Response to "Interactive comment on 'Multi-scale modeling study of the source contributions to near-surface ozone and sulfur oxides levels over California during the ARCTAS-CARB period' by M. Huang et al."

by Anonymous Referee #1, Received and published: 5 January 2011

We would like to thank both reviewers for their comments and suggestions. Both of them mentioned that the quality of figures needed to be improved. We increased resolutions of all figures and compiled them in an easy-reading way.

This paper uses a multi-scale (12 km and 60 km resolution) modeling framework to a assess near-surface ozone and sulphur oxides over California during a week in June 2008. It uses observations from NASA DC8 flights and ground based sites for model evaluation and focuses on southern California. In particular the impact of local biogenic, forest fires and maritime emissions are estimated. Further the impact of long range transport from Asia is also considered. Unfortunately the results are rather poorly constrained given the uncertainties in the emission inventories. In several cases the comparison with observed concentration data is poor (or not shown) and yet the model is then used to examine source contributions. This undermines the overall conclusions.
On the whole the presentation is good, but the text in all of the figures is too small to read in print (I had to zoom in on the electronic version) and in several cases more detail is needed about exactly how the data were treated.

The model performance is a reflection of our current capabilities. We have added additional information to the paper regarding the model performance. Please see the details below.

We also added adjoint sensitivity analysis for SO_2 in section 3.7.

Specific comments
P27779, L 12. The abstract appears to suggest that the enhancement in of surface SO4 from Asia is quite large, but this only occurred during one flight and does not represent what was observed during the other flights.
Yes that referred to a specific long-range transport episode starting to influence U.S. west coast from June 22, and the impacts on SC domain are shown on Jun 24. We modified the abstract.

P27784, L 10-19. More is needed on the ground based sites and instrumentation. Firstly it is not clear which sites are in which networks. Sites are plotted in various figures, but it is not clear which networks they come from, therefore whether they are urban or rural. This is critical when comparing observations with models. Which networks do the "six SC sites" in Figure 2 come from? How do they relate to the networks in Figure 8b or in the map of 8c? Secondly more is required about the instruments used at the ground-based sites, beyond reference to a web site.

The six SC sites (shown in Fig 1a) provided observational O_3 and SO_2 (hourly) concentrations as model comparisons. SO_4 measurements were taken from various networks including STN (mostly urban), IMPROVE and CASTNET sites (rural and remote), which are used in Fig 8. None of the CASTNET sites belong to SC domain in this study, only one (San Gabrial) belongs

to IMPROVE, other four are IMPROVE sites. Complete information for six surface sites are tabulated in the supplement. (http://www.arb.ca.gov/qaweb/sitelist_create.php).

The figure 8 (revised version) clearly shows the SO_2 comparisons at SC CARB sites, SO_4 comparisons at all CA IMPROVE/STN/CASTNET sites.

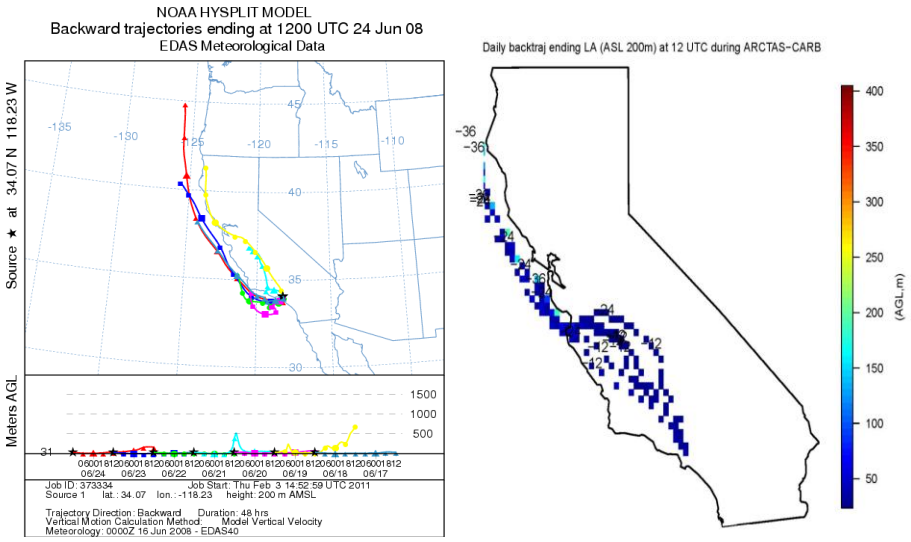
P27787, L 8-9. Define exactly what is meant by the flight time average. This is confusing given that flights occurred on more than one day. Did each flight occur exactly at the same time of day? Similarly define exactly what you mean by averaged daily maximum. Are these just of the days of the flights or the whole week?
Flight-time: 15-24 UTC; which was describe in line 1-3 "Three (18, 22, 24 June) out of the four flights took measurements over the SC area during approximately 15:00–24:00 UTC (08:00 a.m.–05:00 p.m. LT)". They are now also defined multiple times in text and figure captions.

