

Responses to Reviewer Comments

Modeling inter-continental transport of ozone in North America with CAMx for the Air Quality Model Evaluation International Initiative (AQMEII) Phase 3

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The authors thank the reviewer for their helpful comments. Below we respond to each and note our changes to the manuscript.

Anonymous Referee #1 Received and published: 11 April 2017

General comments: The paper analyzes several model approaches for estimating the impact of long-range ozone transport, including use of: brute-force sensitivity tests, chemically-reactive tracers, and inert tracers. The regional CAMx modeling for the U.S. (12km) is based on well-established inputs from the AQMEII Phase 3 effort and sufficient information is presented regarding the model's ability to replicate observations from the simulation period (2010). There are several areas of focus within the manuscript: 1) the impact of boundary conditions on simulated ozone within the U.S., 2) parsing these boundary condition impacts by height, 3) assessing the impacts of 20% reductions in emissions globally and from East Asia, 4) a limited comparison of the boundary conditions in CAMx to another regional model (CMAQ), and 5) comparisons of the various model approaches (e.g., sensitivity vs. reactive tracers) in estimating boundary impacts. The key takeaway from the manuscript is that regional models will be sensitive to biases and errors in boundary conditions, especially in the inter-mountain States in the western U.S. The overall quality of the paper is good and the subject matter is of keen significance to the air quality management community. One general commentary on the manuscript is that there are a number of instances where a finding is made and then several hypotheses are offered for why the finding might be what it is, without any followup analyses to assess the merit of the various hypotheses. Examples include: section 3.1 ("potential causes are ..." deposition, halogen chemistry, mixing), section 3.2 ("other factors must be contributing (not examined here)"), section 3.7 ("factors contributing to these differences may include ..."). Recognizing that no manuscript can be exhaustive, the authors are encouraged to reassess if more analyses are possible in the scope of this work to determine the causes for these modeled features. We especially encourage additional analyses in section 3.7 which, in its current form, raises as many questions as it answers. To the extent, that resources do not permit additional analyses, the authors are encouraged to limit the number of "dangling" hypotheses; either by saving them all for Section 4 as a sort of "next steps" list, or by deleting the sections with conclusions without identified causes.

Response 1: We agree that further investigation into difference between CAMx and CMAQ (and other model) results will be useful and note that US EPA is leading this effort and their paper will be submitted to this same Special Issue. We have added their paper to our reference list (Liu et al.). The point of Section 3.7 is to emphasize the magnitude of bias using the inert tracer approach relative to chemically-reactive tracers, and the fact that this bias is much larger than differences between two inert models using the same inputs. We have added the following text in bold to Section 3.7.

“Differences between the CMAQ and CAMx inert tracer impacts are smaller than the differences between inert and reactive tracers in CAMx which exceed 10 ppb (Figure 5), but they are notable, in a range of 4-8 ppb in summer and 2-6 ppb in spring with CAMx being higher. Factors contributing to these differences may include fewer vertical layers in CAMx (26 compared to 35 in CMAQ), which may cause

more numerical diffusion of UTLS O₃ to ground level (Emery et al. (2012), omission of wet scavenging for the CAMx inert tracers, treatment of deep convective transport in CMAQ, or differences in model treatments of O₃ dry deposition. **Liu et al. (this issue) performed multi-model process comparisons with four AQMEII models and draw similar conclusions regarding factors that can contribute to differences in tracer impacts."**

Reference: Liu, P., Hogrefe, C., Bieser, J., Im, U., Mathur, R., Nopmongcol., U., Roselle, S., Spero, T.: Multi-Model Comparison of Lateral Boundary Contributions to Surface Ozone Over the United States, in preparation.

We think including our hypotheses within their respective sections (the way the text is currently written) is useful. As suggested by the reviewer, we have also summarized these hypotheses in Section 4 as suggestions for future work.

