A Plume-in-Grid Approach to Characterize Air Quality Impacts of Aircraft Emissions at the Hartsfield-Jackson Atlanta International Airport

Responses to James Kelley (Reviewer #1)

We thank Dr. James Kelley for providing thoughtful comments. We have responded to each comment below, and have noted the section number for each revision to the manuscript. Each comment by Dr. Kelley is reproduced below, in bold type. Our responses appear below each comment, indented.

The maximum incremental impact of aircraft emissions in the PiG simulation is quite large (42.1 ug/m3), and I have no way of knowing if it is realistic. As the authors mention, their approach of placing fixed emitters at points along flight segments could artificially concentrate emissions and potentially lead to over-estimates of maximum impacts. If the dilution and mixing of the puffs in the airport environment were also underestimated due to use of grid-scale meteorological information, overly concentrated puffs could produce large over-estimates in maximum impacts. The model evaluation in this study (and most other PiG studies) uses routine network observations that do not provide data in highly concentrated regions of the plume, and so the PiG treatment is never evaluated against observations for the conditions where maximum impacts occur. On p. 113 (para. 2) and elsewhere, the authors highlight the value of their approach for examining the maximum impacts on air quality. These types of statement should be better qualified given the wide range of uncertainties in emissions and other inputs along with the general lack of fine-scale evaluation of PiG models in the literature. In section 6, where the authors suggest areas for future work, they should also place an emphasis on evaluating PiG simulations with in-plume data from previous and future field campaigns. If PiG models are to be used for estimating maximum impacts in practical applications, then we need to develop more confidence that the sub-grid-scale algorithms and processes are operating correctly.

> The figure you cite for the maximum incremental impact of aircraft (42.1 µg/m³) is the maximum concentration reported in a single puff in a single hour across the entire model run. In our opinion, this extreme outlier is not a good reflection of the typical maximum impacts of aircraft. Figure 6 shows that the maximum puff concentrations near the airport, *when excluding outliers*, tend to be about 1.75 µg/m³. Outliers are defined as values more than 1.5 times the inter-quartile range above the 75th percentile and below the 25th percentile, collectively representing 0.7% of a normally distributed dataset (MATLAB's standard definition for outliers). We believe that these figures are a better reflection of the likely or typical maximum impacts from aircraft. We have edited the caption of Figure 6 to explain the mathematical definition used to characterize outliers in our study.
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> You highlight our comment that using an emitter to represent a segment along a flight path may artificially concentrate emissions. This is true, but this assumption

is unlikely to result in one or two extreme outliers, since this effect would be present in all timesteps and would apply to all puffs. Rather, the more likely consequence of this assumption would be to increase concentrations of all puffs by a far smaller amount. An extreme outlier may be explained by a very rare conjunction of physical factors that cause many puffs to overlap and merge, or possibly a combination of chemical conditions that led to high production of PM$_{2.5}$.

In Figure 7, which depicts the speciated composition of maximum puffs and grid cell contributions, we wish to emphasize the speciated breakdown rather than the maximum values. After reading your comment, we realize that because Figure 7 depicts outliers, it may not be the best choice to depict speciated composition. Therefore, we have swapped Figure S2 (that is, the second figure in the Supplementary Materials document) with Figure 7, now showing median, rather than maximum conditions in the main paper. We believe Figure S2 provides a better illustration of typical puff magnitude while still conveying important information about the speciated breakdown of puff contents. Readers who are interested in understanding the speciated breakdown of outliers may now find this information in the Supplementary Materials document. In addition, we have added a sentence to the caption of Figure 7 (now moved into Supplementary Materials) noting that the puffs depicted in each bar are outliers.

We have changed the wording that you highlighted (p. 1113 para 2), to be more qualified and to explicitly mention the need for in-plume measurements to refine and increase the confidence of estimates produced via subgrid-scale modeling. Finally, we added the following new material to Section 6 to emphasize the need to evaluate sub-grid scale processes using in-plume data from field observations: "There were no plume-scale measurements available in the Atlanta region for evaluating the plume processes in AMSTERDAM. Future studies should focus on applying and evaluating AMSTERDAM with in-plume data that may be available from previous or future field campaigns. While this study emphasizes the modeling of concentrations within a plume due to aircraft emissions, we caution that this capability has not been fully evaluated against measurements. Evaluation against measurements from aircraft-related plumes will provide additional confidence in sub-grid scale results presented here."

