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Interactive comment on "The impact of weather pattern and related transport processes on aviation's contribution to ozone and methane concentrations from NO_x emissions" *by* Simon Rosanka et al.

Anonymous Referee #2

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This work aims to provide insight into the effect that aviation NOx 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.

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 to other 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 al 2017c I recommend that such an analysis be added to Section 5.

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).

The approach used appears to quantify the direct effect that aviation NOx and H2O 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 NOx 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.

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?

At the end of section 2.1, it is stated that "50 trajectories [are initialized] at 6, 12 and 18 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?

Many of the conclusions seem to treat correlations as causal links (e.g. section 4.3). Some conclusions – such as that "during summer the O3 formation is limited by the background NOx concentration, whereas in winter low HO2 concentrations limit the total O3 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. More generally, much of the analysis is not very quantitative – such as page 14, lines 3-5 which states that a visual inspection of a correlation makes it "evident" that winter pattern 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.

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 HO2 reacting with O3 and NO, respectively". However, OH and HO2 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 HOx are. I am therefore skeptical of the claim that H2O is only a minor source of OH. I recommend that the authors rephrase this discussion to be about the OH/HO2 ratio and the production and loss of HOx than to talk separately about OH and HO2.

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).

The claim that "the standard deviation of background concentrations [of ozone and NOx] 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 NOx inventory, now 20-25 years old. Since total aviation NOx 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.

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.

What happened to WP2 in Table 1?

References

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