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 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 ~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 sulfate. We revised the manuscript to highlight the possibility of missing anthropogenic emissions that contribute to the sulfate under-prediction in southern California (in the 3rd paragraph of section 3.1).

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 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 (in the 2nd paragraph of section 3.3).

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.
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 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

Anonymous Referee #2

The manuscript by Hu et al., is what appears to be the first in a series of papers to use an air quality model, in this case the "UCD/CIT" model, to develop PM exposure indices for health studies. This one deals with model evaluation. The model domain covers California and some surroundings. They find that while PM2.5 mass is reasonably accurately simulated, nitrate, organic carbon and sulfate are on the low side, while dust is high. They then conduct sensitivity analyses to help explain the issues.

General Comment: The use of air quality models in health studies is a growing trend, though one should tread cautiously. As this paper shows, there can be large biases involved (which is likely a bigger concern than the errors), particularly when those biases are not thoroughly investigated. In this case, organic carbon, sulfate and nitrate are biased low. As noted below, much of the issue is laid to the emissions, but the modeling approach may have concerns here as well.
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.

In response to the first general Reviewer comment, we completely agree that air quality model results should be treated cautiously when used in health studies. That is exactly the objective of the manuscript: to evaluate the accuracy (or bias) in the temporal and spatial variations in the model predictions and to identify the features of air quality model results that could be used in exposure assessment. We identify air pollutants that agree well with ambient measurements and believe those pollutants should be considered in health studies with higher confidence. We also examine the possible causes for biases in predicted organic carbon, sulfate and nitrate. However, we do not think that errors in emissions are the only cause of the model bias. For organic carbon and nitrate, we think uncertainties in meteorological inputs (wind, mixing depth, and relative humidity) and uncertainties in chemical mechanisms are also important factors. We believe these points are clear in the paper.

Specific Comments: Their sensitivity analysis is not well motivated and done in a rather cursory fashion. They suggest that the reason for the low nitrate is that the RH is low from WRF. That may be the case, in which case one should figure out why RH is low in WRF. Instead, they raise the RH by 30%. The nitrate then increases. It is suggested that they find out why the base meteorological model is providing biased results, and have it corrected in a fashion that will capture linkages important to the meteorology. For one, if you increase RH, shouldn't you get more rain? Arbitrarily increasing RH as an output does not provide this natural link. Instead, they cap the RH at 95%. More rain will lower, not increase nitrate. Thus, their approach is a bit one-sided. How well did WRF simulate rain? So far as issues with the meteorological model, they also increased the friction velocity by 50% to decrease an overestimate of wind speed, but leading to a negative bias in wind speed. Whilst they can cite two of their own papers, this would strike me as to atypical practice or everyone would follow suit. Is this generally accepted? Further, given it leads to a negative bias, is this not too much? Also, might the RH problem be linked to this? Should they use a different meteorological model?
Response: In previous work we have conducted a comprehensive study in which we have tested different physics schemes in WRF simulations for California and evaluated the meteorological

results. This results of this study have been publish in a previous paper [1]. Based on that analysis, we chose the schemes that yielded the 'best' model performance and applied those schemes in the current study. We then evaluated the accuracy of the predicted meteorological parameters that are important for air quality predictions. The results show that the 'best' WRF schemes still have bias, and that there are no models/schemes that yield perfect results. Improving the components of WRF to correct these biases is a major research effort that is simply beyond the reasonable scope of the present study. We believe our WRF predictions are representative of the state-of-the-science in the field, and focus on how the remaining biases in the predicted meteorological fields affect the air quality predictions in the current study.

We found that WRF under-predicts RH in California in the current study, consistent with other studies in California [2, 3] Therefore, we have conducted the RH sensitivity simulations to examine how this bias in RH affects nitrate performance. We increased RH by 30% in the air quality model simulations as an upper bound estimate of the nitrate sensitivity to RH. Only RH values were modified in the air quality calculations in the sensitivity simulation, thus, rain and other meteorological parameters were not changed. Our results show that the RH under-prediction does indeed influence nitrate concentrations. This provides motivation for the meteorological community to address the problems in the WRF model, but this task is beyond the reasonable scope of the current study. The 5th paragraph in the section 3.2 was revised to include more information discussed above.

We found the surface wind was over-predicted by WRF, especially for wind speed less than 3 m/s, which is a typical condition for high PM pollution events. Many other studies in California have reported similar findings [1, 3-5].This problem still exists after we utilized the four dimensional data assimilation techniques available in WRF [6]. This problem prevented the accurate prediction of events with high PM concentrations which cause the most severe health problems. A recent study (Mass, C.F, not published, personal communication) found that increasing the surface friction velocity (u*) by 50% improved the surface wind predictions in a complex-terrain domain that covers the state of Washington. We tested this method in the Central California Ozone Study (CCOS) [7] and found improved model performance. In the current study, we conducted 1-year simulations in 2000 to examine the effects of increasing u* by 50%, and the results were included in a previous paper [8]. This method reduced the bias in surface wind and improved air quality predictions. Even with the change, winds less than 1.5 m/s were still over-predicted. Therefore, we conducted the wind speed bias vs. concentration bias analysis to examine how remaining wind bias affects PM performance in the current study. Once again, the results of that sensitivity analysis indicate that wind speed bias is an important driver of uncertainty in model predictions. This provides motivation for the community to improve the WRF model, but enhancing the WRF model to correct this bias is beyond the reasonable scope of the current study. The 1st paragraph of section 2.2 was revised to include more information discussed above.

