but low HO_x concentrations limit the ~~total~~ efficient production of O_3~~gained~~.

3.3 Influence of water vapour on the total CH_4 depletion

In comparison to O_3, only the total CH_4 depletion is of interest, due to its longer atmospheric lifetime. A high O_3 concentration ~~may lead~~ leads to a high CH_4 depletion (Spearman rank coefficient of 0.66) since O_3 is a major source of OH, accelerating the depletion of CH_4 (Reaction R4). However, the moderate Spearman rank coefficient indicates that other factors additionally control the CH_4 depletion process. Our results show that a high foreground CH_4 depletion is only possible if the background

OH concentration is high. ~~Air parcels with high~~ When looking at OH the fast cycling between OH and HO_2 has to be taken into account. Analysing the recycling probability (r) of OH is useful to account for this cycling. Here, we define the recycling probability, following Lelieveld et al. (2002), as:

$$r = 1 - \frac{P}{G} \tag{2}$$

in which P is the primary production of OH via:

$$H_2O + O(^1D) \rightarrow 2OH \tag{R6}$$

and G is the gross OH production additionally considering the secondary production of OH. In general, when r approaches 100 % the formation of OH becomes autocatalytic. When r approaches 0 %, all OH is formed via Reaction R6. Based on a perturbation study, Lelieveld et al. (2002) identified that for recycling probabilities above 60 % the chemical system becomes buffered and that NO_x perturbations in this regime have only little impact on OH. The top of Fig. 8 shows that a high CH_4 ~~depletion experience a three times higher total~~ depletion is only possible if the recycling probability of background OH ~~production in the background compared to air parcels with low~~ is below 60 %. When the major source of OH is from Reaction R6 (r approaches 0 %), the formed OH is not recycled via NO_x, accelerating the depletion of CH_4~~depletion. In the modelling approach used, three sources of hydroxyl radicals were tagged: (1) the reaction of water vapour with exited oxygen atoms (), (2) the reaction of hydoperoxyl radicals () with ozone and (3) with nitrogen oxide. Figure 8 shows the relative contribution of each major source to the~~ . The major source of foreground OH is the formed O_3. A high background OH ~~production for low and high~~ recycling probability does not necessarily mean that foreground O_3 is efficiently produced. The foreground formation of O_3 is limited by background NO_x and HO_x during summer and winter, respectively. Figure 8 (bottom) shows the relation between the total CH_4 depletion ~~. Here, air parcel with a total~~ and the O_3 magnitude for recycling probabilities below 20 % (most left box-plot in top of Fig. 8). Thus, the possible low O_3 formed limits the CH_4 depletion ~~of more than 5 ppb are considered to have a high~~ at low OH recycling probabilities, explaining the high spread in the total CH_4 depletion ~~. It becomes evident that the contribution to background differs significantly and almost twice as much is produced due to the reaction involving water vapour for the high~~ in that regime. It can thus be concluded that a high depletion of CH_4 ~~depletion case.~~ is only possible if the major formation of OH is due to Reaction R6.

Globally, Lelieveld et al. (2016) estimate that about 30% of the tropospheric OH is produced by ~~this reaction, taking additional sources into account. This suggests that air parcels experiencing high depletion are on average exposed to a higher water vapour content than the average troposphere. The~~ Reaction R6. This reaction is limited by the availability of $O(^1D)$, formed from the photolysis of O_3, and H_2O. In this study, the highest CH_4 depletion rate occurs in tropical regions close to the surface (between 0°and 20°N and below 850hPa), which is dominated by hot and humid weather conditions. This region is known to have high HO_x ~~(+)~~ concentrations due to active photochemistry and large OH sources and sinks. Here, the contribution of OH being produced by water vapour is highest (Lelieveld et al., 2016). ~~The importance of water vapour is further strengthened by a strong correlation~~ A strong correlation exists between the average specific humidity along the air parcel trajectory and the total depletion of CH_4 (mean Spearman rank coefficient ~~above 0.71).~~

Table 2. Spearman rank coefficient ~~for~~ of all ~~relations~~ identify ~~within in this study given~~ relations for each individual weather pattern~~taken into account~~. All correlation factors related to the time and the maximum O_3 mixing ratio are calculated based on mean values for the time span between emission and the time of the O_3 maximum. The correlation between the total CH_4 depletion and specific humidity is based on the mean between time of emission and the time when the total CH_4 depletion is reached.

