Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-601-RC1, 2019 © Author(s) 2019. This work is distributed under the Creative Commons Attribution 4.0 License.

Interactive comment on **"Tracer study to estimate the transport of cruise altitude aviation emissions in Northern Hemisphere"** ***by***
Lakshmi Pradeepa Vennam et al.

Anonymous Referee #1

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The authors are thankful to the reviewer for a thorough review and for raising several interesting and valid points that provided us an opportunity to clarify several aspects of this manuscript and improve it overall. Below are our responses to the reviewer comments and revisions to the manuscript when applicable.

Comment: Currently I am not able to judge whether the model is able to adequately model the dynamics of the tropopause layer. From Figure 3 I guess that the model vertical resolution is around 1 km and the model top is at 20 km. Please give a reasoning why CMAQ is capable to correctly treat the transport of species emitted at around 10 km. There is some indication that quite some counter-gradient transport might happen, since the model transports quite some amount of the tracer to altitudes of around 20 km, exceeding the surface values (Figure 3). How do you explain this transport pathway to such high altitudes in such short time?

Response: The CMAQ model has been used for numerous regional and global applications and is capable of fully capturing horizontal and vertical transport near tropopause and upper troposphere. In this study the model layer top is ~50 mbar and the vertical structure is similar to recent hemispheric CMAQ (H-CMAQ) studies (Mathur et al., 2017; Hogrefe et al., 2018), and now included as a table in Supplementary information. We used the same WRF meteorological data as were used in these prior studies with no layer collapsing which maximizes the consistency between H-CMAQ and WRF, and further reduces the vertical diffusion of the inter-continental transport. The UTLS dynamics are highly dependent on the reanalysis data used in the nudging and further details of the WRF configuration are discussed in Xing et al., 2015. Additionally in this application, higher vertical resolution (< 1km) was also used above the boundary layer (i.e., free and upper troposphere) than traditional CMAQ regional-scale applications to better resolve the tropopause dynamics. Mathur et al., 2017 briefly discussed the motivation behind 50 mbar model top (44 model layer structure) and highlighted that this layer structure is less diffusive in entraining the free-troposphere tracers to boundary layer. Recent hemispheric CMAQ studies extensively evaluated the free-troposphere and upper troposphere vertical profiles of various pollutant concentrations with in-situ aircraft measurements (Mathur et al., 2017; Vennam et al., 2017) and ozonesonde data (Xing et al., 2016; Hogrefe et al., 2018). These studies have indicated that H-CMAQ shows good vertical representation in the UTLS region when compared with observations, and vertical profiles look comparable to some other global models that consider the fully coupled stratosphere-troposphere dynamics. Overall this discussion reinforces our justification that H-CMAQ is capable of capturing the tropopause and upper troposphere

transport which is the central focus of this study. However, we acknowledge and mentioned these lines in the revised manuscript "*we acknowledge that there is no detailed assessment of the UTLS dynamics with this configuration and recommend that this be investigated further in future H-CMAQ studies.*"

We attribute the upward transport partly to the strong winds aloft that can horizontally transport and can eventually vertically advect some of the tracer mass to higher model layers. Also note that in Figure 3 we are averaging all horizontal grid cells concentrations at each altitude to calculate the mass fraction at each layer. Thus, in few grid cells if there is a upward draft it can transport the tracer mass from cruise altitudes to higher altitudes.
To address the reviewer's comment we incorporated these lines in the revised manuscript *"Some of the upward flux from cruise altitudes in such short time could also be due to vertical advection scheme that was used in our simulations. In few sub-tropical regions and Arctic region as shown in Figure 4, we are even injecting the CAAE tracers above the tropopause region (as the tropopause is lower in arctic regions ~ 8 km) which can vertically mix the tracer to upper altitudes. As discussed in Vennam et al., 2017 some of the CAAE can get transported along the isentropes to higher altitudes when higher isentropes show an upward pattern."*