Specific comments: The two most likely "policy-relevant" conclusions to be cited from this manuscript are that 1) boundary conditions impacts on the Denver area average 57 ppb on the days with the highest MDA8 O₃, and 2) that a 20% reduction in emissions from East Asia will have < 1 ppb impact on surface O₃ in the U.S. Particularly for that first conclusion, the manuscript would be improved if more detail was provided about the robustness of the conclusion. For instance, it is not clear to this reader whether the city-specific analyses are based on a single site or an aggregate of sites within an area. Additionally, given the note in the paper about the high modeled bias on the the H4MDA8 O₃ day (observed O₃ = 50 ppb while BC impacts alone > 70 ppb), it would be helpful if the model bias/error values for the top 30 subset of days were also included in table 3 or elsewhere. Given the paper's conclusion that biases/errors in the boundary conditions will affect regional concentrations, it is imperative to understand what the biases/errors are on these Top 30 days before too much weight is assigned to the 57 ppb conclusion in Denver (i.e., if there's a positive bias in O₃ over those 30 days, then that specific estimate of the role of BC may also be overestimated). The Denver area is notoriously hard to model. Are the authors comfortable that the 12km CAMx modeling is properly capturing the meteorology ("Denver cyclone") and other daily-varying conditions that lead to a complex mix of local/regional/natural/international contributions in this area? It's hard to discern that from seasonal-average tables of bias and error.

Response 2: We thank the reviewer for this suggestion. We have added quantile-quantile (Q-Q) plots to the SI (Figure S8; Denver example is shown in Figure R1). Q-Q plots compare the independently sorted (time-unpaired) values of observed and modeled concentrations and are useful for evaluating whether the model can reproduce the distribution of observed values (perfect model performance would show all data points along a 1:1 line). Our Q-Q plots suggest good model distributions (e.g., data points nearly match the 1:1 line) for Los Angeles, Sacramento, Phoenix, Denver, Dallas, Houston, Pittsburgh, and Philadelphia. In Denver, the model captures the ozone distribution below 65 ppb well and slightly under predicts ozone above 65 ppb, but it over predicts the highest MDA8 ozone. We have added the following statement (text in bold) to the second paragraph of Section 3.1.

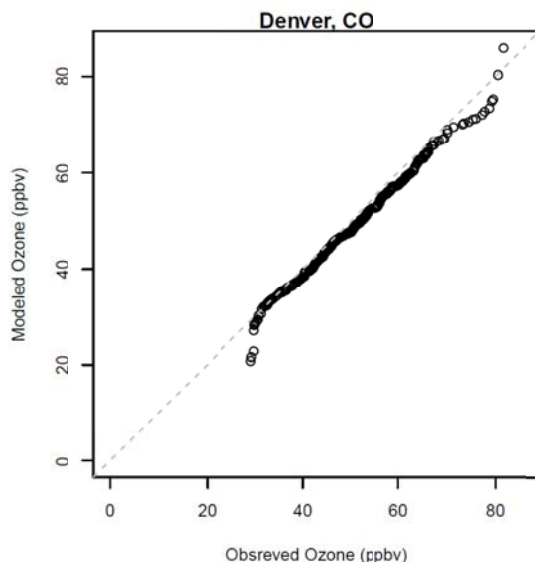


Figure R1. Example of Q-Q plot for Denver

“We additionally provide quantile-quantile (Q-Q) plots (Figure S8) which compare independently sorted (time-unpaired; space-paired) observed and modeled O₃ for each city. The Q-Q plots suggest good model distributions (e.g., data pairs near the 1:1 line) for Los Angeles, Sacramento, Phoenix, Denver, Dallas, Houston, Pittsburgh, and Philadelphia.”

The city analyses are based on a single site that shows the highest H4MDA8 within the metropolitan statistical area as described at the end of Section 2.2 and in Figure S1. The selected AQS site ID is provided in Table S3.

Per the finding that there is a near-linear relationship between the O₃ changes in the boundary conditions and the surface O₃ changes in the western U.S., might there be a more direct way to visualize this finding than the 16-panel plots? Seems like scatterplots of delta O₃ vs. delta tracer would show this conclusion more directly (by region, if needed). Alternatively, perhaps spatial maps of percent O₃ or tracer changes (as opposed to absolute change) would make the point more directly.

Response 3: We added two scatter plots to Figure 7 (See Figure R2 below) to help explain the information offered in this figure. The two scatter plots show the relationship of delta O₃ vs. delta tracer for Denver in spring and summer. The scatter plots also illustrate the regression analyses that we performed to develop Figure 7. Columns 3 and 4 in Figure 7 show the regression parameters for each surface grid cell and match the scatter plots for Denver. We have revised our text (shown in bold) in the second paragraph of Section 3.6 to better describe this plot.