P27787, L17-25. The text here focuses on the differences between the 12 and 60 km resolution simulations and does not really emphasise enough the discrepancies with the observations. E.g. that neither model run simulates the full range of observed ozone concentrations (i.e the lowest or highest values) during the flights nor the lowest (night-time) concentrations observed at the ground-based sites. Further some explanations for these discrepancies should be discussed and any implications to the results of the sensitivity studies considered. Is this related to emissions, resolution, boundary layer dynamics, choice of sites (urban or remote), etc?

Model discrepancies are caused by: model resolutions, model input uncertainties, as well as physical and chemical mechanisms. A major contributor is model input uncertainties, from such as emission inventories and to a less extent, meteorological fields. Overall, models overpredict summertime US near-surface O_3 (not just ours).

Specifically,
1) We cannot confirm nighttime model NO predictions as there were no NO measurements during these periods. However, we found the model underpredicted NO_2 for nighttime, which indicates that the insufficient titration is one of the possible reasons for night time overpredictions on O_3.

2) Another possible factor is the transported background, which will be demonstrated below:



The daily back-trajectories during the flight week-based on 12km WRF flow fields (ending 12 UTC, 5am local time at the LA site in SC) (right), is shown together with the trajectories generated by HYSPLIT (left, http://ready.arl.noaa.gov/HYSPLIT.php) based on EADS.

Both trajectories indicate that, 12 h and 36 h before the ending times (which are 00 UTC, 17 local time), the air-masses were at the Central Valley and/or at the north coast areas, below 200m. The model shows a tendency to overpredict O_3 over the coastal areas, therefore, transport brought high level of background O_3, contributing to the overprdiction of O_3 at nighttime/early morning.

P27789, L7-10. The sensitivity calculated using Eq. 1 uses only model data, but is said to be for "each of the one-minute flight data below 1000 m". Please be more specific about exactly what this means. i.e. is this simply for the time and location of each flight data point?

Final version of flight data were created by NASA--they merged measurements at different time resolutions in 1-min time resolution. Here we extract model results (using interpolation) for corresponded species concentrations at that location/time. This was described in section 2.1.

P27791, L13-15. Where data from different teams have been combined, some comment needs to be made as to how this was done given that there is clearly a difference between the measurements made by the different teams.

First, we calculated SO_2 and SO_4 profiles separately. As there were two SO_2 and two SO_4 teams, if there are two measurements, we took the average. If there was only one, we used it directly. If there is none data, it was treated as NA. Then we calculated SO_x=SO_2+SO_4, for locations with no SO_2 or SO_4 data, it was treated as NA. It's been added to text.

P27791, L25-28. The comparison between the observed and modeled sulphur for the 60 km resolution simulation is extremely poor. The comments made here all are about the relative predictions made by the two different resolution simulations. Surely it is important to point out here that the 60 km resolution run completely fails to simulate

the observations.
We have added a paragraph at the beginning of section 3.3 to clarify the role of 60 km simulation. The 60 km simulation was completed first, we found consistent significant negative biases by comparing with observations, and were strongly motivated to conduct the 12 km simulation to better evaluate the impacts of model resolution, emission inventories and other factors on simulated SO_x concentrations.

P27792, L5-6. Again here it has to be made clear that the 60 km run fails to predict the observed sulfur.
Please see above.