Comment: While understanding the experimental set-up, I do not understand how to correctly interpret the simulation results. The quantity "Mass Fraction (surface layer)" is to zero at the beginning of the simulation. During the simulation the values increase constantly (as explained by the authors). The concentration increases everywhere in the model domain without limits and the ratio between the concentrations in the surface model layer to the column will converge to the model layer air mass to the column air mass and hence independent from the emission location. Latter because the difference between concentrations of model grid points concentration is getting small compared to the absolute steadily increasing concentration. Hence it looks arbitrary to me to take out any specific point in time. I think there is a principle problem in the interpretation of the results for this simulation set-up, which can only be resolved by a change in the simulation set-up. One possibility is to define a sink at the surface (deposition, ...) in a meaningful way. The simulation will converge to a quasi-steady state. This has also the advantage of having the possibility to check whether the results are in steady- state after three months. Referring to Grewe et al 2014 Figure 9, the water vapor temporal evolution for cruise emissions show larger time scales, which might question the assumption of achieving a steady-state after 3 months (or actually 2 months, see below).

Response: To interpret and discuss our results we selected "Mass Fraction" metric to highlight the tracer magnitudes in various altitudes relative to the total column. Since the emissions are released near the cruise altitudes, mass fraction near the surface should be zero at the beginning of the simulation. These cruise altitude emissions get transported with time to different model grids and altitudes, which is likely to increase the concentrations in the model domain. We agree with the reviewer that the concentrations can keep increasing near the surface if we run the model for longer time periods, as we did not consider any sink process in our tracer simulations. However, it is precisely for this very reason that we ran the model for 3 months in each season to quantify the

magnitudes of the cruise altitude tracers that could get transported during the typical transport time of pollutants in the atmosphere. We acknowledge that not having sink is one of the limitations in our study but we intentionally structured it such that we are focusing mainly on the tracer transport and isolate any deposited tracer mass on the surface. The change in the simulation setup and/or change in the model will give different results as the transport processes are highly dependent on some of these factors. To address reviewer's comment we added these lines in the revised manuscript "*A key limitation in this study is that we did not consider any sink process in the tracer simulations and incorporating sink process might have given us an opportunity to run for longer time periods instead of three-month simulation. We envision future studies to address this limitation to further advance our understanding the role of CAAE on surface air quality*."

We feel the 3-month simulation period that we considered is a reasonable time period as pollutants takes 1 – 2 days for vertical transport from PBL to the surface, ~1 week from mid-troposphere and ~1 month from tropopause (Jacob D.J, 1999, Figure 4-24). The horizontal transport in subtropics takes ~2 weeks and the transport from subtropics to tropics or towards poles takes ~1 – 2 months (Jacob D.J, 1999). Stohl et al., 2002 clearly indicated intercontinental transport occurs on timescales of weeks to 30 days and transport from lower stratosphere to lower troposphere can occur in range of ~90 days. Liang et al., 2009 demonstrated that it takes one month to cross the tropopause, one month to transport from upper troposphere to middle troposphere, and another one month to get transported to lower troposphere. Therefore, taking into consideration these relevant atmospheric transport timescales from the literature, we carried 90 days continuous tracer run in our tracer modeling to capture intercontinental, cross tropopause, and upper troposphere to lower troposphere transport processes.

Comment: Quantities are biased by the model resolution. Results are not given independently from the model resolution (see below), which inhibits a proper interpretation, even if the comment B wouldn't apply.

Response: The modeling results can vary with the considered spatial extent and temporal scales. We agree that the quantities could have been biased with the model resolution, but here we converted the concentrations in each layer to mass per area units (molecules/cm^2) considering the height of the each layer and compared with the total tracer column (molecules/cm^2) to estimate the relative fraction of tracer in different altitudes. We interpreted our results in mass-based units, which is an appropriate metric for the analysis that we showed in this paper.

Specific comments Abstract

Comment: p1 / l11 Please specify a bit more what kind of tracer you are referring to: inert gas- phase, inert particles with sedimentation, ..., what loss processes?

