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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-term particulate matter modeling for health effects studies in California – Part 1: Model performance on temporal and spatial variations" *by* J. Hu et al.

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Anonymous Referee #1

This paper presents results from a 9-year model simulation of air quality in California that includes chemically-resolved PM2.5 concentrations. Model predictions are compared to published field observations and generally show excellent agreement, both in capturing seasonal trends and absolute concentrations of several atmospherically relevant gas- and particle-phase species as well as total PM2.5. These results are of great importance as the adverse health effects of particulate matter have been shown

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Interactive Discussion

Discussion Paper

to depend not just on mass loading, but on particle size and composition. The availability of accurate long-term, large-scale model predictions will help fill the significant spatial and temporal gaps in ambient particle measurements, and aid efforts to correlate aerosol-related health effects with specific particle-phase species. This work is novel as it combines long-term modeling over a large area with high-spatial resolution in the most populated areas, and full particle size and component resolution. The results are clearly presented, and address a relevant question within the scope of ACP. It is recommended that this manuscript be accepted and published after consideration of several comments.

Response: The authors thank the reviewer for the constructive comments and suggestions that help improve the quality of the paper. We have revised the manuscript to address the reviewer's comments and made response to each comment in this file. The comments are in regular fonts and the responses are in red.

General comments/questions:

1) Can the spatial and/or temporal variation in the sulfate bias say anything as to it's source? It's stated that one possible contributor is the missing DMS from the ocean. However, in figure 9e, the model does a pretty good job along the coast except for a few sites in southern California, where anthropogenic sources would be expected to dominate sulfate formation.

Response: We agree with the reviewer that anthropogenic sulfur sources are likely incomplete in southern California. The sulfate concentrations at the sites in the southern California are $\sim$2 to 3 times higher than in the northern California. Sulfate is under-predicted (with MFBs of around -0.5 to -0.6) at most of the sites along the coast in the northern California, and this is likely due to the missing ocean emissions. Sulfate under-predicted by an even larger amount (with MFBs around -1.0) at the southern California sites. We think some anthropogenic sulfur sources are likely missing in addition to the missing ocean emissions which produces the large under-prediction of

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sulfate. We revised the manuscript to highlight the possibility of missing anthropogenic emissions that contribute to the sulfate under-prediction in southern California.

2) On a related note, under conditions of low background particle concentration, nucleation from gas-phase sulfuric acid can be an important source of seed aerosol particles. Is this process included in the model? If so, is any correlation observed between the low sulfate bias and low PM2.5 bias generally in cleaner areas? If it is not included, could this account for some of the observed low sulfate bias? (Fig. 9e appears to show a small, constant under-prediction of sulfate over inland northern California and along the eastern border where aerosol mass is low to start with.)

Response: Nucleation is not included in the current model. Nucleation is important for number concentrations, but does not contribute significantly to the mass concentrations. In the low sulfate areas (northern California and along the eastern border as the reviewer mentioned), predicted sulfate mass concentrations are about half of the measured concentrations, and this discrepancy cannot be explained by nucleation.

3) The general finding that longer averaging times improves model performance is useful, especially as it relates to removing the effects of random measurement errors. However, would the 'smoothing out' of actual variations in PM2.5 concentrations affect the correlation of model results with health effects in epidemiological studies since short-term PM exposure is known to cause health issues? (Pope III, C. A., and D. W. Dockery. 2006 and references therein).

Response: Yes, the longer averaging times smooth out short-term PM variation that could be useful in some epidemiological studies that focus on short term changes in health effects. Our analysis indicates that air quality model predictions are less accurate with the averaging times of 1 hour or 24 hours, and we have conveyed this limitation to potential users of the data. To get more accurate pollutant predictions at shorter timescales would require more accurate representation of emissions, meteorological conditions, and atmospheric chemistry at these time scales. Many intensive studies

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that manually corrected input data have focused on high temporal resolution for short periods (generally less than 1 month), such as the California Regional PM10/PM2.5 Air Quality Study (CRPAQS) (Ying et al., 2008). It is currently impractical to carry out such efforts for a ∼10 year modeling period in which there are a large number of special events that are not represented perfectly by automated meteorology and emissions models. The atmospheric modeling field continues to refine tools that can capture and accurately represent these special cases. For example, the current study includes automatic detection and incorporation of wildfire emissions into the modeling system. This feature was not generally available in previous studies. Future advances will detect transportation patterns responding to traffic accidents or holiday traffic jams, drought effects on biogenic emissions, etc. These future advances will improve our models to have more accurate predictions in both short- (<1 month) and long- (> 1 month) averaging times but they are currently unavailable. At the present time, we have the greatest confidence in model results for health effect studies where >1 month averaging times are useful. This discussion has been added in the revised manuscript.

Specific comments/questions:

Page 21009, lines 2-4. Could the model not also be greatly over- and under-predicting moderate actual concentrations, as long as it did so with approximately equal frequency or over-predict consistently in certain areas and under-predict in others?

Response: We do not think that is the case because nitrate is consistently under-predicted in most sites.

Page 21012, lines 24-29. 'OM' has not yet been defined. Also, on page 21001 line 17, 'OC' is defined as 'organic compounds,' however it appears to be used in this paragraph (and in Fig. 5?) to refer to organic carbon, while 'OM' is used to refer to the total organic mass. If 'OC' does refer to organic carbon throughout the text and figures, the discussion of this conversion should be moved up, maybe to the methods section. If not, a different abbreviation should be used for organic carbon.

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Response: 'OM' now is defined as organic matter, and 'OC' definition is changed to organic carbon.

Page 21024, line 16. If the bias is also driven by missing DMS emissions, this should be changed from SO2 to more general sulfur emissions.

Response: Yes, we changed 'SO2 emissions' to 'sulfur emissions' in the revised manuscript.

Page 21032, Figure 5. The text in the figure is too small to be read easily. I would recommend splitting this into multiple figures. Also, labels (a) - (h) should be defined in the figure caption.

Response: We have split Figure 5 into two figures (Figure 5 and Figure 6) in the revised manuscript and add the labels in the figure captions.

Page 21035, Figure 8. Units for concentrations and inverse wind speeds in brackets should be indicated in figure caption.

Response: Units are added in the figure caption now.

Minor editorial comments

Page 21000, line 4. Is the intended phrase to use "state-of-the-science," ? Response: Corrected.

Page 21003, line 21. 'hpa' should be 'hPa' Response: Corrected.

Page 21014, line 3. Extra period at the end of the sentence Response: Corrected.

Page 21023, line 5. 'atmosphere' is misspelled Response: Corrected.

Page 21035, Figure 8. The legend for 1/u [0,4] and the black dot to its right could be closer. Response: Corrected.

References Provided by Reviewer 1: Pope III, C. A., and D. W. Dockery. 2006. "Health Effects of Fine Particulate Air Pollution: Lines That Connect." Journal of the Air & Waste

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Management Association 56 (6): 709–42.

References provided by authors in Response to Comments: Ying, Q., Lu, J., Allen, P., Livingstone, P., Kaduwela, A., Kleeman, M., 2008. Modeling air quality during the California Regional PM10/PM2.5 Air Quality Study (CRPAQS) using the UCD/CIT source-oriented air quality model - Part I. Base case model results. Atmospheric Environment 42, 8954-8966

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 20997, 2014.