Author's replies to the reviewers

Impact of aircraft NOx and aerosol emissions on atmospheric composition and associated direct radiative forcing of climate

Terrenoire et al.

We thank both reviewers for their comments on our manuscript and for the time spent on their reviews. Please find below a detailed point-by-point reply to the comments and suggestions to reviewer #1 and reviewer #2.

Review 2 of the "Impact of present and future aircraft NOx and aerosol emissions on atmospheric composition and associated direct radiative forcing on climate" by Terrenoire et al.

This study explores and gathers various aspects of the impact of aircraft NOx emissions on atmospheric composition and climate utilizing a series of present and future aviation projections, and an updated global chemistry-aerosol-climate model, LMDZ-INCA. In addition to the well-established effects of aircraft NOx emission, this paper also investigates the impacts that arise from the formation of nitrate and sulfate aerosols that constitute a novelty of the publication as it has been addressed in only a few modeling studies so far. It is also associated with large uncertainties, which might be worth highlighting more in this publication.

The paper is well-written and scientifically sound. The applied methods are valid and clear. The findings of the study are of interest to the community. I recommend this paper for publication, preceded by a few minor comments.

We thank the reviewer for the positive comments. We have revised the abstract and the conclusion and in particular we have tried to emphasize the uncertainties associated with the aerosol forcings in these sections.

General comments:

Abstract reads more like a summary. It is too long, with too many technicalities, and it is difficult to follow. I would suggest authors consider re-writing it, concentrating on the main findings and their implications.

The abstract has been revised and is now shorter by about 50%.

The text is full of details, which on one side is understandable taking into account the number of experiments that were performed for this study. On the other hand, maybe authors can help to level this complexity of the paper by preparing a kind of 'takeaway figure'. Figure that would summarize the main results, highlight the novelty of this study, etc. I believe the paper might gain the readability by having such a figure, but I leave this decision with the authors.

The conclusion section has been revised. It is now shorter by 1/3 and more focused on the take-away messages that we now enumerate in order not to repeat the previous sections and to provide the take away messages. Please note also that a new Table 3 has been added in our manuscript, providing the labelling and key information on the various model simulations performed in the study.

Specific comments:

Lines 90-94: some references can be useful here

Two references have been added in the manuscript on these issues: Freeman, S., D. S. Lee, L. L. Lim, A. Skowron, and R. Rodriguez De Leon, Trading off aircraft fuel burn and NOx emissions for optimal climate policy. *Environ. Sci. Technol.* 52, 2498−2505 (2018) and Prashanth, P., R. L. Speth, S. D. Eastham, J. S. Sabnis, and S.R.H. Barrett, Post-Combustion Emissions Control in Aero-Gas Turbine Engines. *Energy and Environmental Science*, DOI: 10.1039/D0EE02362K, 2021.

Line 219: motigtaion ---> mitigation

The typo has been corrected.

Line 239: the list (e.g., as a Table) of performed experiments, maybe with their short description, might be helpful

A new Table 3 has been added in our manuscript, providing the labelling and key information on the various model simulations performed in the study.

Lines 464: why your "present-day" is 2004 here, while in the previous section is 2006?

It is indeed 2006. The typo has been corrected.

Line 490: the description of methane RF calculation and all the methane-induced components are missing? Maybe part of your section 6.2 can be moved to 3.4, or vice versa, for consistency.

The method used to calculate the methane forcing has now been moved to Section 3.4.

Line 528: Figure 5 (and subsequent Figures) why May? Why not July, or JJA? On the other hand, your RF numbers are based on annual averages, so maybe your chemistry analysis can show the annual means too?

As shown in Figure 3 the maximum perturbation in ozone associated with aircraft emissions is calculated in May in this model. This figure was specifically introduced to illustrate the full seasonal cycle, before focusing on the minimum (January) and maximum perturbations (May). May was specifically chosen to illustrate this maximum. In contrast to some models showing a maximum in July or in summer, another category of models (including LMDZ-INCA) shows a maximum in late spring. This maximum reached in May at high latitudes is the result of a combination of a more intense photochemical activity in summer combined with a more intense transport poleward in spring. As a consequence, the peak is reached at the end of the spring season. We have added a sentence when describing Fig. 3 in order to better explain this and the choice of May is better justified when introducing Fig. 5. For the zonal mean perturbations we would like to keep this illustration of the minimum and the maximum. For the future mitigation scenarios we only illustrate the maximum in order to have a clear signal and not duplicate the number of figures, but the minimum perturbation is also provided in the text. This way, the seasonal cycle is illustrated in Fig. 3, the zonal perturbations show the min and max

perturbations, and the budgets and forcings provide the annual mean perturbations more important for climate purposes. We hope this provides a consistent set of illustrations.

