Response to Reviewers
An Updated Modeling Framework to Simulate Los Angeles Air Quality. Part 1: Model Development, Evaluation, and Source Apportionment

Review 1:

In this manuscript the authors present a comprehensive exploration of air quality in the LA basin, a complex region influenced by unique atmospheric dynamics and diverse emission sources. As a prologue to a planned future submission exploring consequences of the COVID-19 pandemic lockdowns in more detail, the authors here set out to model regional air quality by applying updated and optimized emissions inventories to the chemical transport model CMAQ, and to evaluate the model against a suite of observations including typical gas- and particle-phase pollutants and their precursors.

On the whole I find this to be an important and well-composed manuscript, providing a compilation of recently identified emission types and associated chemistry into valuable new model/measurement comparisons. The manuscript text is clear and well-composed, as are key figures. However, I do have just a few concerns, mostly related to the treatment of modeled dynamics and its consequences, which I'd like to see addressed before I recommend publication.

Thank you for your thoughtful response and helpful comments. Each question is addressed individually below.

- My primary concern is related to WRF performance in reproducing dynamics and transport. As noted by the authors, wind speed and direction are both crucial components in overall model performance due to their role in determining the transport of pollutants and precursors. The failure of the nested WRF model to adequately represent these features is therefore a pretty big issue in my mind: it's hard to tell how much of the subsequent analyses of chemical composition are impacted by problems in dynamics, casting many of the subsequent conclusions in doubt. I understand that this is a very thorny modeling problem, but I would very much like to see the authors attempt to address this problem in some form, at the very least to try and better understand when and where their WRF simulations are failing, and to quantify the sensitivity of their final results to transport issues.

  The reviewer points out that wind speed is inaccurately predicted, as demonstrated by the large normalized mean bias (NMB) values in Figure 5 in the submitted publication. NMB for all 3 domains is presented in Figure R1. WRF tends to overpredict wind speed at most locations in all 3 modeling domains. This suggests that the model error lies with the input reanalysis data, and less with the model configuration. This further suggests that to improve model simulations, new reanalysis data should be used or observational nudging should be engaged when running WRF. However, using new reanalysis data may introduce error to other meteorological fields, whereas temperature is well-predicted by this model setup. Please see the response to the third point below, where we re-ran CMAQ to test the sensitivity of the model to the WRF-predicted wind speed. We concluded that a reduction of wind field error cannot improve the modeled O_3 and $PM_{2.5}$.

  Figure R1 was included in the manuscript as Figure S10, and the conclusions made here have been integrated into the main text in Section 3.1.1:

"Wind speed and direction tend not to be predicted well, with high bias and high scatter, but the error is highly variable between sites (Figure 5). Wind speed and direction error will potentially affect the transport between grid cells, and their impact on modeled pollutant concentrations is investigated in Section 3.2. To understand the source of wind speed error, the NMB was quantified in all 3 modeling domains (Figure S10). Wind speed did not improve appreciably as the model resolution increased, and the spatial distribution of error remained consistent. This suggests that the model error lies with the input reanalysis data, and less with the model configuration. This further suggests that to improve model simulations, new reanalysis data should be used or observational nudging should be engaged when running WRF. However, using new reanalysis data may introduce error to other meteorological fields, whereas temperature is well-predicted by this model setup."