“We examine more closely the relationship between changes in O₃ and reactive tracers in the EAS scenario. In each surface grid cell, we regress hourly O₃ changes against reactive tracer concentration changes (summed over boundary height ranges) to compute slope and r **as demonstrated in the two scatter plots for Denver in spring and summer**. Slope and r values of 1 indicate that the O₃ changes are explained entirely by the changes in O₃ BC reactive tracers. **The delta total O₃ and delta tracer O₃ relationship is near-linear at Denver with a slope of 0.87 and r of 0.9. The 16 panels in Figure 7 show the regression parameters for each grid surface cell and match the scatter plots for Denver.** The slope and r (Figure 7, 3rd and 4th columns) values have similar spatial patterns in all seasons. In winter and fall the slope values are near 1 with r of 0.8 to 1 across the US **suggesting strong influence of O₃ transport from Asia during these seasons**. In spring, strong correlation (r = 0.8 to 1) is seen in the Western US but areas in the Eastern US have a slope lower than 0.2 and r lower than 0.4 indicating that the O₃ BC tracers

can explain only a fraction of the total O₃ change. The lowest correlation ($r < 0.2$) is in the summertime over the Southeastern US in a region where the EAS scenario produces almost no change in surface O₃ indicating that transport from Asia becomes unimportant. High correlation in the Western US in all seasons emphasizes the influence of O₃ transport from Asia in this region.”