Response: We expanded our description of a passive tracer described in the introduction section. To enhance clarity, we explicitly included the phrase *"no chemistry and loss processes"* right next to passive tracer.

Comment: p1 / l12 Please explain in more detail what "tracer mass fraction means" in this con- text. Everything which is emitted in the atmosphere will eventually be deposited at the ground. From this perspective 100% would be expected. Near the surface is crucial. The smaller the volume the lower the percentage?

Response: To improve the clarity, we deleted the word "fraction" in the abstract (as we explained the metric clearly in the methodology section) and rephrased those lines as following "*Our results from Northern hemisphere simulations highlight that only < 0.6% of CAAE tracer mass get transported to the surface after 90 days of transport time considered in this study*" in the revised manuscript. In this context, tracer mass fraction is average percentage of CAAE tracer that can reach the surface through transport at worst-case conditions (no sink, no chemistry and continuous emissions).

We agree that everything that was emitted in the atmosphere will eventually get transported to the surface or undergo chemistry before getting deposited to the ground. With this setup, if we ran the model continuously for few months that would be the case. However we ran the model for four 90-day periods to represent each of the four seasons as a new model simulation. This gave us the ability to study the fate and transport of cruise altitude tracer in the 3-month period span during each season when chemistry and loss processes are turned off.

Comment: p1/l14 why "even"? It seems that the authors have expected something else. Please clarify this.

Response: We deleted the word "even".

Comment: p1/l16 Unclear. If something is emitted at 12 km it will always be deposited downwind. There is no direct instantaneous downward transport.

Response: Yes we agree with the reviewer that any pollutant emitted in the higher altitudes eventually gets transported, undergoes chemistry, gets deposited in the atmosphere and of course does not always undergo direct instantaneous downward transport. Since the main objective of this study is to quantify the amount of the passive tracer that get transported to the surface from cruise altitudes, so here in this line we are quantifying the source region CAAE contribution on the receptor region due to transport. To address this comment we included additional information in the revised manuscript "*The tagged source tracer simulations illustrated the source-receptor relationships and showed that ~10 – 50% source contributions can occur in downwind receptor regions due to transport when all other atmospheric processes are turned off*".

Introduction

Comment: p1/l20-22 There are indeed a couple of passive tracer studies, which were not included here, but a comparison might have been of interest. They are often not directly referring to the surface as a receptor region, but Figures are often including this information.

* Velthoven et al Atmospheic Environment 1997, is referred to below.

* Köhler, I., Sausen, R., Reinberger, R., Contributions of aircraft emissions to the at- mospheric NOx content, Atmospheric Environment, Volume 31, Issue 12, 1997, Pages
1801-1818,

* Danilin, et al. Aviation fuel tracer simulation: model intercomparison and implications, Geophys. Res. Lett., 25 (1998), pp. 3947-3950

* Schoeberl, M.R., Morris, G.A., 2000. A Lagrangian simulation of supersonic and subsonic aircraft exhaust emissions. Journal of Geophysical Research 105, 11,833–
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* Rogers, H. L., H. Teyssedere, G. Pitari, V. Grewe, P. van Velthoven, J. Sundet, Model intercomparison of the transport of aircraft-like emissions from sub- and supersonic aircraft, Meteorol. Z., 3, 151-159, 2002.

* Grewe, V., Reithmeier, C. and D.T. Shindell, Dynamic-chemical coupling of the upper troposphere and lower stratosphere region, Chemosphere: Global Change Science, 47, 851-861, 2002.

Some of this is referred to later in the text, but should be clarified already here, since the impression is given that those studies do not exist.

Response: We thank the reviewer for bringing these papers to our attention. Firstly, we did not include some of this literature in the paper as few of these papers focused on the supersonic emissions which is outside the focus area of our study. But however we appreciate the reviewer's suggestion, and thus included few of them as shown below in the introduction text whenever they seem relevant. However, none of the papers cited above quantified the % of CAAE that can get transported to the surface.