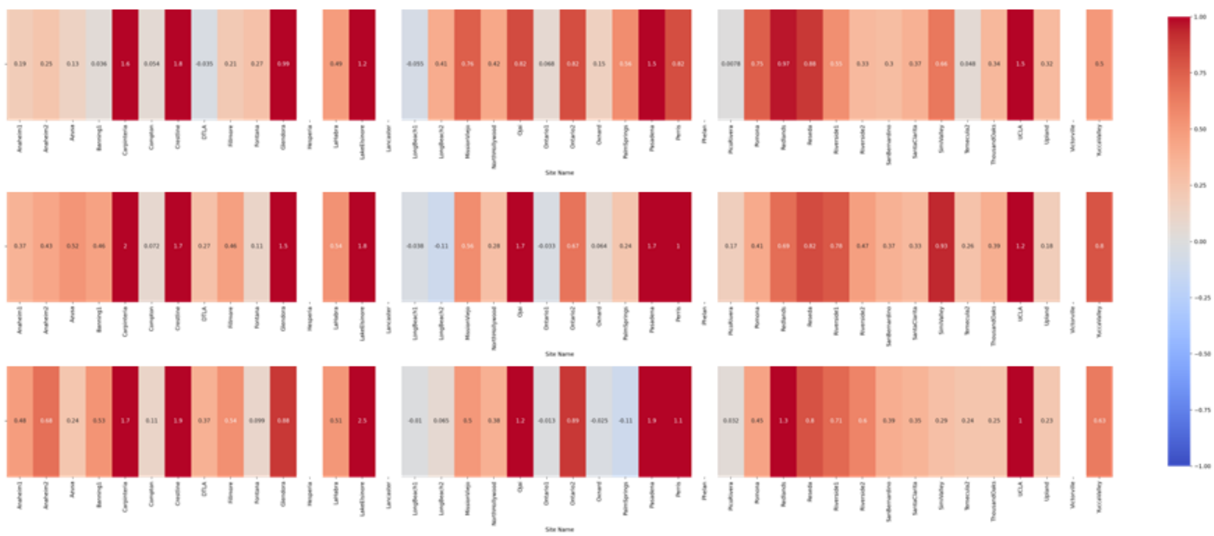

Figure R1: Normalized mean bias (NMB) of WRF wind speed in the 3 nested WRF domains: top) d01, middle) d02, bottom) d03.

- On a related note, I was curious about the chosen WRF nesting scheme used here, which employs a 16:4:1 km grid size scheme. My understanding is that WRF best practice is to use an odd ratio less than 7 (typically 3:1 or 5:1) for spatial dimensions of nested grids to minimize interpolation errors. I'd be very curious to see whether a shift to 15:3:1 km grid sizes might help to improve WRF output here.
  As stated in the above response, the model meteorological errors are very likely caused by the input reanalysis data. This suggests that the downscaling of that data, in any form, would carry through the error to the inner domains. We do not predict that changing the grid ratio would drastically improve the wind speed predictions.

- Since the intended focus of this paper is not on WRF model performance itself, I also wonder whether it would be better to use meteorological data assimilation techniques (or even established high-resolution meteorology data products such as HRRR) to further improve modeled wind speed and direction, providing a clearer focus on emissions and chemistry.

Please see the response to the question below. As those results demonstrate, wind speed does not cause noticeable bias in the error of species such as ozone or $PM_{2.5}$. Hence, we expect that improving wind speed predictions using a tool such as HRRR will not have a large impact on predicted pollutant levels.

- Regardless of how well these (or other) techniques might improve model performance, their use would also allow for a kind of sensitivity study to assess the impact of transport on modeled chemistry. It would be helpful and informative to assess CMAQ predictions under alternative (hopefully improved!) meteorological methods such as those described above, and to compare that performance against predictions made using original meteorology, thereby estimating sensitivities.

As suggested by the reviewer, we have now conducted additional sensitivity study to assess the impact of transport on modeled chemistry. The WRF wind speed was reduced by 25% (i.e., scaled by a factor of 0.75) in an effort to correct for some of the bias. A reduction of 25% was chosen to represent the correction required to bring modeled wind speed into the range of observed wind speed, as represented by the values in Table S1 from the submitted manuscript SI. CMAQ was re-run using the new meteorological files to assess the impact of improved wind speed on pollutant predictions. The results are presented below in Figures R2 and R3. The values in Figure R2 can be compared to the values in Figure 5 in the main text. Wind speed improved appreciably in response to the 25% reduction in their values throughout the domain. In spite of improved wind speed, modeled $PM_{2.5}$ only exhibits small improvement, while modeled O_3 did not respond much. This suggests that wind speed does not have a large effect on modeled pollutant concentrations, and bias in those concentrations is more likely caused by errors in modeled chemistry and/or emissions.