Line 657: since you discuss the future scenarios run with interactive chemistry, it would be interesting to know how future climate change might influence your results. What is the sensitivity of aviation forcings to future climate? Is the use of present-day meteorology justified here?

Indeed, in this study, we follow the methodology adopted in most of previous work and assume unchanged meteorology between present-day and future simulations. The aircraft perturbation is rather small and using a fixed meteorology allows to focus on the impact of aircraft emissions and to compare the present-day and future impact of aircraft emissions. A few studies accounted for climate change in their transportation (aircraft) perturbation studies (Olivié et al., 2012; Huszar et al., 2013). However, the impact of future climate change on aircraft forcings is not isolated in these studies (and it is probably difficult to do so). There is clearly a potential impact of future changes in upper tropospheric temperature, humidity and dynamics on aircraft perturbations but these effects need to be investigated more systematically in forthcoming studies. The importance of these changes at the 2050 time-horizon is probably small but this needs to be investigated. This limitation is now stressed in our conclusion (see below) and is mentioned as a perspective for future activities. In addition, this limitation is also briefly mentioned in the "Model set-up" section and in the paragraph spotted here by the reviewer.

Following previous work methodology, in order to better emphasize the impact of aircraft emissions on atmospheric composition, the impact of future climate change on aircraft perturbations is not considered in our simulations performed based on a present-day, unchanged, climate in 2050. The simulations performed by Olivié et al. (2012) and Huszar et al. (2013) did account for future climate change, however, the impact of future climate on atmospheric composition changes due to aircraft emissions is not isolated. Changes in upper-tropospheric temperature, humidity, and dynamics have the potential to affect the response of the atmosphere to aircraft emissions. These perturbations are likely to be more pronounced at a longer time-horizon than the 2050 timeframe considered in our simulations, but this topic is clearly a subject to be investigated in forthcoming studies.

Line 682: wouldn't it be more suitable to discuss the effects of the mitigation scenarios based on the annual change, not a one-month response?

As mentioned above, for the zonal mean perturbations we would like to keep this illustration of the minimum and the maximum of the perturbations. For the future mitigation scenarios we indeed illustrate the maximum solely in order to have a clear signal and not duplicate the number of figures, however the minimum perturbations in January are also cited in the text. In this way, the seasonal cycle is illustrated in Fig. 3, the zonal perturbations show the min and max perturbations (or only the max with the min cited), and the budgets and forcings provide the annual mean perturbations more important for climate purposes. The choice of May is better justified when introducing Fig. 5.

Line 795: 7.2 ---> 7.3

We checked this value and confirm that Holmes et al. (2011) in their Table 2 do provide a decomposition factor of 21.6 ± 7.2 mW/m^2/TgN.

Line 833: having fixed surface CH4 concentrations means your CH4 and OH interactions are constrained, so it is not obvious how you derived your CH4 feedback factor.

This methodology based on fixed surface methane perturbations has been described in Prather et al. (2001) or in various IPCC reports and has been commonly used and applied in numerous studies including Holmes et al. (2011) and Fiore et al. (2009). We have applied this commonly used method in our study and derive a feedback factor f=1.36 in agreement with this previous work and the recent IPCC AR6. Other methodologies exist (Khodayari et al., 2015; Pitari et al., 2016) as mentioned in our manuscript and the fixed surface mixing ratio method was found accurate to within 10%. This sentence has been revised in our manuscript in order to reemphasize this limitation.

Khodayari et al. (2015) concluded that for the simulations with fixed CH4 at the lower boundary condition, the parameterization based on Eq. (1) using the global mean lifetime approach overestimates the change in CH4 by 8.6% compared to the change calculated directly from the model using CH4 surface emissions. The overestimation is 12.1%-20.0% if using other lifetime approaches. They concluded that the parameterization based on Eq. (1) is good to within ~10% when using the global mean lifetime approach.