Correlation factors		Winter				Summer		
		WP1	WP3	WP4	WP5	SP1	SP2	SP3
Time of O_3 maximum	Vertical wind velocity	-0.67	-0.77	-0.69	-0.61	-0.66	-0.66	-0.57
	850 -250 hPa layer thickness	-0.74	-0.66	-0.61	-0.63	-0.59	-0.63	-0.56
	Mean temperature	-0.78	-0.84	-0.80	-0.79	-0.86	-0.82	-0.81
Maximum O_3 mixing ratio	Mean background NO_x	-0.08	-0.37	-0.43	-0.66	-0.73	-0.76	-0.78
	Mean background HO_2	0.76	0.71	0.71	0.38	0.38	0.21	0.40
Total CH_4 depletion	Specific humidity	0.81	0.82	0.82	0.75	0.74	0.71	0.72

~~While is gained most foreground~~ of 0.77). In particular, air parcels with a low OH ~~is produced by reacting with (see Figure 1). In this period 35% and 42% of background OH is produced by reacting with and , respectively. Due to the higher air parcel altitude during the gain, is only a minor source of .~~

~~The importance of as an source increases with increasing foreground concentration. In Figure 1, this can be clearly seen by an increasing~~ recycling probability and thus a high CH_4 ~~depletion rate between day five and ten. The larger background depletion due to higher concentrations at lower altitudes explains why air parcels with an early maximum due to a strong downward transport and a high total gain experience a larger total~~ depletion are characterised by high specific humidity and high incoming solar radiation. Therefore, a ~~larger cooling effect due to a larger~~ high depletion of CH_4 is only possible ~~for air parcels with a high formation by and if those air parcels are then transported into regions which are dominated by a high water vapour content~~if the air parcel is transported towards low altitudes and latitudes. The transport into tropical regions, ~~which are characterised by those conditions,~~ occurs mainly due to the subsidence in high pressure systems (see ~~Section~~ Sec. 3.1).

3.4 Inter seasonal variability

Within this study specific weather situations (for graphical representations see ~~Irvine et al. (2013, Figure 7 and Figure 8)~~Irvine et al. (2013, , and Frömming et al. (2020)) were analysed. Table 2 shows an overview of all correlations analysed within this study for each distinct weather pattern. Vertical transport processes until the O_3 maximum, represented by the vertical wind velocity, correlate reasonably well within both seasons. However, during summer the correlation tends to be lower. Summer pattern 3 has the lowest correlation coefficient and the highest mean downward wind velocity. The pattern is characterised by a high pressure blocking situation, resulting in an overall high layer thickness~~. This weakens the distinction between high and low pressure systems which explains the~~ , resulting in a weaker correlation. ~~Thus it is~~ It is thus expected that differences in each indi-

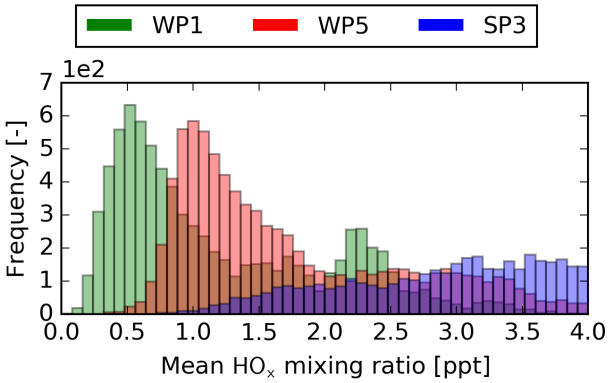

Figure 9. Histogram for the mean HO_x mixing ratio until the O_3 maximum is reached for WP1, WP5 and SP3. Note that mixing ratios above 4 ppt are not shown.

vidual weather situation (i.e. number, location and strength of the high pressure systems) cause the inter-seasonal variability. Additionally, downward transport during summer is less important for air parcels to experience high temperatures. This also explains the weaker correlation for the 850 to 250 hPa layer thickness during summer. Still, air parcels which stay in a high pressure region experience earlier maxima during summer.