The literature review section touches on many topics tangential to this study but does not mention the sub-grid-scale capabilities available in CAMx or the development of an APT version of CMAQv5. Since this study is based on the AMSTERDAM model, the aerosol and gas-phase chemistry routines (AERO3 and CB-IV, respectively) used are somewhat outdated compared to the more recent treatments available in CMAQv5 and CAMx (e.g., AERO6 and CB6). In the conclusion section, where the authors suggest future work on the AMSTERDAM model, perhaps they should indicate that future work should be based on the CMAQv5-APT or CAMx models that host the most recent chemical mechanisms.

Note that AMSTERDAM is an APT version of CMAQ, and was the only (and latest) version available at the time of this work. However, we have now mentioned the upcoming CMAQv5-APT in Section 2.1 with the following new text: "An updated version of the APT module has been implemented in the latest release of CMAQ v5.0 (with several updates to the overall modeling system, including an enhanced aerosol module AERO6), and it is expected to be publicly available later this year."

We have also added the following paragraph to the portion of the Conclusion (Section 6) that discusses this issue: "Since the AMSTERDAM model was created, enhanced aerosol and gas phase chemistry routines have been developed for CMAQ. AMSTERDAM uses aerosol treatment AERO-3 and chemical mechanism Carbon Bond IV (CB-IV), while the latest versions are AERO6 and CB05 in CMAQv5. Additionally, the Regional Atmospheric Chemical Mechanism version 2 (RACM2) has been recently implemented in CMAQ v5 (Sarwar et al., 2013). Future researchers may wish to incorporate these routines into AMSTERDAM, or use a different subgrid-scale process in a model that natively uses these routines."

p. 1092, line 3: As discussed above, attention should be focuses on the PiG approach rather than the AMSTERDAM model specifically because AMSTERDAM does not have the latest chemistry routines.

We have changed this line to refer to a plume-in-grid process rather than AMSTERDAM specifically.

p. 1095 line 1: The Foley et al. (2010) reference for CMAQ is not appropriate as the version of CMAQ used here pre-dates CMAQv4.7 described by Foley et al.

You are correct: the version of CMAQ that was used in the construction of AMSTERDAM was v4.6, not v4.7. We have removed the Foley et al. (2010) reference. Also, for clarity, we have added the following sentence to the start of Section 3 ("Modeling Approach") : "This configuration is based on CMAQ v4.6, uses the Carbon Bond IV chemical mechanism for gas-phase chemistry, and uses the AERO3 module for aerosol treatment."

p. 1095, line 11: The artificial dilution of emissions in grid models could also lead to over-predictions in the case where oxidants from the background environment are artificially mixed into the plume and, e.g., enhance ozone formation.

We have changed the wording in Section 2.1 to indicate that the artificial dilution can result in "over- or under-prediction of pollutant concentrations" rather than "over-prediction of pollutant concentrations."

p. 1096, lines 9-10: The authors state that "when puffs are sufficiently large or dilute, it is no longer worthwhile to track them separately from the surrounding air." Please indicate the physical and chemical criteria used to make this determination in AMSTERDAM.

We have corrected the manuscript to indicate that only a physical criterion is used in AMSTERDAM. In Section 2.1, we now say:

"When puffs are sufficiently large, AMSTERDAM no longer tracks them separately from the surrounding air. AMSTERDAM makes this determination by using one of two physical criteria. The primary criterion is met if the horizontal size of the puff accounting for spread in both the x and y directions is equal to the grid cell size. The secondary criterion is met when the horizontal size in any one direction is 16 times the grid cell size in the same direction. For most applications, the dumping occurs based upon the first criterion, and the second criterion was implemented to address elongated puffs that would otherwise get transported to longer distances leading to instability in the model. When either criterion is met, the puffs' contents are added to the grid cells where the puffs are located (based upon the centroid of the puffs), and the puffs themselves are removed from the model."

p. 1096, lines 25-30. Is the version of CMAQ-APT used here publicly available?

Yes, the AMSTERDAM model (including the CMAQ-APT configuration) is publicly available on the CMAS Center's website at http://www.cmascenter.org. We obtained it from the model author, Prakash Karamchandani (who is thanked in the Acknowledgements section of the paper) in advance of the public release. We have updated the Acknowledgements to say: "We also thank Prakash Karamchandani and the Electric Power Research Institute (EPRI)'s Eladio Knipping and Naresh Kumar for making AMSTERDAM available to us before its public release." During the course of this study, we identified and reported eleven bugs in AMSTERDAM. We have been informed that all of these bugs have now been fixed. An updated version of the APT module has been implemented in CMAQ v5 and is expected to be publicly released later this year.

p. 1101, lines 23-25: If the authors have 2005-based information to drive the emissions for the airport why choose to model 2002?