P27793, L24. How is VOC age calculated? Which VOCs are used? What assumptions are made about OH concentrations, for example?
Details can be found in: Tang, Y., et al. (2004), Multiscale simulations of tropospheric chemistry in the eastern Pacific and on the U.S. West Coast during spring 2002, J. Geophys. Res., 109, D23S11, doi:10.1029/2004JD004513. "we also define a volatile organic compound (VOC) age using ethane as an indicator that is related to ethane emission and decay rate." T=constant×log(HC1/HC3), HC=ethane and ethene

P27794, L18. At such long VOC ages, it is likely that the VOCs observed were at low concentrations, possibly close to detection limit. If so does this affect the uncertainty in these calculated ages?
As described above, VOC age is a relative ratio, and is based exclusively on model calculations, so detection limit is not an issue.

P27795, L9. Equation 3 assumes that CO and SOX behaves similarly. E.g. have similar lifetimes and thus can be transported similarly across huge distances. Can this be justified?
We added the words "upper limits" in text. SO_x usually have shorter lifetimes than CO. Under long-range transport events that are associated with "dry fronts", a large fraction of SO_x can be transported across the Pacific.

P27795, L23- (Fig 10). Quite clearly the SOX emissions from the CARB EI and NEI inventories are very different with the NEI being substantially lower than the CARB, for the SC area. The model results are therefore not exactly surprising.
Not surprising, but quantifying the basic model response is essential to set the stage for subsequent sensitivity studies and scaling factor evaluations.

P27795, L27 – P27796, L1 (Fig 10). The text refers to the data plotted in Fig 10c as being the average emission rates over the six SC surface sites, whilst the Fig caption simply says it is over the SC. Is this an area average, or the average over the 6 sites, and if the latter presumably each emission grid box must still represent an area around each site?
We corrected captions for Fig 10c. It's for the average over six SC sites, locations shown in Fig1a.

P27796, L13-14. The model simulation with the NEI underestimates the SOX observations by a factor of 10. The model simulation with CARB EI underestimates the SOX observations by a factor of 2. There are clearly big issues with the SOX emission estimates for the SC area. In addition there will be model errors, e.g. transport terms, and uncertainties in the observations. The estimate of 40-50% of the SOX coming from shipping surely must have large uncertainties associated with it and thus the validity of any assessment of the effect of maritime emissions must be questioned, or at the least presented as highly uncertain.

Results are obviously uncertain, but it is this great uncertainty in emissions that motivates our study, and our comprehensive modeling yields no satisfactory explanations other than our core finding that large underestimates of maritime emissions are central to the systematic low bias in contemporary SO_x emissions inventories for California.

The typical uncertainties common to all regional chemical transport modeling studies are not so large as to undermine the core findings. We note that previous modeling studies of SC shipping emissions did not validate the model simulations; this study first evaluates model simulations of a range of species using 3D observations, specifically to address these uncertainties. Model errors in transport terms and uncertainty ranges in aircraft and routine monitor observations in this study are exactly the same as those in all regional scale process modeling and regulatory studies, and are not large enough in those cases to question the net results, which often deal with much smaller and more nuanced changes. In this case, any such errors are swamped by the emissions errors. The WRF simulation employed here was driven by reanalyzed regional-scale meteorology (NARR) and thoroughly evaluated with observations and previous modeling works, and the adjoint sensitivity study is conducted to better understand impacts of the maritime/terrestrial emissions and transport of SO_x on surface sites SO_2 levels, vertically and temporally.

P27796, L25-28. Simply scaling the results by the observed/modeled ratio will not necessarily correct the uncertainties imported from the original CARB EI if the errors in the EI vary with emission sectors.

We removed the scaling part by using consistent scaling factors, and agree that the results regarding the contributions from the maritime emissions presented here are of high uncertainties. However, a sectoral, spatial, temporal attribution of errors and uncertainties is beyond the scope of this study. Optimal methods (such as data assimilation) can provide complete emission scaling factor matrix and reduce the mismatches between observations and model simulations, and will benefit from better observational datasets, not only over the SC are but also over the upwind regions such as north and central coastal areas. Our results are a first step toward understanding the underestimated emissions, and future assimilation may be able to more accurately quantify and reduce uncertainty in sectoral emissions.