The mean background concentration of NO_x correlates strongly with the O_3 magnitude for each summer pattern, giving no indication that there is another parameter that controls the total O_3 gain. However, no correlation exists for most winter patterns. This indicated that in winter, when the chemistry is slow at the emission location of mid and higher latitudes, the transport pathway, e.g. towards the tropics, is more important that the chemical background conditions at the time of emission, whereas in summer with active photochemistry the background NO_x concentration plays a role. Only winter pattern five shows a ~~strong correlation. When visually inspecting (not shown) the given correlation, it becomes evident that it looks almost the same as for~~ stronger correlation. At the same time, WP5 has a low correlation with HO_x. Figure 9 gives the frequency of the mean background HO_x concentration for WP1, WP5 and SP3~~(Figure 6) with the main difference that the maximum background concentration is lower but still significantly higher than for the other winter pattern. Winter pattern five also correlates weakly with the mean background concentration~~. In comparison to WP1, WP5 is characterised by higher HO_x concentrations with most mixing ratios above 1 ppt (WP1 mean: 1.2 ppt, WP5 mean: 1.8 ppt; WP1 median: 0.89 ppt, WP5 median: 1.4 ppt). In general, mixing ratios above 1 ppt allow a high O_3 formation (see Fig. 7). This indicates that for most air parcels in WP5, HO_x is not the only limiting factor. At the same time WP5 is characterised by higher NO_x mixing ratios (WP1 mean: 18.1 ppt, WP5 mean: 31.6 ppt), explaining the higher correlation with NO_x. This indicates that in the case of WP5 the maximum O_3 concentration is limited by a combination of HO_x and NO_x. The chosen example ~~of winter pattern five~~ day in EMAC for WP5 occurs at the end February, whereas the other winter patterns are initialised in December or early January. This indicates that ~~the controlling influence of changes~~ HO_x becomes a less limiting factor towards spring. This suggests that our results are only valid for ~~the~~ both analysed seasons and further research is necessary to identify the controlling factors in spring and autumn.

Specific humidity is clearly the controlling factor of the total CH_4 depletion for all weather patterns taken into account. Again the correlation is weaker for summer which ~~can be explained by~~ is due to the generally higher H_2O concentrations. This results in a lower variability in the specific humidity which weakens the correlation analysed. Here, ~~winter pattern five~~ WP5 again behaves like all summer patterns. This indicates that O_3 and CH_4 concentration changes due to emissions in spring are most likely controlled by mechanisms identified for summer.

4 Uncertainties and discussion

Our results indicate a large impact of transport patterns on ozone and methane concentration changes ~~resulting from~~ due to aviation NO_x emissions. This is both a highly complex interaction of transport and chemistry~~and~~ , and a relatively small contribution of ozone and methane concentration changes against a large natural variability. Hence, a direct validation of our results is not feasible. However, the main processes, such as transport and chemistry can be evaluated individually - at least in parts. ~~Hence~~ In the following we will discuss ~~in the following~~ some aspects of this interaction and discuss the ability of EMAC to reproduce observations.

An important aspect in our study is the model's transport. Short-lived species which only have a surface source, such as ^{222}Radon (^{222}Rn, radioactive decay half-lifetime of 3.8 d), are frequently used to validate fast vertical transport characteristics. Jöckel et al. (2010) and in more detail Brinkop and Jöckel (2019) showed that the model is able to capture the ^{222}Rn surface concentrations and vertical profiles, indicating that the vertical transport is well represented in EMAC.

The horizontal transport is difficult to evaluate and observed trace gases, which resemble the exchange between mid and high latitudes and the tropics, are not available. However, Orbe et al. (2018) compared transport timescales in various global models, e.g. from northern mid-latitudes to tropics, which differed by 30%~~or interhemispheric transport , which~~ . The interhemispheric transport differed by 20%. The authors concluded that vertical transport is a major source of this variability. More research is needed to better constrain models with respect to their tropospheric transport timescales. A more integrated view on the variability of aviation related transport-chemistry interaction is given by a model intercomparison of NO_x concentration differences between a simulation with and without aircraft NO_x emissions (Søvde et al., 2014, see their supplementary material). Here, we concentrate on the winter results to reduce the chemical impact to a minimum. The results clearly show a very similar NO_x change of the 5 models (including EMAC) peaking around 40°N at cruise altitude in winter with a tendency to have a downward and southward transport to the tropics. However, the peak values vary between 55 pptv and 70 pptv.

Chemistry, or more specific, the concentrations of chemically active species is evaluated in detail in Jöckel et al. (2010) and Jöckel et al. (2016). In general, EMAC overestimates the concentrations of tropospheric ozone by 5-10 DU in mid-latitudes and 10-15 DU in the tropics. Carbon monoxide on the other hand is underestimated, though the variability matches well with observations. The tropospheric oxidation capacity is at the lower end of model's estimates, but within the models' uncertainty ranges. Jöckel et al. (2016) speculate that lightning NO_x emissions or stratosphere-to-troposphere exchange might play a role. It is important to note that variations, which are caused by meteorology are in most cases well represented ~~(e.g. Grewe et al., 2017a, their sec. 3.2)~~(e.g. Grewe et al., 2017a, their Sec. 3.2).

