

Responses to Reviewers Comments on “Dynamic Adjustment of Climatological Ozone Boundary Conditions for Air-Quality Forecasts” by P. A. Makar et al.

We thank the reviewers for their insightful comments on our work. We hope that our responses below and in the revised manuscript will address their concerns. In the following, the reviewers’ original comments are provided in italics, followed by our response in regular font.

Anonymous Referee #1

(1) One of the findings of this paper is that the best overall performance of ozone in the troposphere was achieved using the no mass consistency correction (but with dynamic tropopause adjustment of course) and the best surface ozone performance with the vertical wind correction. To my understanding, AURAMS utilizes the semi-Lagrangian numerical scheme that conserves mixing ratio for pollutant transport, then applies an additional global mass adjustment to improve mass conservation property of the scheme. Therefore, additional mass consistency adjustment schemes introduced (either OPT2, or OPT4) may not be necessary if original AURAMS transport is truly mixing ratio and mass conserving. Note that OPT3 is an incomplete form of mass consistency correction, so its test is of no use. If the redundant correction is applied, now I am worried if the system still can conserve mixing ratio? Has any test performed to ensure if the different approaches utilized has any merit to be included in the set?

AURAMS carries out advection using the following hierarchy, within a given time step:

- (1) If the vertical advection mass consistency correction is used, this is applied to the velocity fields prior to calling semi-Lagrangian advection.
- (2) 3D backtrajectories are calculated using the wind fields
- (3) If the mass consistency corrections making use of the advection of density or of (density x Jacobian) are required, these fields are advected, and the corresponding correction ratios are calculated.
- (4) 3D semi-Lagrangian advection is carried out
- (5) If the mass consistency corrections making use of advection of density or of (density x Jacobian) are required, these corrections are carried out.
- (6) Global mass conservation is applied.

Note that the global mass conservation is applied as a final step, and the mass consistency corrections happen prior to this step. Thus, if the mass consistency corrections result in exact conservation of mass, then the global mass conservation impact will be zero. The text has been modified to make the order of operations in AURAMS more clear.

Re: “*OPT3 is an incomplete form of mass consistency correction*”

It might be better to state that “OPT3 is an ad hoc empirical method”, in that it was “presented without theoretical explanation” (Byun 1999b). However, it has been used in the past (Chang et al, 1997), so we feel it is worth including here for completeness’ sake. We’ve added the Chang reference and the quote to Byun in the modified manuscript.

Re: “... *different approaches utilized* ...”. The current work in the submitted manuscript is only our latest chapter on this issue. Earlier testing on the vertical velocity calculation methodology (OPT2) was reported in Gong et al. (2003). At the time these earlier tests were carried out, a zero-gradient boundary condition was used, which in later work (Tarasick et al., 2007, Samaali et al., 2009) was found to be inadequate; ozone concentrations in the middle to upper troposphere were being severely underpredicted relative to ozonesondes. When a realistic

climatological ozone profile was used as the model boundary condition, the problem with excessive downward transport of ozone became clear. This in turn prompted the investigation leading to the paper currently under consideration. This sequence of events has been clarified in the literature portion of the revised paper, and the Gong et al. (2003) reference has been added.

(2) It is not obvious why the surface ozone positive bias should improve if the corrected vertical wind has tendency to bring down higher ozone from above. I am very concerned to conclude that OPT2 would be the best in improving the surface ozone prediction while incurring significant positive biases in the free troposphere and upper troposphere. Wouldn't it be just due to a compensating error working in the direction of reducing the biases? Isn't the transport process the most important factor here as the PM2.5 improves most with the OPT4 as shown in Table 3?

Re: “*why the surface ozone should improve*”. We suspect that this is a compensating error: the very large concentrations in the upper troposphere due to the excessive downward transport of ozone also result in interpolative “undershooting” of the ozone closer to the surface (recall that we use semi-Lagrangian advection and the erroneous mass transport in the upper part of the model domain results in a steep concentration gradient in the middle troposphere). We have modified the text to discuss this point.