Line 875 and Table 5: your long-term ozone estimate is much smaller than what can be found in other studies, e.g., 47% (Wild et al., 2001), 58% (Kohler et al., 2008), 42% (Hoor et al., 2009), 51%, 43% (Pitari et al., 2016) of the CH4 RF. Most of the latest aircraft studies include 50% (IPCC AR4, Myhre et al., 2013) in their calculations. The same applies to your stratospheric water vapor; most studies calculate it to be 15% (Myhre et al., 2007) of the CH4 RF, while yours is around three times smaller. These need some clarification or/and justification.

The methane indirect forcings that we calculate (paragraph now moved to section 3.4) are 116 $mW/m^2/ppmvCH_4$ for the long-term O_3 forcing and 27 $mW/m^2/ppmvCH_4$ for stratospheric H_2O. In the recent AR6 Chapter 7 (Section 7.6) these forcings are estimated as 140 ± 70 $mW/m^2/ppmvCH_4$ for long-term ozone and 40 ± 40 $mW/m^2/ppmvCH_4$ for stratospheric water vapour. This implies a forcing smaller by 17% for ozone compared to the IPCC best estimate but well within the provided confidence level. For stratospheric H_2O, the forcing is subject to a larger uncertainty, and we derive a forcing lower by 32% compared to the IPCC best estimate but also well within the (large) confidence level. The reference and the IPCC values now available have been added in our manuscript in Section 3.4.

Line 906: based the Etminan ---> based on the Etminan

Typo corrected.

Line 946: and 2004 ---> and 2005

Typo corrected.

Line 888 and Table S3: the CH4 increase via the Etminan parametrization you report is 15%, and it is smaller than what other aircraft studies calculate (e.g., Grewe et al., 2019), Skowron et al., 2021). Any explanation for that? At the same time, a few paragraphs above (line 822), you mention the 24.5% increase. It is confusing.

The methane forcing we calculate (without indirect effects except OH feedback) of -11.02 mW/m^2 (Table 6) is actually larger than Skowron et al. (2021) (i.e., -8.4 mW/m^2, their Table 2) reflecting the uncertainty on OH changes in our models (LMDZ-INCA in this study and MOZART3 in their study). The effect of the new Etminan parameterization is 22% in their case. It is more complicated to compare with Grewe et al. (2019) because they start from a methane forcing based on a different emission inventory (Lee et al., 2009) but they derive an increase of the forcing of 23% when the revised Etminan formula is used. The 15% increase provided in our study refers to Table S3 and to the <u>total methane</u> forcing (including the indirect ozone and water vapour effects). The impact of the revised Etminan formula on the methane forcing only (no indirect effects other that OH feedback) is also equal to 23% in our calculations. The 24.5% increase has been calculated by Etminan for a halving of the present-day methane concentration and is cited as an example. A reference to Etminan is provided to avoid confusion when the 24.5% value is cited and the 15% value for the total methane forcing is also better stressed. In addition, the 23% increase for the methane direct forcing only is now mentioned for clarity.

Line 1055: considering significant uncertainties associated with ERF/RF factors (especially those related to NOx as based on just one study according to Lee et al., 2021), wouldn't it be better to compare RF numbers? It would also be consistent with Holmes et al. (2011) comparison. Another aspect is the nature of these studies, Lee et al. (2021) and Holmes et al. (2011) are multi-model ensembles, so maybe presenting also ranges that they report together with their best estimates might help your comparison to be more feasible.

This is indeed an excellent idea in order to illustrate the large uncertainties on the forcings. Table 7 and the corresponding text have been updated in order to include the RFs in the comparison. In addition, the ranges provided by Lee et al. (2021) are now also provided. Since the total forcings provided here are different from Lee et al. (2021) (not the same terms are included in this study), we recalculated the 5-95% percentiles based on the excel spreadsheet provided by these authors in order to use their discrete pdf distributions of the RFs and ERFs. A Monte-Carlo sampling similar to the one used by Lee et al. (2021) with 1 million sampling has been used to generate the total uncertainty. Please note that in this Table 7 we only provide the values of Lee et al. (2021) often cited as a reference publication. The Holmes et al. (2011) results (only available for NO$_x$) are however used and compared to Lee et al. (2021) and to the calculated forcings in Table S4 dedicated to the NO$_x$ terms.