This paragraph has been copied into the main text in Section 3.2, and Figures R2-R3 have become Figures 7 and 8 in the main text.

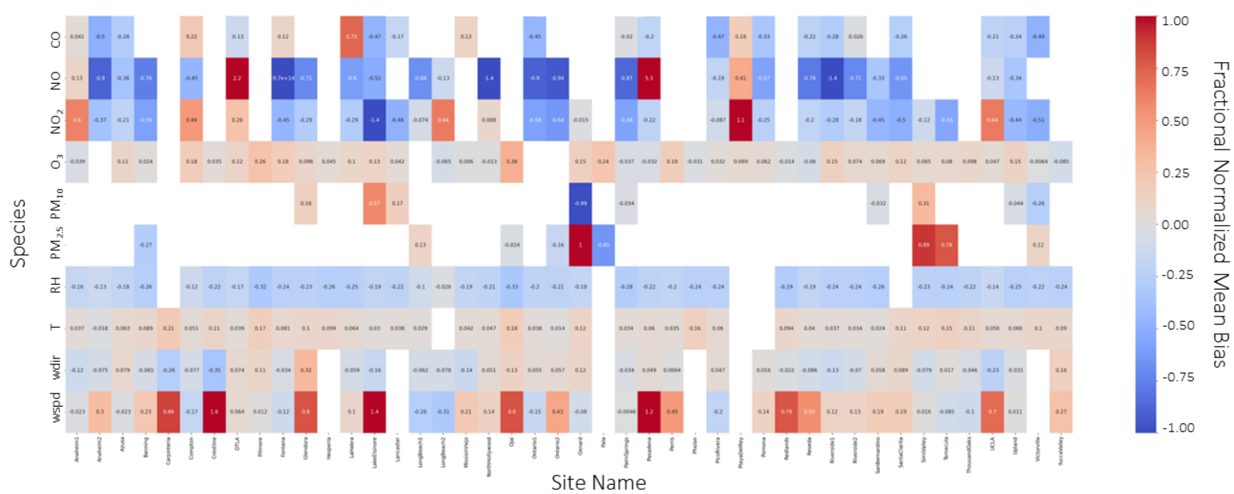

Figure R2: Fractional NMB of pollutants (rows) at all EPA AQS sites (columns) in the LA domain using daily-average values April 1-30, 2020. Empty boxes represent sites without measurements of the given pollutant.

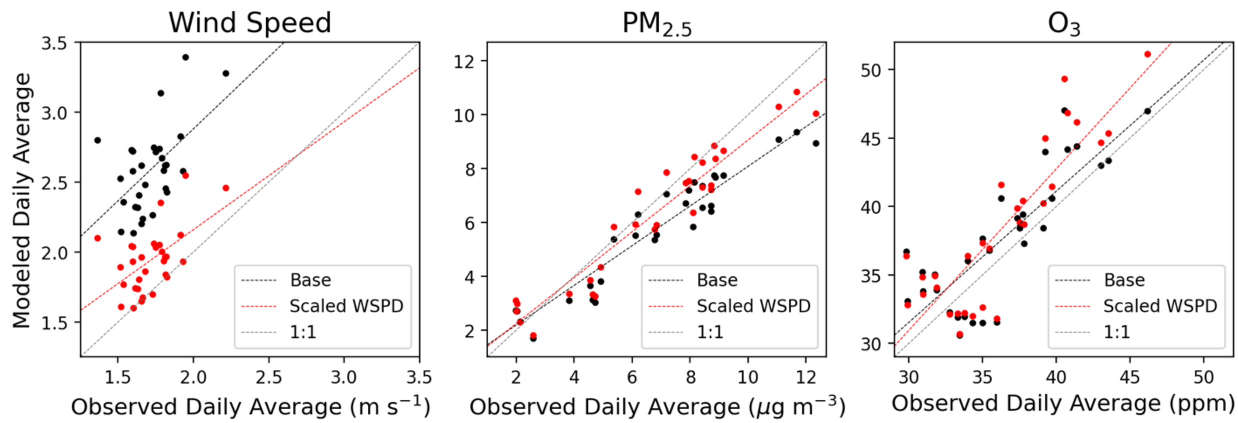

Figure R3: Daily-averaged modeled versus observed values of (left) wind speed, (middle) O_3, and (right) $PM_{2.5}$. Black markers and lines represent data from the "base case" wind speed simulations. Red markers and lines represent data from the scaled (i.e. scaled by 0.75) wind speed simulations. Gray line represents the 1:1 modeled:observed line.