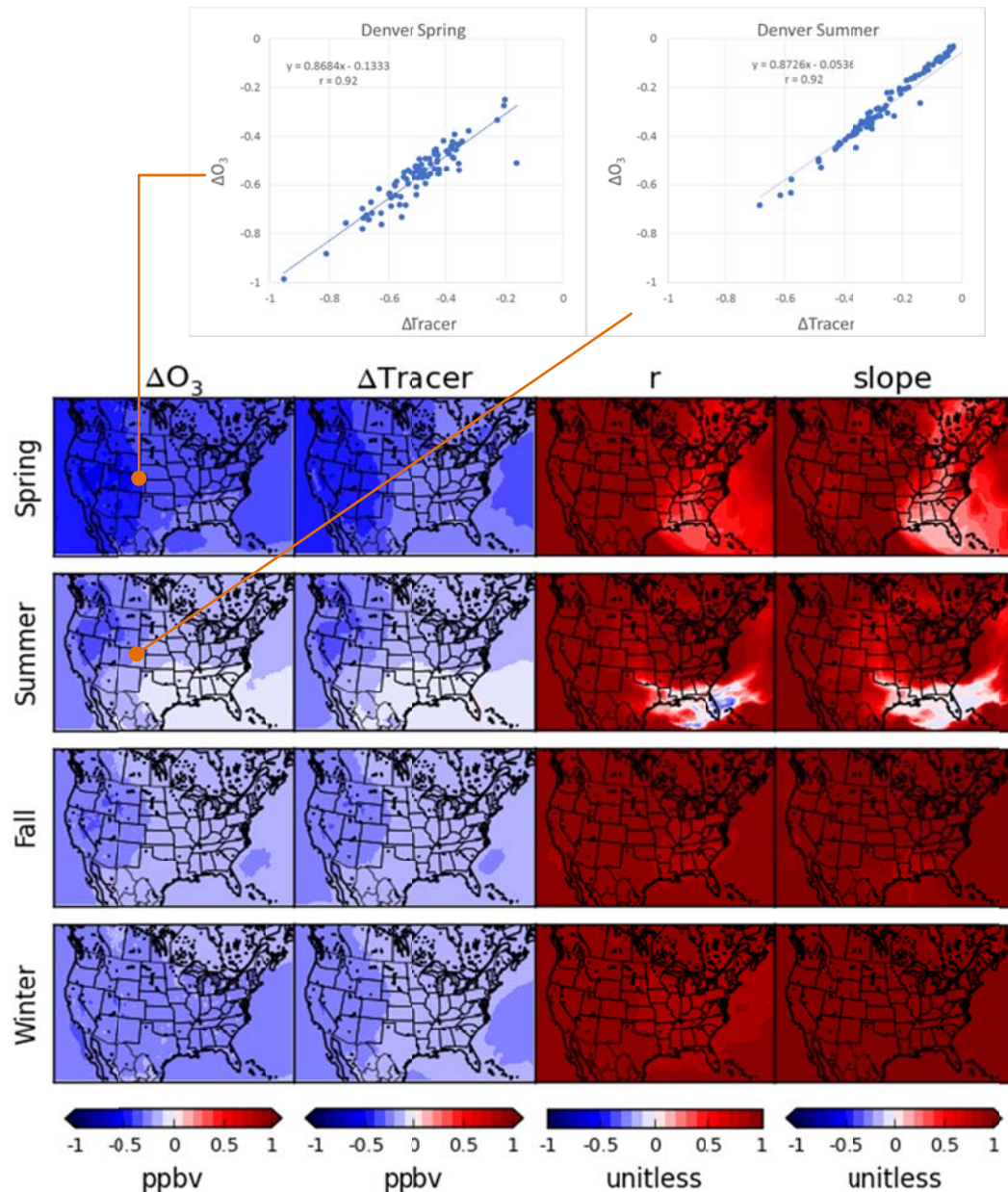


Figure R2: Scatter plots on the top show delta daily average O₃ (y-axis) and reactive tracer BC O₃ contribution (x-axis) for Denver in spring and summer for 20% reduction in East Asia emissions (EAS scenario). The 16 panels summarize the same information showing seasonal delta total O₃ (left column) and reactive tracer BC O₃ contribution (column 2) for each grid surface. The correlation (r) and slope of a linear regression of column 2 against column are shown in columns 3 and 4, respectively.

Per Figure 4, can the plot be modified to show the count of data points in each box/whisker. If there are some boxes with less than some small number of data, perhaps those should be combined into a larger range w/ more statistical robustness.

Response 4: We have included the count of predicted data points in Figure 4 as suggested by the reviewer. We prefer not to group data into larger ranges because it creates uneven spacing and loses detail. Additional modification to Figure 4 is described in our Response#5 below.

Would it be possible for the authors to comment on an additional possible conclusion from Denver/Fig 4? It appears to me that the model is overestimating the BC \rightarrow total O₃ slope in this area (in Phoenix as well). The BC/total slope in the model appears to be close to 1 (i.e., what distinguishes high days from low days in Denver is BC contributions), whereas the observations suggest something much flatter (i.e., what distinguishes high days from low days in Denver is something other than BC). This seems like a potentially important finding. Once the model exceeds 50 ppb, the BC terms are large (and appear to be overdone).

Response 5: We agree that the model tends to overestimate BC on the highest days and this is the reason we look at multiple ozone metrics including summer average and Top 30 days. The model is not perfect, but it captures the ozone distribution at Denver quite well based on the Q-Q plot we added to the SI (Figure S8 in the SI; Figure R3 in this document). The conclusions we can draw from Figure 4 are limited because BC contributions cannot be derived from measurements. We have decided that including observations in Figure 4 is confusing and taking away the key point we want to make (i.e., different pattern of how total O₃ changes as the modelled BC contributions increase seen in the west and the east). So we removed observations (yellow bars) from this plot and revised the relevant text in Section 3.3 as shown in bold below.