They suggest that the poor sulfate results are due to the uncertainties in the sulfur dioxide emissions. My understanding is that air quality models in the US are best at sulfate because the sulfur oxide emissions are well characterized as they arise from fuels, and fuel usage and composition are well known, or the emissions are measured. Here, they say the emissions are

uncertain. They should provide a better justification for this conclusion. Are such emissions very uncertain and why? What other demonstration of this is there, particularly given the findings from other studies? Also, a low sulfate might increase the nitrate, but the nitrate is also low.
Response: In the eastern US, emissions of sulfur dioxide are large because of the combustion of coal to generate electricity and the use of transportation fuels with relatively high sulfur content. The sulfur dioxide emissions are relatively accurate because the small number of large sources are well characterized. In California, all large sources of sulfur dioxide emissions have been eliminated during more than 3 decades of emissions controls. The remaining sulfur emissions are small and are not well characterized. The minor emissions from point, mobile and industrial sources are well documented, but the emissions from other sources such as residential, agricultural and natural sources still contain great uncertainties.

Under-prediction of sulfate concentrations in California has been reported in other regional modeling studies [3, 9], using different air quality models (e.g., CMAQ, WRF-Chem). The common findings in sulfate under-prediction across different studies using different models indicate that the sulfate problem is not due to the choice of air quality model. A global model study using GEOS-Chem in which ocean DMS emissions were included showed a better sulfate performance [10]. Therefore, we attributed the some of the sulfate under-prediction to the missing natural sulfur emissions in the manuscript. Further analysis, as suggested by reviewer 1, indicates that missing anthropogenic emissions (such as residential or agricultural emissions) is also likely contributing to the sulfate problem in the southern California. The manuscript has been revised to include the above discussions in the 3rd paragraph of section 3.1.

In the eastern United States where ammonia can be a limiting factor, decreased concentrations of sulfate free ammonia which is then available to form ammonium nitrate. In the present study, ammonia is generally present in excess in both the San Joaquin Valley and the region east of Los Angeles, meaning that nitrate formation is not dependent on sulfate concentrations. The nitrate under-prediction in the current study is caused by other factors, such as the uncertainties in emissions, under-predicted WRF relative humidity, and/or the chemistry mechanism, as discussed in the manuscript. Under-prediction of nitrate in California has also been reported in other studies [3, 9, 10].

Given it is a long term simulation, a question arises as to how well does their model simulate deposition over that same period. I have seen evaluations of other air quality models for deposition. A publication of such an evaluation for the model they have chosen would be of interest. They should provide an update here if possible. This should be done, particularly, considering the modification to the friction velocity and the low RH bias, which probably means a low bias in wet deposition, which should lead to higher pollutant concentrations.
Response: We thank the reviewer for the constructive suggestion and we think that an evaluation for the model performance on deposition would be interesting. Unfortunately, deposition data were not saved in the current model simulations. Thus we cannot perform such analysis at this point.

They suggest that many of the problems are due to emissions. This should be better demonstrated. One can get very similar problems if some other issue is driving the problems. What if some other process is not being captured correctly? For example, might it be that the model is

dispersing material too rapidly/not rapidly enough? I would have liked to see more support using other approaches, e.g., from recent tunnel studies or satellite data. Their paragraph, starting "Figure 5b and c: : :" says things work in some places and times, but not other places and times. They conclude the emissions are uncertain, which is a potential cause. The model may also lead to those same biases due to the parameterizations of physical processes, and, indeed, one might find the latter a more likely happenstance. Given the complex meteorological situation in the area, it could be quite challenging for a model to correctly capture dispersion. They hint at this possibility, but do not give it the importance it deserves, and how it might be addressed, or how it might impact the use of model results.

Response: We agree there are other issues besides the uncertainties in emissions and meteorological inputs that need to be considered, such as some uncertainties in the chemical mechanism. In the manuscript, we have pointed out that uncertainties in the nitrate and SOA chemistry mechanisms are also possible causes for bias in the OC and nitrate predictions. Model grid resolution is another potential issue that may contribute to the bias in the model predictions when comparing the 4X4 km grid concentrations to point monitor measurements [8].

We have tested our model approach for individual processes, including horizontal advection, diffusion, dry deposition, and wet deposition. The test results were included in a previous study [8]. In summary, the biases due to the numerical parameterizations of physical processes are small. We agree there are uncertainties in the dispersion calculation that contribute in a minor fashion to the biases in model results. But we think these uncertainties are mainly due to the difficulty in accurately predicting wind fields in an area with such complex topography. The numerical solvers of the pollutant dispersion do not introduce significant error.

