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## 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.

## **Anonymous Referee #1**

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This paper uses a multi-scale (12 km and 60 km resolution) modelling 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 longrange 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

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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.

## 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.

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.

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?

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?

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?

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.

P27791, L25-28. The comparison between the observed and modelled 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.

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

P27793, L24. How is VOC age calculated? Which VOCs are used? What assumptions are made about OH concentrations, for example?

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?

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?

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.

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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?

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.

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

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).

## **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.

P 27782, L 23. I thought ITCT was simply "Intercontinental Transport and Chemical

Transformation".

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

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?

P27785, L 25. Please provide a reference or further description of the CARB emission inventory.

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.

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.

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

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.

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?

P27791, L18. Which three regions?

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

P27794, L15. Define THD. Where is it?

P27794, L23. If this is not shown what is being referred to in Fig 9a?

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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.

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

P27798, L8-12. These comments on long-range transport and Asian impact on O3 are not conclusions from the work presented in this paper.

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.

Tables and Figures

Table 1. Several acronyms need defining.

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).

Fig 1. Text of axes and titles too small. Arrows too small. Need a scale for arrow size. Give local time in caption.

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

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?

Fig 4. Text of axes, titles and legends too small. (b) needs units of ppb. Suggest provide local time.

Fig 5. Text of axes too small.

- Fig 6. Text of axes, titles and legends too small. Suggest provide local time.
- 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.
- Fig 7, g-i. Text of legends too small. Define TR.
- Fig. 8. C, Which are STN and which IMPROVE sites?
- Fig. 9. Text of axes, titles and legends too small. (a) inclues VOC age. (c) and (d) flight altitude included. (e) and (g) need to be clear that the altitude limit refers to the flight leg. (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. Need to explain what the red circles 1, 2 and 3 refer to. (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?
- Fig 10. Text of axes, titles and legends too small.
- Fig 11. Text of axes, titles and legends too small.
- Fig 12. Text of axes, titles and legends too small. (b) difference between 12 km cases ? base and no-maritime?
- Fig 13. Text of axes, titles and legends too small.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 27777, 2010.

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