Except for information on the total number of flights disaggregated by engine type, all of our input data represent the year 2002. This included the temporal distribution of flights throughout the modeling period (i.e. ratios of activity between different hours of the day, days of the week, and months of the year), background emissions, meteorology, initial conditions, boundary conditions, etc. It would not have been possible to easily obtain all of these inputs for the year 2005.

In order to help readers better understand the impact of this issue, we have added the following sentence (and reference) to Section 4.1: "In 2005, Hartsfield-Jackson Airport had 972,248 aircraft movements, an increase of 9.2% over the 889,966 aircraft movements in 2002 (Airports Council International, 2013), so our results may show $PM_{2.5}$ impacts slightly higher than would be expected if 2002 flight data had been available."

p., 1103, lines 20-30: Please identify the 8 TOG components of the emissions input to AMSTERDAM and the amount of mass for each in the emissions table

In the body of the paper, we have added the following line and reference in Section 4.1 listing and explaining the 8 TOG components: "The eight representative VOC species as defined in CB-IV (Gery et al., 1989) are PAR for paraffins, OLE for olefins, TOL for toluene and other monoalkyl aromatics, XYL for xylene and other polyalkyl

aromatics, FORM for formaldehyde, ALD2 for higher aldehydes, ETH for ethene, and ISPD for isoprene products." We also updated Table 1 as suggested to include their masses.

p. 1107, line 16 (Fig. 2): It would be worthwhile to overlay the observed values in Fig. 2.

We have updated Figure 2 to include an overlay of monitors with observed concentrations in the domain.

p. 1108, lines 3-5: Can this explanation of reduced sulfate due to reduced OH be examined by comparing OH concentration in the different modeled cases?

As you suggested, we performed an analysis of OH and sulfate concentrations in different modeled cases. While we found that OH and sulfate both decrease far from the airport, the spatial pattern of the OH declines does not agree with the spatial pattern of sulfate declines. Accordingly, we have removed the language from the paper (in Sections 5.2 and 6) suggesting that this is the explanation for the observed sulfate declines.

p. 1108, lines 18-22: Please explain the reasoning here more clearly. I am not sure what reactants are being referred to, and I am not sure how coagulation of EC would increase the total EC mass.

We have replaced the word "reactants" with "gas-phase precursors to secondary aerosols" in Section 5.3 in order to clarify the type of reactants being discussed here.

We have deleted the sentence regarding coagulation, which would not necessarily be expected to increase aerosol mass (and, in fact, might increase the deposition rate).

p., 1109 lines 1-5. Please clarify the discussion about different temperatures for the puffs compared to the surrounding grid cell. The methods section indicates that standard gridded meteorological inputs are provided to AMSTERDAM. How is the model obtaining or estimating higher temperatures for the puff compared to surrounding air?

Puffs in AMSTERDAM have many physical properties, including temperature. If a puff's temperature is higher than the temperature of the surrounding air, then the puff may rise due to buoyancy. The temperature of the surrounding air, not the temperature of the puff, is derived from meteorological data.

The temperature of puffs at the time of emission is simply an input parameter that is set by the model user. In the case of our study, we used temperatures ranging from 648K for taxi emissions up to 843K for takeoff activities to represent the aircraft exhaust, and we now provide these values in Section 5.3.

p. 1109, lines 8-9: Looks like the increase in nitrate is dampened by the increase in sulfate that acidifies the particles.

These lines address the effects of the PinG process. Both nitrate and sulfate are formed with or without this process; our findings show that implementing the process does more to boost sulfate formation than nitrate formation. It is possible that the concentrating effect of using a PinG process causes the inorganic chemistry of the sulfate-nitrate-ammonium aerosols to favor formation of ammonium sulfate first.

We have added the following lines to Section 5.3 of the manuscript: "The increase in sulfate and nitrate concentrations may be due to the PinG process transporting inorganic gas-phase precursors away from urban areas and to areas where more ammonia is available to neutralize the sulfate and nitrate. Additionally, since this is a summer simulation, high temperatures favor the formation of ammonium sulfate over ammonium nitrate since the latter is semi-volatile."

p. 1109. lines 10-22: It is hard to understand exactly what is going on here based on the description. It would help clarify the discussion if the authors could investigate the differences in deposition for the species to determine if that explanation is valid. If coagulation has a significant impact, then I wonder if the emissions size distributions are accurate and if the size resolution of the AMSTERDAM PM routines is adequate to accurately represent these processes.

