Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-601-RC2, 2019

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 #2

Received and published: 13 January 2019

Comment: The paper discusses the dispersion of emissions of aviation from cruise altitudes in the Northern Hemisphere atmosphere. The paper considers the emissions as given in a data set as provided by FAA and Volpe for the ACCRI project [Wilkerson et al., 2010; Brasseur et al., 2016]. The emissions are treated as passive tracers, without any removal process in the atmosphere. The emission are followed over several seasons (3 months periods), starting from zero initial concentrations. The model considers tracer transport by advection with the resolved wind field of a hemispheric global model and by diffusion from a convective mechanism. I do not know this model and information is given in this paper about this model only in terms of a few references and some resolution information.

Response: We thank the reviewer for the thorough review and providing valuable comments that helped us to improve the manuscript. We provide below detailed responses to the review comments, and revisions to the manuscript where required.

The Community Multiscale Air Quality (CMAQ) model used here is primarily developed by the U.S. EPA, and has several thousands of users across the world from over 50 countries. According to Google Scholar, there are over 5000 publications since the year 2000 that refer to this model, and the model has gone through five different external peer reviews of its science during the development stages. As of date, there are over 25,000 downloads of the model across the world. An overview of the model, its science components and applications, and the global user community is available at: https://www.epa.gov/cmaq. A near-comprehensive list of peer-reviewed publications from 2000 – 2018 is available at: https://www.epa.gov/cmaq/cmaq-publications-and-peer-review.

To address the model and resolution information comment, we added some more detailed description about the model and the resolution used in methodology section as shown below "*The state-of-the-art EPA's Community Multi-Scale Air Quality (CMAQv4.7.1,doi:10.5281/zenodo.1079879) chemistry-transport model (Byun and Schere 2006) was used over a Northern hemispheric-wide domain at a grid resolution of 108 × 108 km^2 that has spatial extent as shown in Figure 1 with 44 vertical layers of variable thickness between surface and 50 mb (Table S.1). The CMAQ model has been extensively used in numerous urban-to-regional scale air quality studies globally for both*

research as well as regulatory applications to study the formation of several pollutants including ozone, fine particulate matter and air toxics [Foley et al.,2010; Appel et al., 2017; Astitha et al., 2017; Zhang et al., 2018]. In this study we used the new hemispheric CMAQ (Xing et al., 2015; Mathur et al., 2017) that has the capabilities to address long-range and intercontinental hemispheric pollution transport. The extended hemispheric CMAQ has also been evaluated against surface as well as aloft observation data (Mathur et al., 2017; Vennam et al, 2017; Hogrefe et al., 2018) and model processes were also examined for the new larger spatial scales. We turned off all chemistry and deposition processes in the model and turned on only the transport processes to perform the tracer simulations."

Comment: So the paper studies how a passive tracer emitted from a more or less continuous source near the tropopause accumulates in and gets distributed over the atmosphere within 3 months periods for various seasonal meteorological conditions. The paper studies the mass fraction of tracers in model layers and in various source and receptor domains including a surface layer (of unknown vertical thickness).

Response: In the revised manuscript we included the vertical structure (as mentioned in the previous comment) used in the model along with the surface layer thickness information in the Supplementary document Table S.1 and also referenced the previous studies that provided detailed layer description in the manuscript.

Comment: The paper aims to investigate physical processes in transporting cruise altitude emissions in the atmosphere. However, I cannot learn anything about physical processes except that they vary with season and altitude, and that convection may be important in summer. That is not new.

Response: The main objective of this paper is to quantify the magnitudes of CAAE that reach surface layer due to transport and the influence of CAAE from source regions on the receptor regions. However, in a recent study (Vennam et al, JGR 2017), we were able to carefully quantify the air quality contributions (O_3 and $PM_{2.5}$) of CAAE at the surface both in North America and in Northern Hemisphere and show the vertical transport pathways through isentropic analysis. Since CAAE occur mainly near the tropopause, a region where isentropic mixing/transport is important and highly influenced by potential temperature, Vennam et al (2017) studied the isentropic-based aircraft-attributable concentrations for all seasons to understand the transport processes. The detailed physical processes responsible for the transport in general have been studied as part of the algorithm development in CMAQ and published elsewhere (Pleim et al., 2007a,b). However, Vennam et al did not separate the role of transport alone in how CAAE may affect surface layer concentrations.