Re: “*very concerned to conclude that OPT2...*”. This was precisely our point, so we have tried to clarify the manuscript in this regard. We are NOT recommending the OPT2 method based on our tests; rather, our intention was to show that this method may get what looks like good results at the surface, but when you look in the column, these “good” results are happening for the wrong reasons. We also wanted to show the dangers of using surface bias alone as the sole criterion for judging model performance.

(3) Usually predicted O3 shows positive bias in lower concentration range. In such a case, the offset in the regression between the simulated and observed can affect most of the statistical values utilized here. Can we really use these incongruent measures to judge if one approach is better than the others? Bottom line is that considering all the input uncertainties in both meteorology and emissions, the best approach (of mass correction) should be chosen from the theoretical basis a priori with the in-depth understanding of the model configurations.

Re: “*usually predicted O3 shows positive bias in the lower concentration range...better than the others*”. Our comparisons suggest that at least some of that positive bias in the lower concentration range may result from the manner in which boundary conditions and the manner in which mass consistency and conservation algorithms are applied. However, we must add, that this is the result of tests for our particular combination of meteorological models and air-quality models, and specifically for ozone predictions. In the Discussion and Conclusions section of the original and revised manuscripts, we say this explicitly, “It is important to note that at least some of these results may depend on the particular combination of meteorological model and CTM grids and grid projections, as well as their respective vertical coordinates and resolutions, the advection algorithm employed, and the height of the model top.”. Our use of bias as a statistic for determining model performance is a common operational one for both air-quality and weather forecasting models. We note the fallacy of using bias alone via our comparisons with vertical profiles of ozone concentrations, where we show that the best (surface) bias set of options (OPT2) is clearly causing unrealistic behaviour at other levels in the model domain. We agree with the reviewer that the theoretical basis for a mass correction is a crucial part of deciding what methodology to use. We also feel that comparisons to observations are an equally essential part of evaluating different approaches. We have

modified our conclusions, and the paper abstract, to discuss these points in more detail, stressing that these results may be unique to our particular setup, while also stressing the importance of comparisons to 3D datasets in the evaluation of different methodologies.

(4) Both GEM (meteorological model) and AURAMS use the scaled terrain following height as the vertical coordinates. Compared to other atmospheric models that use a form of hydrostatic pressure coordinate, a terrain-following vertical coordinate tends to have unwarranted vertical motions in the upper troposphere, which may be responsible for the exaggerated stratosphere-troposphere exchange of pollutants, etc. Also, the correction method OPT2 tends to accumulate divergence errors in the lower atmosphere toward the top of the model. Combination of these two may accentuate the effect of the lateral and top boundary conditions at the downwind of Rockies. Quantification of such mass flux must be made to understand the final effects on ozone simulations.

Re: “GEM ... uses a scaled terrain following height as the vertical coordinate”. GEM actually uses a hybrid sigma-pressure vertical coordinate $\left(\eta = \frac{P - P_T}{P_S - P_T} \right)$, whereas AURAMS does use a modified Gal-Chen terrain-following vertical coordinate $\left(\varsigma = \frac{z - z_{\text{terrain}}}{z_{\text{top}} - z_{\text{terrain}}} z_{\text{top}} \right)$, where z_{terrain} is the local terrain height, and z_{top} is the model top.

Re: “...tends to have unwarranted vertical motions in the upper troposphere, which may be responsible for the exaggerated stratosphere-troposphere exchange of pollutants”. The vertical velocity field is supplied by the meteorological model and should not contain unwarranted vertical motions in the upper troposphere. But if this were the case, in our modelling setup, we would expect the OPT1 simulations, which use no mass-consistency corrections and a global mass conservation correction, to show signs of these unwarranted vertical motions. In contrast, we see the opposite: the erroneously high concentration ozone in the upper part of the model domain is only present when one of the approaches for mass consistency is applied; it does not take place when the original wind fields are used.