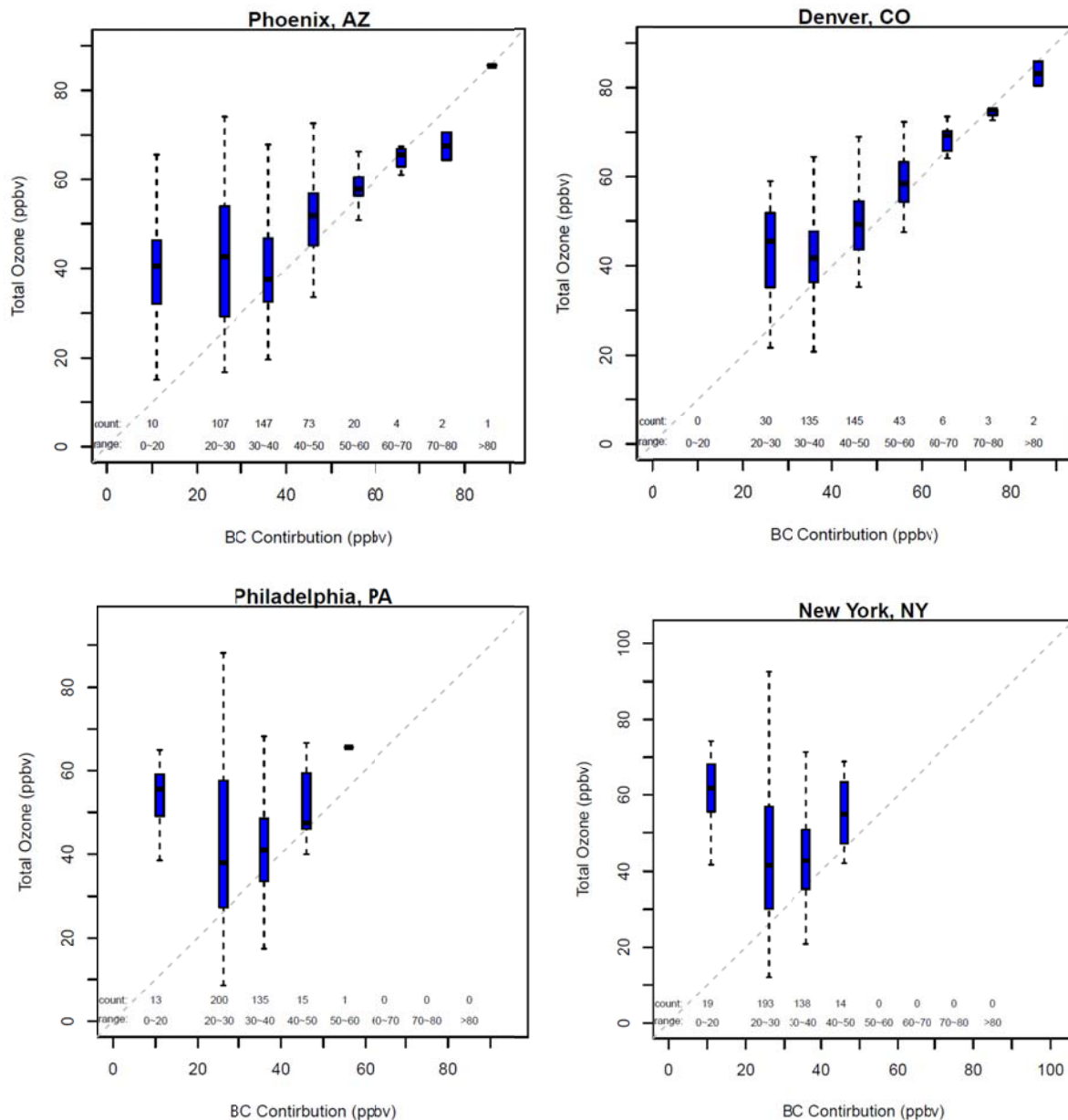


Figure R3: Box-and-whisker plots comparing modeled total MDA8 O₃ to modeled BC contribution in 10 ppb ranges paired in time and space.

“We further investigated how total O₃ changes as the modelled BC contributions increase (in 10 ppb increments) as shown in Figure 4 for several cities. The relationships vary between cities and the model captures this variation with the Western cities (Denver and Phoenix) showing different patterns than Eastern cities (Philadelphia and Atlanta). For Denver and Phoenix in the Intermountain West, total O₃ increases with BC contribution and approaches the 1:1 line at higher BC contribution revealing small groups of days when MDA8 O₃ exceeded 60 ppb (**11 days for Denver and 7 days for Phoenix**) and the modelling indicates that BCs accounted for almost all of this O₃. **In other words, BC contributions alone distinguish high O₃ days from low days.** These groups of **high BC-contributed** days are important because local emission reductions, or even US-wide emission reductions, would be ineffective at reducing O₃. However, there are other days in Denver when total MDA8 O₃ exceeded 60 ppb with modelled BC contribution below 30 ppb (see first and second bars in Figure 4) on which reducing local or US emissions would lower O₃. Air quality managers need methods to identify dates when emission reductions would be ineffective so that those dates can be excluded from emission strategy development. **Nonetheless, these results should be interpreted with consideration given to model performance. As shown in the Q-Q plot for Denver (Figure S8), the model can capture the O₃ distribution quite well, although it underestimates MDA8 O₃ over 65 ppb and overestimates MDA8 O₃ over 80 ppb. For this reason, we encourage making use of multi-day metrics (such as Top 30) rather than a single-day metric (e.g., H4MDA8) “**