In the current study, we were able to clearly illustrate that even at worst-case conditions (i.e., no chemistry or deposition, and with continuous CAAE at cruise altitudes) by using fine-scale horizontal grid resolution (~4 – 6 times finer than typical global models (Whitt et al., 2011)) and finer vertical resolution, only insignificant fraction of CAAE reaches the surface due to transport. Here we were also able to quantify the contribution of CAAE

from source region on the receptor regions that occurred due to intercontinental transport. Both these findings are new and were not addressed in the previous tracer studies (Kohler et al., 1997; Grewe et al., 2010). We thus believe that this paper is a valuable and unique contribution, and advancing the knowledge in understanding the role of cruise altitude aviation emissions on surface impacts.

Comment: The paper claims to be the first in using a "tagged tracer simulation" to quantify source- receptor relationships. Tagging is needed to follow the fate of tracers in a nonlinear system [Grewe et al., 2010]. In this study, the tracer transport is linear in the concentration values. A doubling of the sources causes a doubling of the concentrations. In this case, emissions from various sources can be treated independently of each other and tagging is trivial. Similar studies of the dispersion of NOx as a passive tracer from various sources, with linear chemistry, have been presented, e.g. by Ehhalt et al. [Ehhalt et al., 1992] and Köhler et al. [Köhler et al., 1997], long ago.

Response: Our intent is not to say that this is the first tagging study but to highlight that this is one of the first studies to tag cruise altitude aircraft emissions by region to study the source-receptor relationships of the CAAE tracers. To be more specific, we modified the line in the revised manuscript as following: "*this is the first study to use tagged tracer simulations for these high aircraft activity regions to illustrate the role of intercontinental transport*". The studies that the reviewer cited are different in many aspects (such as model configuration, aircraft emissions and modeling methodology) from our study. Kohler et al., 1997 considered NOx emissions with simplified linear chemistry so it is not a passive tracer study, and we already included some other additional passive tracer study references in our manuscript. Ehhalt et al., 1992 developed a very simplified 2-D approach with uniform vertical wind and their main intention was to demonstrate the importance of aircraft to NOx upper troposphere budget, which is different from what we are trying to address in this study with a 3-D model passive tracer modeling application. Note that these two studies are over 2 decades old, and since then model algorithms, emissions inventories and transport schemes are significantly advanced and improved, so it is important to address some of these research questions with evolving new modeling systems. One such example is aircraft emission inventory; previous studies used old gridded aircraft emission inventories whereas we used the new chorded highly-resolved gate-to-gate aviation emissions inventory from AEDT (Wilkerson et al., 2010) that provides better spatial and temporal representation of this source sector in the modeling system. And finally, given the growth in global aviation activity, in recent years, we find further motivation for a study like ours.

Comment: So, this is an academic study. That would be acceptable if done well. However, I also have technical problems:

Response: We did not understand the reviewer comment about academic study completely. We tried our best to address all specific comments and concerns that are explicitly pointed out by the reviewer in our responses in this document, and in the revised manuscript.

Comment: What is the vertical resolution? How thick is the surface layer? What are the time step sizes? Which process is simulated by asymmetric diffusion?

Response: In the revised manuscript, we added the layer structure in the supplementary info. Since WRF and thus CMAQ follow a sigma-coordinate system, note that the surface layer thickness varies both in space and time, and we thus provided the average surface layer thickness of ~20 meters in the manuscript. The time step size in CMAQ is considered small enough to ensure positivity and numerical stability of the solution, which satisfies the Courant- Friedrich Lewy (CFL) condition (Byun et al., 2006). Typically the time step size is 12 minutes, and the model runs the chemical and physical processes for each of this timestep and gives both an average and instantaneous concentration at the end of each hour. The asymmetric convection model (Pleim et al., 2007a,b) considered simulates the vertical diffusion in the model and Yamartino scheme (Byun et al., 2006) is chosen for the vertical and horizontal advection processes.

Comment: Page 4, line 3: why do you mention water vapor. Why not CO2?

Response: We mentioned water vapor as it is one of the highly emitted pollutant followed by NOx and CO2. In the revised manuscript to provide additional insight to the readers we included all these three pollutants. Revised line "*we considered emissions of NO_X as our passive tracer since it is one of the highly emitted pollutants at cruise altitudes from aircraft, besides CO_2 and water vapor*". Furthermore, both water vapor and CO_2 are related to potential climate impacts, and given the focus of our study on surface layer concentrations (potentially related to air quality and public health), we had initially omitted CO_2.