We agree that the description you highlight is unclear and have re-written the relevant part of Section 5.3 to improve clarity by discussing sulfate and EC separately, rather than together. We also put greater emphasis on the transport explanation and a reduced emphasis on the deposition explanation for EC concentration changes (and we no longer restrict the transport explanation to the case of increasing concentrations) in Sections 5.3 and 6. We did check deposition estimates between the various simulations for EC and ASO4, and in general they do increase near the airport with PinG. However, there is no clear spatial pattern, and we agree that this may not be the only possible answer.

Regarding AMSTERDAM PM routines and emissions size distributions, we didn't change the default model assumptions as part of this study. However, based upon particle measurements from aircraft exhaust (Kinsey et al., 2010), we think that the size distributions for aircraft emissions need to be adjusted, where more emissions are in the Aitken mode than currently assumed. Accordingly, we have added the following new text in Section 6: "CMAQ-AMSTERDAM currently assigns 99.9% of black carbon (PEC) and primary organic aerosol (POA) emissions (from all sources including aircraft) to the accumulation mode (particles of size between 0.1 and 2.5 μm with geometric mean particle diameter (GMDV) of 300 nm), and 0.1% to the Aitken mode (particles of size < 0.1 μm with GMDV of 30 nm). A series of commercial aircraft engine measurements performed under the Aircraft Particle Emissions eXperiments (APEX) 1-3 showed that the GMDV for aircraft emissions range from 9.4 to 37 nm for various engines under different test conditions (Kinsey et al., 2010). It is clear that the aircraft emissions are closer to what CMAQ assumes as the Aitken mode rather than the accumulation mode. Future work should explore assigning aircraft emissions to the Aitken mode and assess the impacts of this change on predicted aerosol concentrations."

p. 1109, lines 11-12: Since some of the impacts considered in this paper are very small, one wonders if the combination of the PiG model and host model is mass conservative. Has anyone ever examined the mass conservation characteristics of this model?

> AMSTERDAM, which is the combination of CMAQ with a plume-in-grid treatment, does conserve mass between these two steps. Emissions that enter into puffs are subtracted from the emissions going into the underlying grid cells a priori. Deposition processes are part of the CMAQ Chemical Transport Model (CTM) and do not remove mass from puffs, so all of the mass that was added to the puffs is still present when the puffs are merged into the grid. No mass is created or destroyed during the merging process. Once the puffs are merged into the grid, mass can be removed via deposition as normal.

> We are aware of mass conservation tests for sulfur and nitrogen species (the emitted species) in early versions of APT implemented in MAQSIP (prototype model for CMAQ; see Mathur et al., 2005) and CMAQ, and documented in an external peer-review of CMAQ-APT performed by a team of investigators that among others, included one of the co-authors of this manuscript (EPRI, 2003). But based upon discussions with the developers of AMSTERDAM, no recent attempts have been made to characterize or evaluate mass conservation since then.

p. 1111, line 3. Do all pollutants have the same criteria? Does it make sense in this application to dump elemental carbon to the grid because gas-phase chemical conditions have matured?

> This is an excellent point. Yes, all pollutants have the same criteria, and that is based only on the physical size of the puffs. We have clarified the manuscript (in Section 2.1) to state that the dumping of concentrations from puffs to grids occur only based upon physical criteria and not chemical criteria (Also see our earlier response on a related issue). While AMSTERDAM simulates gas-phase chemistry in CB-IV, it was not updated to look at chemical criteria to determine maturity for aerosol chemistry due to the complex set of reactions involved. So, the version of AMSTERDAM that we used is configured not to use any chemical criteria to determine maturity of puffs and relies only on physical criteria, as described in detail earlier, for puff handover.

Figures 3-5. The units in the captions and on the figures do not match.

> This has been fixed.

Figure 8. This plot is very difficult to read. I would recommend reducing the number of curves in the figure to about 3.

> We have updated Figure 8 to include results only at 3 distances – 4km, 36km and 68km – to enhance readability. Also, to be consistent with Figure 7, which now shows daily median (instead of daily maxima concentrations), we present median concentrations in Figure 8 too. The timeseries of daily maximum concentrations is now available in the Supplemental Information, as Figure S3, to go along with Figure S2 depicting the speciated information for $PM_{2.5}$.

References Cited in this Reviewer Response:

Kinsey, J.S., Y. Dong, D. C. Williams, R. Logan (2010). Physical Characterization of the fine particle emissions from commercial aircraft engines during the Aircraft Particle Emissions Experiment (APEX) 1 -3, Atmospheric Environment, 44:2147-2156.

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