Re: “Also, the correction method OPT2 tends to accumulate divergence errors in the lower atmosphere toward the top of the model.”. Yes, this is correct and is a result of the discretization used in the relevant equations; errors will accumulate upwards for the OPT2 methodology. We have modified the text describing the different approaches to accentuate this point. It is worth noting that the OPT3 and OPT4 approaches do not explicitly accumulate errors upwards, yet they also display a similar tendency to exaggerate the downward transport of ozone from above.

Anonymous Referee #2

General Comments:

This paper presents and assesses a number of techniques for interfacing regionalscale air quality models of limited horizontal and vertical spatial extent with climatological profiles of ozone used as boundary conditions for the regional models. These profiles extend into the stratosphere, so the corresponding values of ozone are consequently quite high, and care must be taken in specifying these high values as boundary conditions unless the appropriate strat/trop dynamics

are modeled well. Regarding that point, it would assist the readers to include a figure illustrating the vertical structures/ layering used in the GEMS and AURAMS models, and to the extent that the layer structures differ between the models, how that affects the dynamics and vertical fluxes in the chemical-transport model.

A figure showing the GEM and AURAMS layers relative to sea level has been included in the revised manuscript.

The various interfacing configurations were described well, and the testing results were presented in a clear and insightful manner. Taking into consideration and normalizing for the difference in height between the climatological tropopause and the GEMS modeled tropopause seems prudent as far as interfacing with a climatological ozone profile. However, one wonders whether the various methods tested in some way are simply used to balance the errors induced by the deep upper layers used in AURAMS and any dynamic inconsistencies induced by disparate layer matching between the meteorological and chemical-transport models.

As we noted in the Discussion and Conclusions section of the revised manuscript, the methodologies from the literature tested here were intended to be applicable to different combinations of meteorological and air-quality model coordinate systems. Incompatibilities between air-quality and meteorological model coordinate systems is one of the reasons why one might expect mass consistency problems in the resulting wind fields, and one of the justifications for the application of mass-consistency corrections. Apparently, for our combination of meteorological model, air-quality model, and boundary conditions for ozone, the corrections do not always work (and actually made the ozone results worse).

Was any consideration given to using potential vorticity as a stratospheric ozone tracer to provide estimates of upper level ozone concentrations above the climatological profile limit?

We have examined this possibility in another project using the FLEXPART modelling system, appearing in the same special issue of ACPD (He *et al.*, 2010), already referenced in the original manuscript. Accordingly, we have added a note in the Introduction referencing He *et al* (2010) in the context of potential vorticity as a proxy for ozone. One disadvantage of potential vorticity is that it shows good correlation with ozone concentrations in the stratosphere, but this correlation weakens considerably in the troposphere.

Specific Comments:

page 13654/line 19 – The North American domain analysis covers the summer 2007 period. Certainly the summer is the time for greatest interest in surface ozone concentrations. However, the exchange between strat/top is at its most dynamic in the spring. It would be interesting to see a similar analysis for that time of year, and whether the results differ from those shown here.

We agree with the reviewer; we would expect to see a larger impact in the springtime. However, the simulations required to do these comparisons take approximately 7 weeks of processing time each on our current supercomputer (the jobs are run concurrently to reduce the overall wall-clock time), and our observations were for a measurement intensive in June/July of 2007. We have included a comment to the manuscript that the spring season may be of greater interest in our discussion of possible future directions for the research.

page 13659/8 – Did the higher resolution case studies use the same model configurations and vertical structures as the coarser resolution North American domain study?

We have clarified this point in the text, in the description of the meteorological model. The highest resolution simulations (2.5 km) use a different physics package within the meteorological

model; convective parameterizations are no longer required at that resolution. The vertical resolution was constant for all model horizontal resolutions. The text has been modified to clarify this point.

Figures 4,6 – Plots are too small or too busy to discern any details.

The size of these figures has been increased: Figure 4 (new Figure 5) has been broken up into five images instead of three, and Figures 5 and 6 (new Figures 6 and 7) into two images instead of one.