- Why were lightning NO_x and windblown dust sources omitted? While they may not dominate in this urban region, they still seem like sources better included than excluded. (If the omission was due to a lack of trust in modeled wind speeds and convection, this seems like all the more reason to improve these elements.)
  Dust makes up a small fraction of total PM loading. Hayes et al. (2013) showed that in Pasadena, dust makes up only 1.6% of total PM_1 by mass. Natural emissions are the lowest source of PM emissions (CARB, 2023), so windblown dust is a minor contributor to total PM. However, it is possible that muting the dust scheme could cause underestimations of $PM_{2.5}$ and PM_{10}. Previous work suggests that crustal elements, i.e. dust elements, do not have a large impact on modeled ammonium and nitrate concentrations (Ensberg et al., 2013), so omitting these emissions should not have a large impact on other inorganic aerosol or gas-phase species. Previous work (e.g. Choi et al., 2009) has shown that lightning NO_x is nearly negligible over Southern California. Lightning NO_x and windblown dust sources were also omitted to be consistent with prior works investigating the Los Angeles Basin (e.g., Murphy et al., 2017; Qin et al., 2021). This paragraph has been copied into Section 2.1.3.

- This is a more subjective and minor request, but I also would like to suggest that the authors consider an alternative and consistent color scheme for the panel maps of Fig. 6. Rainbow schemes such as this one produce artificial bands across specific color ranges, making it harder to consistently evaluate differences in the maps across concentration thresholds. It also looks like some of the maps use discretized color bins while others are continuous. Unless there is a specific reason for this, please make this a consistent decision one way or the other.
  The color scheme has been updated in Figure 6. All figures now use discretized color bins.

Aside from these concerns I am confident that this will be an important literature contribution deserving of publication. My hope is that improved model dynamics will allow for a stronger profile of emissions and chemistry, along with the potential to quantify their associated sensitivities.

Thank you for your review!

References

CARB. (2023). *Criteria Pollutant Emission Inventory Data | California Air Resources Board.* https://ww2.arb.ca.gov/criteria-pollutant-emission-inventory-data

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Review 2:

This work describes a model framework for investigating air quality and sources of pollution in the Los Angeles region. As a region with complex emissions, understanding the sources of air pollution is critical to improving air quality in the area. The model framework and validation against observations is exceptionally well-described and the work fits well within the scope of ACP. While I think the article could essentially be published as is, additional discussion on the limitations of this model framework based on the analysis of model error would provide more context on how to interpret future results. Therefore I would recommend publication and would encourage the authors to consider addressing the following things. I look forward to part 2!

Thank you for your thoughtful response and helpful comments. Each question is addressed individually below.

1. While the evaluation of the model against available measurements is quite thorough and well-described in the text, there is little discussion of how this evaluation impacts the interpretation of the results as well as the strengths and weaknesses of the model. A comprehensive assessment of how all the biases in specific pollutants could affect results would be largely speculative and thus is unnecessary, but I believe a brief discussion on how the model's performance against measurements could affect big picture results would be appropriate. Below I provide a few questions that struck me as meaningful to address, however they need not all be.

- Does the good representation of POA, but poor representation of SOA mean that generally, this model will predict OA better near sources and diminish in its effectiveness further away? Yes, this is true. Unfortunately, there are not sufficient observations of OA composition to better characterize this phenomenon. This point has been addressed in the first paragraph of Section 3.3:

  > "The accurate representation of POA and poorer representation of SOA suggests that OA is better represented near source regions and diminishes in its effectiveness with distance from sources."