P27798, L3-6. The conclusions made about the impact of maritime emissions on the VOC-limited and NOX-limited state are dependent on the model reproducing the observed NOY and O3/NOY. It is important to first demonstrate that the model can reproduced the observed state (presumably there are observations available from the DC8

flights).
We evaluated the NO_y as well as O_3/NO_y model results along all DC8 flight tracks, modeled are similar as observed, which is described in detail in a forthcoming manuscript (Huang et al., 2011, in preparation). In the revised paper, we have added a few sentences to the text, briefly describing the validations.

Technical Corrections
It would be helpful to provide a map of California annotated with the key locations referred to in the text, measurement sites and major cities etc. This would be helpful to those of us less familiar with the geography of the region.
All sites locations related to this study (CARB, IMPROVE, CASTNET,STN) are shown in Fig 1a and Fig 8. We also prepare a complete list of site descriptions for the six CARB sites shown in Figure 1a. (Please see supplement)

P 27782, L 23. I thought ITCT was simply "Intercontinental Transport and Chemical Transformation".
I think they are interchangeable. (reference: http://www.esrl.noaa.gov/csd/ITCT/)

P27783, L 24. "2" should by "22" – date of 3rd flight.
Corrected.

P27785, L 1-7. Although it is stated the different LBC are used for the 2 different resolutions then next couple of sentences only explain how they differ for gases and aerosols. How do they vary for different resolutions?
Time and spatial resolutions are summarized in Table 1 in detail.

P27785, L 25. Please provide a reference or further description of the CARB emission inventory.
The CARB EI was received in July, 2009, by personal contact with co-authors from CARB (Kaduwela, Ajith and Cai, Chenxia).

P27786, L 11-17. The diurnal variation in the BL height was not immediately clear to me, because of the time zone. Although in the titles of the plots in Figure 1 the local time is given, the text is far too small. LT should also be defined as local time. I would also suggest including LT in the text here.
Done. Now shown in both text and figure titles.

P27787, L 13-15. This doesn't really make sense as the magnitudes are different. It would be better to state that the patterns are similar.
In the revised text, we described 12 km and 60 km base case O_3 separately.

P27787, L18. "Compared to the 60 km simulations, the 12 km simulations ....."
Please see above.

P27789, L12. You state that "Both resolutions show that ...", but at this point you have only mentioned that these sensitivity runs were done for the 12 km resolution runs (P

27787, L28) and it is not until the next paragraph (P27789, L21) that you state that these runs were also performed at 60 km resolution. This needs to be clarified.
We corrected the text.

P27790, L23-25. This statement is ambiguous. Is the point that the fire and biogenic emissions have a greater affect outside the SC area than inside it?
Yes.

P27791, L18. Which three regions?
SC, SF and CV. We reworded.

P27793, L17. Suggest providing local times as this is what is important in this case.
Done.

P27794, L15. Define THD. Where is it?
Trinidad Head. (NOAA's sounding location to sample air entering western US: -124.16W, 40.8N, elev. 20m). We spelled it out.

P27794, L23. If this is not shown what is being referred to in Fig 9a?
China CO% contributions are not shown but we did show the VOC age to make our point.

P27796, L1. You refer to flight leg 3. Figure 9 has, in several panels areas circled and labelled as 1, 2 or 3. Presumably these are the flight legs, although this is not stated in the caption, and is confused by some labels being both 2 & 3 and some pointing more to spikes in data than a flight leg and sometimes with two 2s in a single plot.
We modified this in text. See detailed explanations in response to previous questions.

P27796, L12. Surely section 3.7 is about the effects of maritime emissions, not just maritime SOX emissions.
Done.

P27798, L8-12. These comments on long-range transport and Asian impact on O3 are not conclusions from the work presented in this paper.
That's a transitional sentence to summarize the content of this paper. The paper focuses on transport and local emissions contributions to SO_x and local generations of O_3 contributed from several sectors. The transported O_3 was described in a previous paper as cited.

P27799, L18-24. This paragraph is out of place in the conclusion as it refers to work not mentioned previously in the paper. It should come earlier.
We moved this paragraph to section 3.3 and added a concluding paragraph.

Tables and Figures
Table 1. Several acronyms need defining.
GFS: Global Forecast System
NARR: North American Regional Reanalysis
MODIS: Moderate Resolution Imaging Spectroradiometer

Other acronyms such as STEM, RAQMS,NEI, CARB are spelled out in text.