The top boundary of the model is of concern. It is only 5000 m. For such long simulations, and in such complex topography, aren't there periods where various processes might occur that lead to exchange above 5000 m, e.g., convective storms, atmospheric waves over the mountains in the domain? Might stratospheric intrusion play a role?1,2 They should conduct a sensitivity study to assess how the lower model boundary condition impacts their results. It would also seem that the low model height might negate any modeled role of air craft or lightning emissions.

Response: Measured planetary boundary layer height (PBLH) data showed that PBLH in California are generally lower than 2 km [1]. We have done sensitivity simulations to study the effects of changing the model height from 4000 to 5000 m. The results indicate less than 3% difference in general [8]. A recent modeling study in Europe used 14 layers up to ~6000 m and yielded satisfactory results [11]. Note that the use of relatively shallow vertical domains is only appropriate in regions with well-defined air basins and might not be appropriate for locations in the eastern U.S. or other regions with moderate topography. The information is added in the 3rd paragraph of section 2.1. The choice of a model top at 5000m in the current study was an appropriate approach to limit the computational burden of the calculations conducted for California.

We agree that there are certain periods when exchange above 5000 m occurs. A few case studies have examined the stratospheric intrusion of ozone in California [12-14]. The stratospheric contribution can be significant for springtime ozone, but has less impact in summer and winter [12]. Nevertheless, our predicted surface ozone is in excellent agreement with measurements. Including the stratospheric intrusion process will improve the day-to-day variations in the ozone

predictions during those springtime events, but it should not change the majority of the ozone performance or the monthly/seasonal variations of ozone predictions.

Long range transport can also influence air quality in California [15]. The impacts are determined in boundary conditions in our model simulations. In the current studies, we interpolated the ambient measured upwind concentrations and seasonal background concentrations from literature as boundary conditions [16]. An ongoing effort is underway to use global model predictions to provide temporal-spatial varying boundary conditions for our model. This will improve our model ability to capture the influence of long range transport on local air quality and will be documented in future studies.

High-altitude aircraft emissions and lightning emissions are currently not included in the official emissions inventory provided by the state of California for the current study. Some studies indicated that aircraft emissions could have great impact on surface air quality, especially for ultrafine particles [17, 18]. Future studies should consider the aircraft emissions as they become available.

They say "For the first time, a _ decadal: : :" I do believe that the US EPA has conducted air quality modeling over the whole US, which includes California, for a fairly long time period and may have used that for health analyses. I am not sure to what degree they have used their results for health studies, but that should be checked. Even if they have not, the "for health effects studies" is not that relevant in this case since this really is a model evaluation, and I am not sure if the "for health effects studies" changed the model evaluation analysis appreciably. How does this effort differ from any other evaluation of a long term model application? Also, the whole domain they use is not 4 km, just a subset, so they should alter their lead-in sentence, e.g., add "with populated regions modeled at a resolution as fine as 4 km".
Response: To our best knowledge, this is the first study in California to model 9 years of air quality with 4km resolution covering the major population areas for health effect studies. As discussed in the Introduction section, we try to address the temporal, spatial, and/or chemical limitation problems in the ambient measurement data that are traditionally used in health effect studies. So we use air quality models to address the limitations of using measurement data alone. For that purpose, in addition to a traditional model evaluation, we focus on identifying the model features that could be used in health effect studies, such as what months/seasons, what areas, and what chemicals in the model predictions are accurate; and in what averaging times, model predictions are suitable for health effect studies. The accurate model results should be considered first in the health effects. We have emphasized our purpose in the 3rd paragraph of section 1, and added two paragraphs to discuss the implications of the model results for use in health studies in the section 3.3 in the revised manuscript.

We have further modified the sentence to "with 4 km horizontal resolution over populated regions" in the Abstract and Conclusions.

Not sure if "These results will be improved in future studies." belongs in an Abstract, or even in the paper. It begs the question, why aren't they improved here? Is this paper premature? Also, it does not appear in the paper.
Response: We delete the sentence from the Abstract in the revised manuscript.

In the end, too much of the less than desired agreement with observations is put to the emissions and the meteorological model as opposed to potential issues with the model itself. Further, they need to provide some idea of what should be done. In terms of model evaluation, they should look at the European AQMEII effort.3 (They might also look at other model-based air quality model-health studies in Europe and elsewhere. 4,5) The US EPA has looked at model evaluation under emission uncertainty.6 I would suggest they look at those efforts a bit more. They should consider raising their model domain height given the complexity of the terrain and the potential for longer range transport, convective storms and stratospheric intrusion to be of an issue. They should try to figure out why their meteorological model provides inadequate inputs in its base formulation, e.g., the need to slow winds down and increase RH.

Response: We thank for the reviewer's suggestion. Discussions on several potential issues, e.g., model uncertainties and domain height, have been added in the revised manuscript (see the previous response to individual comments). We also thank the reviewer for the references. The references suggested the importance of evaluating model results in both the temporal and spatial aspects, which supports the analysis approach in the current study. Some of the references have been cited in the 3rd paragraph of section 1 in the revised manuscript. We focus on discussing how the bias could affect the PM predictions using sensitivity analysis, instead of enhancing the meteorological model. We think extensive future efforts should be directed to meteorological model improvements, but that work is beyond the reasonable scope of the current study.

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