Comment: More general, why do you talk about NOx emissions when you simulate the emissions as a passive tracer? NOx has a lifetime of typically 5 days in the free troposphere, and often much shorter near the surface. Thereafter, most NOx is converted to HNO3 and other species after a few days. CO2 would be closer to the passive tracer concept.

Response: The reason we considered NO_x emissions is to better represent the spatial representation of the CAAE emissions that are related to surface air quality and we also mentioned this point in the revised manuscript. "*The rates of emissions of these tracers were based on actual cruise altitude NO_X emissions estimates from AEDT, we considered NO_X to better represent the spatial as well as temporal variation of aviation emissions in the upper layers of the atmosphere*" . Furthermore, whether it is NO_x or CO_2, once it is treated as a passive tracer, the lifetimes are immaterial.

Comment: I am sceptical about the conservation properties of the model in this study. The paper talks about the amount of a species measured in moles. But I would expect that one should discuss a conservative concentration measure like the molar mixing ratio of the tracer (number of moles of the tracer per mole of air).

Response: The CMAQ model is formulated with vertical advection and diffusion schemes; the transport algorithms are well tested to ensure that the continuity equation is calculated to meet convergence conditions and mass is conserved (Byun et al., 2006).

We did not understand clearly the reviewer comment regarding species measured in moles. In CMAQ, the input emissions are in terms of moles for gas-phase species (or grams for particles) and converted into concentrations internally in the model. The final model outputs are in terms of mixing ratio (parts per million by volume, ppmV) and we considered these quantities to calculate mass fraction and tracer contribution in the results.

Comment: Fig. 1 presents emissions in units of moles/s. This is a species abundance source rate. In order to assess this, one needs to know the respective air volume in which the emissions occur.

Response: We may have inadvertently confused the reader here. To avoid further confusion and for completeness, we updated Fig1 (included at the end of this document) with annual emissions total plot (and changed the units to tons per year) in the revised manuscript. In CMAQ the emissions are in moles or grams (based on gas or particulate matter) and the model converts the emission units into output concentrations (mixing ratio, ppmV) considering all the necessary units conversion including the air volume.

Comment: When computing mean values, do the authors weigh the results with the volumes or do they add concentrations from small grid cells (near the poles and near the surface) with the same weight as sources from large grid cells (in the tropics and in the upper atmosphere)? That is not clear presently.

Response: We did not area-weight the concentrations and we equally weighted all the grid cells. However, since we converted the mixing ratio (concentrations) into molecules/cm^2 (converted into area basis instead of volume) we did consider the height of the grid in the units conversion. So that should take into account the depth of the grid in the computing mean values for each model layer and the same applies for the total vertical column calculation. Redoing the calculations with area weights is a potential refinement to this approach in the future.

Comment: Fig. 3, winter, shows a maximum of mass fraction forming in the lower troposphere, i.e., in a region without sources. How can that happen? Yes it can happen temporarily when advection dominates relative to diffusion processes. When averaged over longer period, it should not happen. However that is not discussed. I have the impression that the model violates conservation laws.

Response: The mass fraction in lower troposphere is due to the transport of the tracer from tropopause to the lower altitudes in the model. In winter, due to high westerly transport the horizontal transport in the cruise altitudes dominates and brings the tracer to low source region which can get transported to the lower altitudes simultaneously. As discussed in Vennam et al., 2017, during winter season higher isentropic surfaces get

closer to the lower troposphere isentropes, which indicates that the vertical downward transport along the isentropes are enhanced. During summer seasons, higher isentropic surfaces show an upward pattern that transports CAAE tracer to lower stratosphere.

The CMAQ model is formulated with vertical advection and diffusion schemes, the transport algorithms are well tested to ensure that the continuity equation is calculated to meet convergence conditions and mass is conserved. Both CMAQ and WRF (model used for meteorological inputs) are formulated to satisfy continuity equation and mass consistent advection (Byun et al., 2006; Pleim et al., 2007a,b).