"The tagging approach (Grewe et al., 2017) and passive tracer modeling were also implemented in a few prior aircraft related studies to emphasize the role of transport on the CAAE emissions as discussed below".

"Few earlier as well as recent studies (Brasseur et al., 1998; Grewe et al., 2017) discussed the relative contribution of air traffic emission source to the total pollutant air quality concentrations using tagging approach and conducted brief analysis on this topic in their overall study. However they did not quantify the amount of CAAE that can get transported to the surface and the role of aircraft emissions in the intercontinental transport which is the focus of this study".

Comment: p2 / l5-19 The text might suggest that aircraft emission tagging approaches were not used previously. However, there are studies 20 years back (Brasseur et al 1998) or recently Grewe et al 2017, who use such approaches. Please clarify the text.

* Brasseur et al, European scientific assessment of the atmospheric effects of aircraft emissions, Atmospheric Environment, 32, 1998, 2329-2418 (Figure 35)

- Grewe, V., Tsati, E., Mertens, M., Frömming, C., and Jöckel, P., Contribution of emis- sions to concentrations: The TAGGING 1.0 submodel based on the Modular Earth Sub- model System (MESSy 2.52), Geosci. Model Dev. 10, 2615-2633, doi:10.5194/gmd2016-298, 2017.

Response: We included some of the 3-D modeling related passive tracer studies in the manuscript that seem relevant to our study. We also want to clarify that our intention is not to suggest that the tagging approaches were not used in aircraft emission studies, rather to highlight that this is the one of the first studies to tag the key aviation emission source regions such as North America, Europe and East Asia and study the source-receptor contributions of these emissions. The two studies (Brasseur et al., 1998; Grewe et al., 2017) that the reviewer pointed out are also now referenced in revised manuscript as shown in an earlier response.

Comment: p3/l5 I suggest to use "atmospheric transport (resolved, parameterised and unre- solved)" instead of "dynamic", since the role of turbulence, diabatic heating, etc. is not investigated.

Response: We agree with this comment, and have incorporated the change as suggested.

Comment: p3 l7-9 I do not understand the importance and significance of this approach. There are several effects mixed. Continuous emission lead to continuous increase of the concentrations. At any time, the surface concentration is a snap shot and mixes emissions at time T0, where a lot of the emitted species may have reached the surface with emissions at time T0+90days, where no contribution to the surface concentration is expected. Further, since the tracer is not deposited it remains in the atmosphere and artificially increases the concentration, which disagrees with the worst case assumption.

Response: Yes we agree that continuous emissions can lead to continuous increase of the concentrations. In this study we consider the same continuous emissions scenario without chemistry/deposition as our worst-case scenario which provides us the opportunity to assess the influence of transport alone on cruise altitude emissions. The main objective of this worst-case scenario is to see how much of the T0 emissions get transported to the surface in a T0+90 days transport time. We believe that any passive tracer simulations are artificial modeling exercises as that wouldn't be the case in the actual atmosphere since both chemical and physical processes drive the fate of the pollutant. Since we did not consider deposition, we intentionally ran the model for only 3-month period (typical transport times as mentioned in the literature) to understand the transport and accumulation of these tracers.

Methodology

Comment: Section 2.1: Nothing is said about the vertical extend of the domain, number of layers and especially the resolution at tropopause levels. This is an extremely important point. Large-scale transport as well as diffusivity of the transport scheme

might give largely different results for too low vertical resolution or a too low model top. The large concentrations at 20 km might indicate such problems.

Response: We agree with this gap in information provided in our Methods, and have now included the vertical structure of the model in the Supplementary information in the revised manuscript. We included additional details regarding the vertical resolution in our response to the first comment in this document. We have 44 model layers with a 600 – 800 meters resolution near the cruise altitudes (9 – 12km), much finer near the boundary layer with 50 – 100 meters resolution, and between the boundary layer and the cruise altitudes the resolution is the range of 200 – 500 meters. So overall the vertical resolution is fairly fine enough to resolve the transport patterns in the atmospheric altitudes of interest in this study.