Ehhalt and Rohrer (1995) already stated that the net-O_3 gain strongly depends in a non-linear manner on the NO_x mixing ratio. This is generally well reproduced in EMAC (Mertens et al., 2018, Figure 5) and even by an EMAC predecessor model (Dahlmann et al., 2011; Grewe et al., 2012, their Figure 4 and Figure 1, respectively). Stevenson et al. (2004) showed the response of ozone and methane to a pulse NO_x emission, which is very similar to our results (Grewe et al., 2014, their Figure 9). Stevenson and Derwent (2009) demonstrated that the NO_x concentration at time of emissions strongly defines the resulting climate impact of O_3. A similar but weaker relation can be found in the current data set (not shown) (Grewe et al., 2014, their Figure 9). For some air parcels the background NO_x concentration is low at time of emission but they are quickly transported to ~~areas with~~ regions characterised by higher concentrations. ~~Thus, they~~ These air parcel experience a temporal high O_3 ~~gain~~ production shortly after emission, due to low NO_x concentrations, but only little total O_3 is formed, ~~explaining~~ due to a high mean background NO_x mixing ratios after emission. This explains the difference between our correlation and the one of Stevenson and Derwent (2009). ~~Therefore,~~ Another aspect is the question how well the REACT4C concept, to model atmospheric effects of a local emission (Grewe et al., 2014), represents global modelling approaches, such as Søvde et al. (2014) or Grewe et al. (2017c). The approach was developed to gain more insights in aviation effects that can conventionally not be obtained. Hence, there are by definition limitations in answering this question. However, four indications can be given, which support the consistency of the modelling approach. First, the transport scheme is reasonably well established (Brinkop and Jöckel, 2019, and above). Second, the chemical response to a local emission agrees well with earlier findings of Stevenson et al. (2004), who simulated the monthly mean response of ozone and methane to a NO_x pulse and which are very similar to the results from this approach (Grewe et al., 2014, their Fig. 9). Third, the use of a trajectory analysis to interpret either observational data or modelling data is well established (Riede et al., 2009; Cooper et al., 2010) . And fourth, a first verification of the resulting global pattern of the atmospheric sensitivity to a local NO_x emission by comparing to sparsly available literature data was promising (Yin et al., 2018). This verification is based on a generalisation of the CCFs by developing algorithms to relate the weather information available at the time of emission to the resulting CCF. These algorithmic CCFs (aCCF, van Manen and Grewe (2019)) allow to predict all weather situations, compared to the limited applicability to a few selected days for the CCFs. An annual climatology of the results from using aCCFs were calculated and compared to conventional approaches (Yin et al., 2018). They conclude: It "shows the variation pattern of the ozone aCCFs matches well with the literature results over the northern hemisphere (the latitude between 30° N and 90°N) and the flight corridor (roughly 9 km to 12 km vertical range)".

From our findings, it can be concluded that not only the atmospheric conditions at the time of emission influences the O_3 gain but rather the region in which the maximum O_3 concentration and the maximum concentration change occur. The findings of Stevenson and Derwent (2009) are also only valid for summer and no winter analysis is provided, making it impossible to directly compare our findings identified in ~~Section ??~~ Sec. 3.2 to available literature. However, indirectly, by the use of generalised aCCFs, first findings indicate a reasonably well agreement of the simulated atmospheric response to local NO_x emission.

To conclude, both transport and chemistry processes are crucial for our results. EMAC is in many aspects in line with other model results, but has some biases in the concentration of chemical species. However, the variability of chemical species, such

as NO_x and O_3 is better represented than mean values, indicating that the interaction between transport and chemistry is reasonably well simulated. ~~Since~~ This result should be robust, since our results show a very strong relation between meteorology and the contribution of aviation emission, and since this interaction is in principle well represented in EMAC~~, this result should be robust~~. However, the strength of ~~, e.g.~~ the ozone response to a NO_x emission in a high pressure system has an uncertainty, which we hardly can estimate. Based on on the results of the model intercomparison by Søvde et al. (2014), we would expect an uncertainty in the order of 25%.