Comment: The paper is good in citing many related studies. In fact, I was not aware on many of them. But it appears somewhat random in the selection of references (those of major and minor relevance for this paper). There are many other important studies which dealt with tracer or aviation emission transport in the global atmosphere earlier or more complete. Examples are as follows: Ehhalt et al. [1992]; Danilin et al. [1998] , Forster et al. [2003], Koehler et al. [1997], Brasseur et al. [1996], Brasseur et al. [1998], Gauss et al. [2006].

Response: We thank the reviewer for pointing out this inconsistency. In the revised manuscript we included some of these additional references, and streamlined the literature section.

Comment: In summary, the paper in its present form does not satisfy the quality criteria of ACP. I just looked at the paper Vennam et al. (JGR, 2017), cited in this paper, which just appeared. It seems that this is far more advanced. It includes the chemical processes that are mentioned in the outlook of the ACPD paper. So, I am not convinced that the present paper is still needed.

Response: We regret that the reviewer states that this paper does not satisfy the quality criteria of ACP. While we are unsure of the specific criteria that may be referred to in this comment, we strongly believe that this is a well-founded study using a model with strong scientific credibility (with > 5000 publications to date with a global user base) on a key emissions source sector with robust conclusions that contributes to the growing body of literature on aviation air quality research, and more importantly a very relevant paper for the ACP audience. Vennam et al., 2017 (JGR) (published online 22 December 2017) studied full-flight aviation emissions (cruise altitude + landing and takeoff) impacts on the surface and the sensitivity of the grid resolution on those impacts. However that study was not able to isolate the influence of transport of cruise altitude aviation emissions (CAAE) on surface. Given that CAAE constitute a significant portion (~75% of fuel burn occurs at cruise altitudes) of total aviation emissions, and uncertainties in the cruise altitude impacts on surface, it is important to study this topic further. And in this present study we were able to specifically isolate the role of transport on CAAE emissions and their contribution at various altitudes and key source regions in the northern hemisphere using a fine scale model resolution (4 – 6 times finer than typical resolution used in most global models) and highly-resolved updated emission inventory with actual radar tracking (Olsen et al., 2013) compared to the previous studies. We thus strongly believe that this

manuscript fits the ACP criteria as it addressed some really key issues in the aviation research and advanced the current understanding of the topic studied here, focused on source – receptor relationships for cruise altitude aviation emissions.

From the ACP Subject areas, our paper focuses on the following shown in bold, again emphasizing the suitableness of this paper to the ACP audience:

Subject	**Gases**, Aerosols, Clouds and Precipitation, Isotopes, Radiation, Dynamics, Biosphere Interactions, Hydrosphere Interactions
Research Activity	Laboratory Studies, Field Measurements, Remote Sensing, **Atmospheric Modelling**
Altitude Range	**Troposphere, Stratosphere**, Mesosphere
Science Focus	Chemistry (chemical composition and reactions), **Physics (physical properties and processes)**

References:

1) Brasseur, G. P., J.-F. Müller, and C. Granier (1996), Atmospheric impact of NOx emis- sions by subsonic aircraft: A three-dimensional model study, J. Geophys. Res., 101, 1423-1428, doi: 10.1029/95JD02363.

2) Brasseur, G. P., R. A. Cox, D. Hauglustaine, I. Isaksen, J. Lelieveld, D. H. Lister, R. Sausen, U. Schumann, A. Wahner, and P. Wiesen (1998), European scientific assessment of the atmospheric effects of aircraft emissions, Atmos. Env., 32, 2329 - 2418. Brasseur, G. P., et al. (2016), Impact of aviation on climate: FAA's Aviation Climate Change Research Initiative (ACCRI) Phase II, Bull. Amer. Meteorol. Soc., 97, 561- 583, doi: 10.1175/BAMS-D-13-00089.1.

3) Danilin, M. Y., et al. (1998), Aviation Fuel Tracer Simulation: Model Intercomparison and Implications, Geophys. Res. Lett., 25, 3947 - 3950.

4) Ehhalt, D. H., F. Rohrer, and A. Wahner (1992), Sources and distribution of NOx in the upper troposphere at northern mid-latitudes, J. Geophys. Res., 97, 3725 - 3738, doi: 10.1029/91JD03081.

5) Forster, C., A. Stohl, P. James, and V. Thouret (2003), The residence times of aircraft emissions in the stratosphere using a mean emission inventory and emissions along actual flight tracks, J. Geophys. Res., 108, 8524, doi: 10.1029/2002JD002515.