Comment: Section 2.2 p4 114 why "only"? Is there a disadvantage in this approach?

Response: No, it is not a disadvantage. It is only to emphasize how we structured the tagged tracer spatial extent.

Comment:

Equation (1): I strongly advise the authors to convert this mass fraction from an extensive to an intensive quantity. In the current version "Mass Fraction (MF-layer)" is dependent on the model resolution, which is not given (see above). For example a thick layer at 500 hPa will give a large number, not because there is a lot of tracer mass in terms of concentration, but solely because the model layer is thick. One possibility is to further divide by the layer thickness to obtain %/km as unit. The consequence is that the results are resolution independent, better to be interpreted and can be compared to other model results. In Figure 3, e.g., the total amount can be obtained by summing up the values. When MF is divided by the layer thickness, the total amount can be obtained by integration.

Response: We should clarify that in the Mass Fraction calculation we did consider the model layer thickness when converting the concentrations to mass per area term and note that the units of mass in each layer is molecules/cm^2. The Mass Fraction % is the relative mass available in each layer with respect to the total mass column (calculated by integrating the concentrations for all layers). The MF% in all layers sums up to 100% , so it is easy and convenient for the reader to understand CAAE tracer % that got transported from cruise altitudes to surface and other altitudes. Therefore we decided to retain the MF metric in the manuscript.

However, to be responsive to the reviewer's suggestion, we calculated the %MF/km metric, and included a new Figure 1 at the end of this document. Overall since we are normalizing with the layer thickness, the values seem small. As the reviewer mentioned the total amount can be obtained by integration. The %/km metric shows higher values near the surface layer than the cruise altitude layer which doesn't look correct as the actual concentrations shows different trend (as shown in Figure 4 of the manuscript).

Comment: p5l11 I think, by taking the last 30 days out of a 90 days simulation period, you implic- itly assume a spin-up of 60 days and 30 days simulation, right? Hence, actually you assume a much smaller mixing time than suggested in Section 2. I suggest being a little bit more specific about the lifetimes used. For example if you assume that 95% of the air is mixed after 60 days, this will result in an e-folding mixing lifetime of 20 days, which is certainly too low. A 60 day e-folding mixing lifetime will result in a 95% mixing after 180 days, which would require a much longer simulation time.

Response: As we mentioned in earlier responses, the transport time that we considered are based on the literature we have cited. We quantified and presented figures for all three months for various metrics, but for few calculations (e.g. tracer mixing ratios presented in Figure 4 [and source-receptor contribution metric presented in Figure 8]), we presented only the last month's values to provide overall final numbers after giving sufficient time for transport to occur in the model.

Comment: Figure 1: Units are actually moles/s/gridbox. Please provide Figures, which are not dependent of the chosen resolution, e.g. moles/s/m2 or moles/s/m3.

Response: We realize that rather than moles it is useful to show the emission plot in mass terms (i.e., tons/year) as shown in Figure 1 of the manuscript. Our intent here is to show the overall spatial representation of the emissions rather than actual quantitative numbers so we feel tons/year/gridbox serves better for this purpose than moles/s/m2. Also, we believe that this approach to present mass/unit time without area normalization has been used in numerous ACP publications. And finally, since these emission plots are vertically summed values (column totals), moles/s/m3 is not applicable here.

Comment: p5 l23: How well do you simulate tropopause folds and the associated strat-trop exchange?

Response: We discussed the tropopause dynamics in reviewer's first comment. As mentioned before, H-CMAQ showed good representation of the pollutant vertical profile when compared with observations. Since WRF and H-CMAQ use a rigid "lid" on the top, they might not fully capture the complete stratosphere-troposphere exchange but as demonstrated by Xing et al (2016), the models are capable of simulating the tropopause folds. However, we do concur that there is a need for future studies with detailed evaluation of UTLS dynamics and tropopause folds.