Table 5. The VOC ages is given very precisely. I would expect considerable uncertainties and over the course of a flight there must by variability (air of different ages sampled).
See explanations above about VOC age calculations.

Plus, we added: Corrections of Table 6: unit in table 6 should be mole/km^2/h, instead of mole/km^2/day.

Fig 1. Text of axes and titles too small. Arrows too small. Need a scale for arrow size. Give local time in caption.
Done. We changed the PBLHT into shadings.

Fig 2. Text of axes too small. One map has islands the other doesn't.
Done.

Fig 3. Text of axes, titles and legends too small. "Average flight time"? How is the observed data from the six sites combined? Exactly what data is extracted from the model to represent the ground sites? i.e. grid boxes, levels?
Figure 3a-d are model output, flight time means 15-24 UTC (8-17 local times). For Figure 3f, at each time step (hourly) , we define observations or model extractions at each site=Obs(i), i=1,2,3,4,5,6, and averaged all sites. All surface sites are in the lowest model level in both 12km and 60km configurations, so only horizontal grid interpolation was used for extracting 12km model results. For 60km extraction, temporal interpolation between 6-hour time steps was also applied.

Fig 4. Text of axes, titles and legends too small. (b) needs units of ppb. Suggest provide local time.
Done. Local time are now provided in both text and figure captions.

Fig 5. Text of axes too small.
Done.

Fig 6. Text of axes, titles and legends too small. Suggest provide local time.
Done- local time provided in both text and figure captions.

Fig 7. Text of axes, titles and legends too small. Suggest the scales of plots a, b and e are the same and also the same for c, d and f. i.e. so you can compare observed values with modelled.
Done.

Fig 7, g-i. Text of legends too small. Define TR.
Done. The TR case refers to 12km no-maritime emission case. Legend has been modified. We also correct the SO_4 unit in Figure 7h.

Fig. 8. C, Which are STN and which IMPROVE sites?
We changed this figure to better show site locations and colored STN and IMPROVE sites with triangles and squares. Please refer to section 2 in text for detailed descriptions and links.

Fig. 9. Text of axes, titles and legends too small. (a) includes VOC age. (c) and (d) flight altitude included.
Done-included in caption.

(e) and (g) need to be clear that the altitude limit refers to the flight leg.
In fact not limited to flight legs, but refer to all outbound flight path. (appox. 17-21UTC, 10-14 local time). This is been added to the text.

(f) Too small to really get any idea of which trajectories are for which parts of the flight. needs units of ppb. Suggest provide local time.
Done. Both SO_2 and SO_4 are shown in ppb now.

Need to explain what the red
Circles 1, 2 and 3 are corresponded to approximate locations in time series and vertical plots (Fig9a-d), where elevated concentrations are observed, DC8 took circles at these locations to study vertical distributions of pollutants, not exactly "flight legs". We modified the text.

(a) and (b) how are the vertical profiles compiled given that the aircraft made several ascents and descents? Similarly how are the model vertical profiles compiled?
The scatter plot of vertical profile composites were compiled from all actual observations along the outbound of June 22 flight path, and the corresponding model products at the same times and locations.

Fig 10. Text of axes, titles and legends too small.
Done.

Fig 11. Text of axes, titles and legends too small.
Done.

Fig 12. Text of axes, titles and legends too small. (b) difference between 12 km cases? base and no-maritime?
Yes. Now replaced with no-maritime case NO_y to better study NO_x-VOC regime.

Fig 13. Text of axes, titles and legends too small.
Done. We also add photochemical age comparisons as e,f for base and no-maritime cases, respectively.

References

Huang, M., Carmichael, G. R., Kulkarni, S., Spak, S. N., Chai, T., Oltmans, S. J., Jaffe, D. A., Streets, D. G., Kaduwela, A., Weinheimer, A. J., Huey, G. L.: Source attribution at western U.S. coastal receptors and the impacts of coastal-inland transport of pollutants and local fires on surface air quality, in preparation, 2011.

Tang, Y., et al., Multiscale simulations of tropospheric chemistry in the eastern Pacific and on the U.S. West Coast during spring 2002, J. Geophys. Res., 109, D23S11, doi:10.1029/2004JD004513, 2004.