6) Gauss, M., I. S. A. Isaksen, D. S. Lee, and O. A. Søvde (2006), Impact of aircraft NOx emissions on the atmosphere – tradeoffs to reduce the impact, Atmos. Chem. Phys., 6, 1529–1548.

7) Grewe, V., T. Eleni, and P. Hoor (2010), On the attribution of contributions of atmospheric trace gases to emissions in atmospheric model applications, Geosci. Model Dev., 3, 487-499, doi: 10.5194/gmd-3-487-2010.

8) Koehler, I., R. Sausen, and R. Reinberger (1997), Contributions of aircraft emissions to the atmospheric NOx content, Atmos. Env., 31, 1801-1818.

9) Wilkerson, J. T., M. Z. Jacobson, A. Malwitz, S. Balasubramanian, R. Wayson, G. Fleming, A. D. Naiman, and S. K. Lele (2010), Analysis of emission data from global commercial aviation: 2004 and 2006, Atmos. Chem. Phys., 10, 6391-6408, doi: 10.5194/acp-10-6391-2010.

10) Olsen, S. C., Wuebbles, D. J., & Owen, B. (2013). Comparison of global 3-D aviation emissions datasets. Atmospheric Chemistry and Physics, 13, 429–441.

11) Whitt, D. B., Jacobson, M. Z., Wilkerson, J. T., Naiman, A. D., & Lele, S. K. (2011). Vertical mixing of commercial aviation emissions from cruise altitude to the surface. Journal of Geophysical Research, 116(D14), D14109.

12) Pleim, J.E., 2007a: A Combined Local and Nonlocal Closure Model for the Atmospheric Boundary Layer. Part I: Model Description and Testing. *J. Appl. Meteor. Climatol.,* **46**,1383–1395, https://doi.org/10.1175/JAM2539.1

13) Pleim, J.E., 2007b: A Combined Local and Nonlocal Closure Model for the Atmospheric Boundary Layer. Part II: Application and Evaluation in a Mesoscale Meteorological Model. *J. Appl. Meteor. Climatol.,* **46**, 1396–1409, https://doi.org/10.1175/JAM2534.1

14) Byun D, Schere KL. Review of the Governing Equations, Computational Algorithms, and Other Components of the Models-3 Community Multiscale Air Quality (CMAQ) Modeling System. ASME. Appl. Mech. Rev. 2006;59(2):51-77. doi:10.1115/1.2128636.

15) Appel, K.W., Napelenok, S.L., Foley, K.M., Pye, H.O.T., Hogrefe, C., Luecken, D.J., Bash, J.O., Roselle, S.J., Pleim, J.E., Foroutan, H., Hutzell, W.T., Pouliot, G.A., Sarwar, G., Fahey, K.M., Gantt, B., Gilliam, R.C., Heath, N.K., Kang, D.W., Mathur, R., Schwede, D.B., Spero, T.L., Wong, D.C., & Young, J.O. (2017). Description and evaluation of the Community Multiscale Air Quality (CMAQ) modeling system version 5.1. *Geoscientific Model Development, 10*(4), 1703-1732. doi: 10.5194/gmd-10-1703-2017

16) Astitha, M., Luo, H.Y., Rao, S.T., Hogrefe, C., Mathur, R., & Kumar, N. (2017). Dynamic evaluation of two decades of WRF-CMAQ ozone simulations over the contiguous United States. *Atmospheric Environment*, 164. doi: 10.1016/j.atmosenv.2017.05.020

17) Foley, K., Roselle, S.J., Appel, K.W., Bhave, P., Pleim, J.E., Otte, T.L., Mathur, R., Sarwar, G., Young, J.O., Gilliam, R.C., Nolte, C.G., Kelly, J.T., Gilliland, A., & Bash,

J.O. (2010). Incremental testing of the Community Multiscale Air Quality (CMAQ) modeling system version 4.7. *Geosci. Model Dev., 3*: 205-226. doi: 10.5194/gmd-3-205-2010

18) Zhang, Y., Mathur, R., Bash, J. O., Hogrefe, C., Xing, J., and Roselle, S. J.: Long-term trends in total inorganic nitrogen and sulfur deposition in the US from 1990 to 2010, *Atmos. Chem. Phys*., 18, 9091-9106, https://doi.org/10.5194/acp-18-9091-2018 , 2018.