Comment: p5 l23ff: The discussion is very speculative. I think you should be able to support your arguments with your simulation results. E.g. showing the tropopause fold etc.

Response: Here instead of calculating the tropopause folds we supported our findings with previous studies that showed the same trend due to tropopause folds. We corroborated our findings with another recent study (Akritidis et al., 2016) that showed the role of tropopause folds occurred at Mediterranean region during summer season.

However, performing a detailed tropopause fold calculation is beyond the current scope of this study, and might not yield anything new.

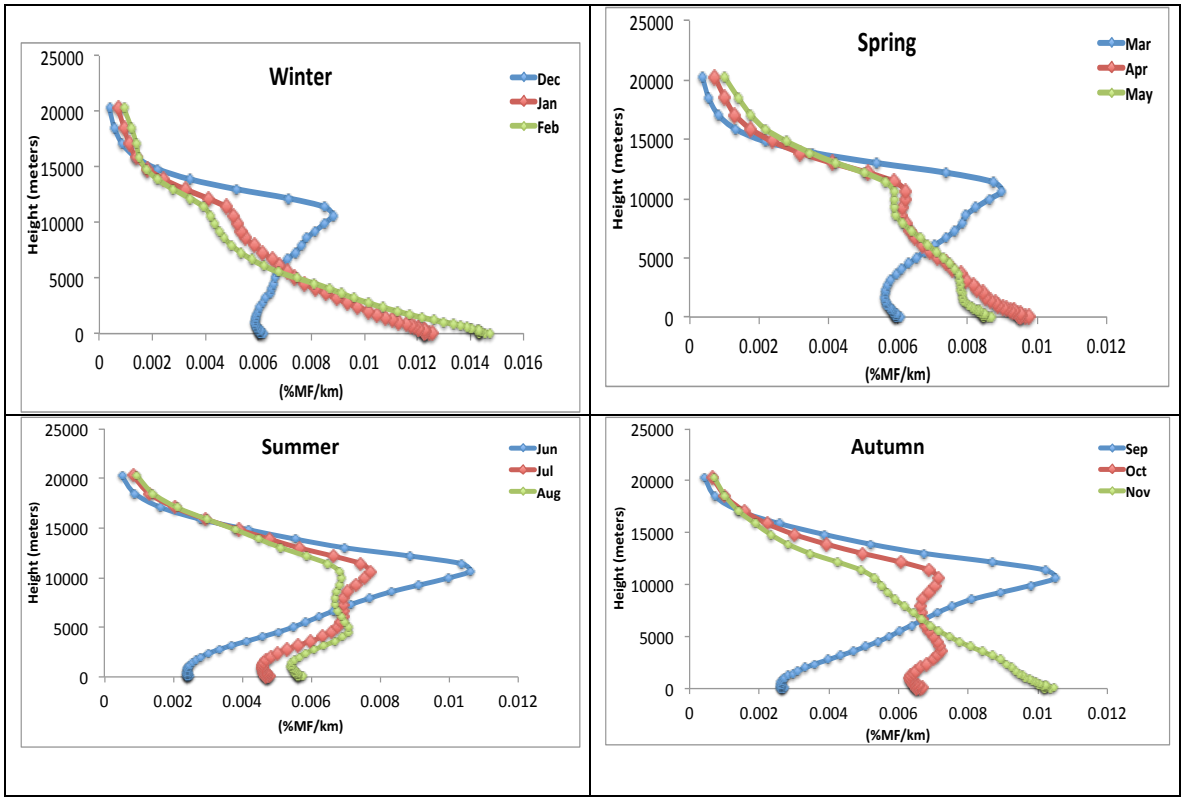

Figure 1: The overall model domain vertical profile of tracer mass fraction (%/layer thickness) at different altitudes (points on the line) in the model for each month by season.

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