5 Conclusions

The possibility to reduce aviation's climate impact by avoiding climate sensitive regions, heavily depends on our understanding of the driving influences on induced contributions to the chemical composition of the atmosphere. In this study, we demonstrated the importance of transport processes on locally induced aviation attributed NO_x emission on ozone and methane concentrations over the North Atlantic flight sector.

~~The resulting climate impact due to changes in ozone concentrations depends on the maximum ozone gained, where only early maxima result in a large ozone concentration change.~~ The induced O_3 change is characterised by the time and magnitude of its maximum and high O_3 maxima are only possible if the maximum occurs early. Transport processes like subsidence in high pressure systems lead to early maxima, due to the fast transport into ~~warmer regions~~ regions with a higher chemical activity. In summer the NO_x-HO_x-relation is limited by background NO_x, whereas in winter the limiting factor are ~~the~~ low HO_x concentrations. ~~High~~ When an air parcel is transported into regions with high NO_x concentrations in summer~~lead to lower changes~~ , a low change in total ozone occurs, since less ozone is formed in the background. In this case most NO_2 is eliminated by forming nitric acid and peroxynitric acid. During winter ~~background limits the total ozone gain,~~ low background NO_x concentrations allow for a high O_3 formation but due to generally lower ~~concentrations in the area of interest~~HO_x concentrations no efficient O_3 formation occurs. Air parcels transported quickly towards lower altitudes encounter ~~higher~~ low NO_x but high HO_x concentrations leading to a higher ozone formation, strengthening the importance of transport processes on the O_3 formation.

The total depletion of methane depends heavily on the background OH concentration. ~~Since the major tropospheric source of~~ If most OH is formed by its primary formation process, which depends on water vapour, and only little OH is recycled to HO_2, a high depletion of CH_4 occurs. Therefore, the water vapour content which the air parcel experiences along its trajectory defines the total methane depletion. Air parcels transported into lower altitudes and latitudes experience higher water vapour concentrations. Thus, atmospheric transport processes also define the total methane depletion. Additionally, only a high total gain of ozone allows a large methane depletion.

Due to the complexity of the problem, we are not able to validate our results. It would be challenging to design a measurement campaign to proof the contribution of aviation NO_x emissions to ozone and methane. The standard deviation of background concentrations are generally considered to be higher than changes induced by aviation NO_x perturbation, making them hardly detectable (Wauben et al., 1997). Our analysis of the model performance however shows that both transport processes as well as

chemical concentrations are reasonably well represented. Our inter seasonal analysis shows that our findings to the importance of background NO_x and HO_x concentrations are only valid for both seasons analysed. ~~During summer background~~ Due to the high variability of NO_x ~~concentrations limit the total ozone gained. We~~ and HO_x concentrations in the troposphere, we expect other important factors to control the total ozone gain in other regions, not analysed in this study~~, due to the high variability of concentrations in the troposphere~~. Based on the findings of Köhler et al. (2013), we expect our results to be valid for most part of the northern extra-tropics. To conclude, further model studies are necessary to fully quantify how transport processes influence induced changes of ozone and methane concentrations in all seasons as well as other regions of interest.

~~Calculating the climate cost for the re-routing of flight trajectories~~ Mitigating the climate impact from aviation by estimating the climate impact and re-route flight trajectories using the same simulation setup (in resolution, time horizon and chemical mechanism used) as in REACT4C on a day to day basis is currently computationally too expensive ~~to be~~ and not feasible at the moment~~, if the same time horizon of 90 days was used as in REACT4C for simulating aircraft effects.~~ . Our results show that transport processes are of most interest when identifying the impact of local NO_x emissions on ozone and methane. Since, purely dynamic simulations without chemistry are ~~less computationally~~ computationally less expensive, the insights gained in this work can be used to allow a more feasible approach by ~~only looking at transport processes . Our results~~ estimating the climate impact based on transport processes and other weather factors (i.e. temperature and humidity). Our results further show that concentration changes of ozone on methane induced by aviation NO_x mainly occur within the first twenty days. Therefore, an alternative approach could be to use shorter simulations to estimate the induced concentration changes and thus reduce computation costs significantly.

Data availability. The data of the REACT4C project used in this work are archived at the German Climate Computing Centre (Deutsches Klimarechenzentrum, DKRZ) and are available on request.

Author contributions. SR and VG designed the analysis and SR carried them out. CF performed the simulations of REACT4C. SR prepared the manuscript with contributions from all co-authors.

Competing interests. The authors declare that they have no conflict of interest.

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