- Could the poor NOx prediction impact the conclusions surrounding the NOx vs VOC-limited ozone regimes?
  The underestimation of NO_x should not affect the conclusions surrounding the inner basin, NO_x-saturated regimes. In that region, a decrease in NO_x results in an increase in O_3. If the NO_x were better-predicted (i.e. higher), this would push the regime into an even more NO_x-saturated regime, and the conclusion would be the same.
  The impact of better-predicted NO_x in the outer regions is less clear. As our results suggest, those regions fall near the NO_x-VOC-O_3 ridgeline (i.e., NO_x-insensitive regime), but we do not know where along that ridgeline. An increase in modeled NO_x may push the regime to a more NO_x-saturated regime, or it could simply move the scenario along the ridgeline and have little impact on model results. This will be important to consider in future work.

- Because of the large biases in certain species does this model's strength lie in predictions of relative changes in species (as the results shown in this work are) rather than predicting absolute values or are there certain species the author's feel confident could be predicted?

Yes, the model likely predicts relative changes and perturbations better than absolute concentrations. This suggests that source apportionment and other studies that compare model scenarios are important.

- Do the large errors in wind speed and direction indicate a systematic bias of air from different areas into the domain region (e.g. higher sea spray aerosol from the ocean vs more agricultural or road sources from in-land) or is the spread too large?
  Wind speed shows a systematic high bias but there is no systematic bias in $PM_{2.5}$ based on location. Unfortunately, there are not sufficient aerosol composition measurements to allow us to investigate whether wind speed bias correlates with bias in specific aerosol species. When wind speed was better-predicted, the bias in $PM_{2.5}$ (and other pollutants) did not improve appreciably (Figure R2), further suggesting that wind speed error does not produce a systematic bias in pollutant concentrations throughout the domain.

2. Are wildfire emissions included? If they are included in one of the emission inventories then that should be made clear.
Wildfire emissions are not included. We chose to model a time period with limited wildfire activity so that the effects of wildfires were insignificant. We have made this more clear in Section 2.1.3:

> "Wildfire emissions were not included as this time period experienced limited wildfire activity."

3. Why were lightning NOx and dust sources not included? Are these just not large emission sources in the area?
Dust makes up a small fraction of total PM loading. Hayes et al. (2013) showed that in Pasadena, dust makes up only 1.6% of total PM_1 by mass. Natural emissions are the lowest source of PM emissions (CARB, 2023), so windblown dust is a minor contributor to total PM. However, it is possible that muting the dust scheme could cause underestimations of $PM_{2.5}$ and PM_{10}. Previous work suggests that crustal elements, i.e. dust elements, do not have a large impact on modeled ammonium and nitrate concentrations (Ensberg et al., 2013), so omitting these emissions should not have a large impact on other inorganic aerosol or gas-phase species. Previous work (e.g. Choi et al., 2009) has shown that lightning NO_x is nearly negligible over Southern California. Lightning NO_x and windblown dust sources were also omitted to be consistent with prior works investigating the Los Angeles Basin (e.g., Murphy et al., 2017; Qin et al., 2021). This paragraph has been copied into Section 2.1.3.

Technical comments:
1. Caption Fig 2: The caption refers to 2 different resolution scenarios as "d01." I assume this is a typo, but as I don't think this notation is used again, it may be unnecessary.
Thank you for noticing this, it has been updated.

References

CARB. (2023). *Criteria Pollutant Emission Inventory Data | California Air Resources Board.* https://ww2.arb.ca.gov/criteria-pollutant-emission-inventory-data

Choi, Y., Kim, J., Eldering, A., Osterman, G., Yung, Y. L., Gu, Y., & Liou, K. N. (2009). Lightning and anthropogenic NOx sources over the United States and the western North Atlantic Ocean: Impact on OLR and radiative effects. *Geophysical Research Letters*, *36*(17). https://doi.org/10.1029/2009GL039381

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