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 #2, Received and published: 25 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.

The authors use a regional chemical transport model driven by WRF meteorology in two different horizontal resolutions to explain the ozone and sulfur oxides measured during the ARCTAS-CARB campaign in summer 2008 and identify the sources for the ozone and sulfur oxides during the period focusing on southern California. Both local (including anthropogenic, biogenic, and fire) and long-range transport contributions to California near-surface ozone and sulfur are quantified. They found long-range transport from Asia did not significantly affect surface air quality in the study region during the period. Two emission inventories are used to study the modeling sensitivity to the emission inventories. Maritime (mostly shipping) emissions are found significantly affecting the air quality over South Coast region of California.

General Comments

I find this paper to be well presented but the experiments could be better designed for authors' purpose for 1) future emission reduction strategies; 2) better understanding of EI's uncertainties. In order to identify the sources of sulfur oxides, the model performance is relatively poor (underestimate surface SC SOx by a factor of 2). Model bias of sulfur simulations makes the authors' conclusion less valued. I would only suggest this paper for publication if the authors address the comments listing below.

Specific Comments

1. In model description part, what are the domains of three model simulations? Should have more description of the model configurations, although you cite a reference.
We have added descriptions and related links in section 2.2.

2. Why are the LBCs for aerosols and gases of 60 km regional scale simulation different (aerosols from STEM tracer and gases from RAQMS)? No description of LBCs in the text for 12 km simulation, although you put it in table 1.
We added a sentence for 12km LBC sources in the text.

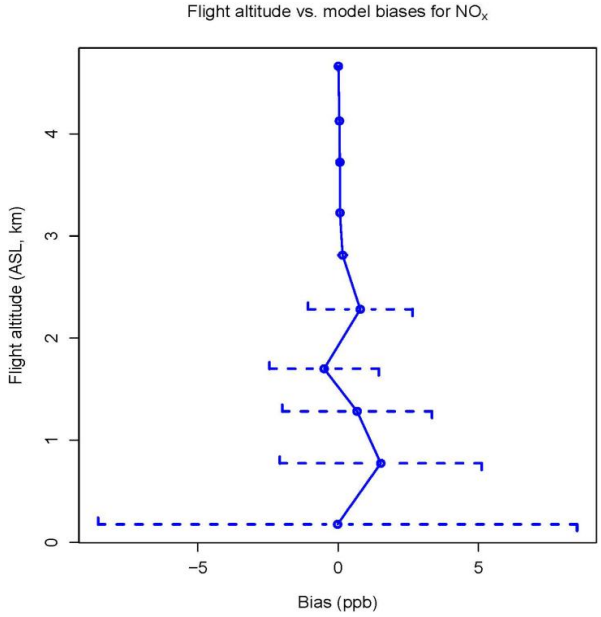
3. I am confused by your conduction of the simulations. The purpose of your study is to investigate the impact of resolution and EI uncertainties on ozone and sulfur simulations. So why shouldn't you design more clean simulations, such as 12 km simulation with both EI's and a 60 km simulation with one of the EI's. It should be better than mixing the effects from resolution and EI together.
We have added a paragraph at the beginning of section 3.3 to clarify the role of 60 km simulation. The 60 km domain was used in forecasting stage in support of the ARCTAS experiment, and is often used in global models (so it is important to evaluate), we found consistent significant

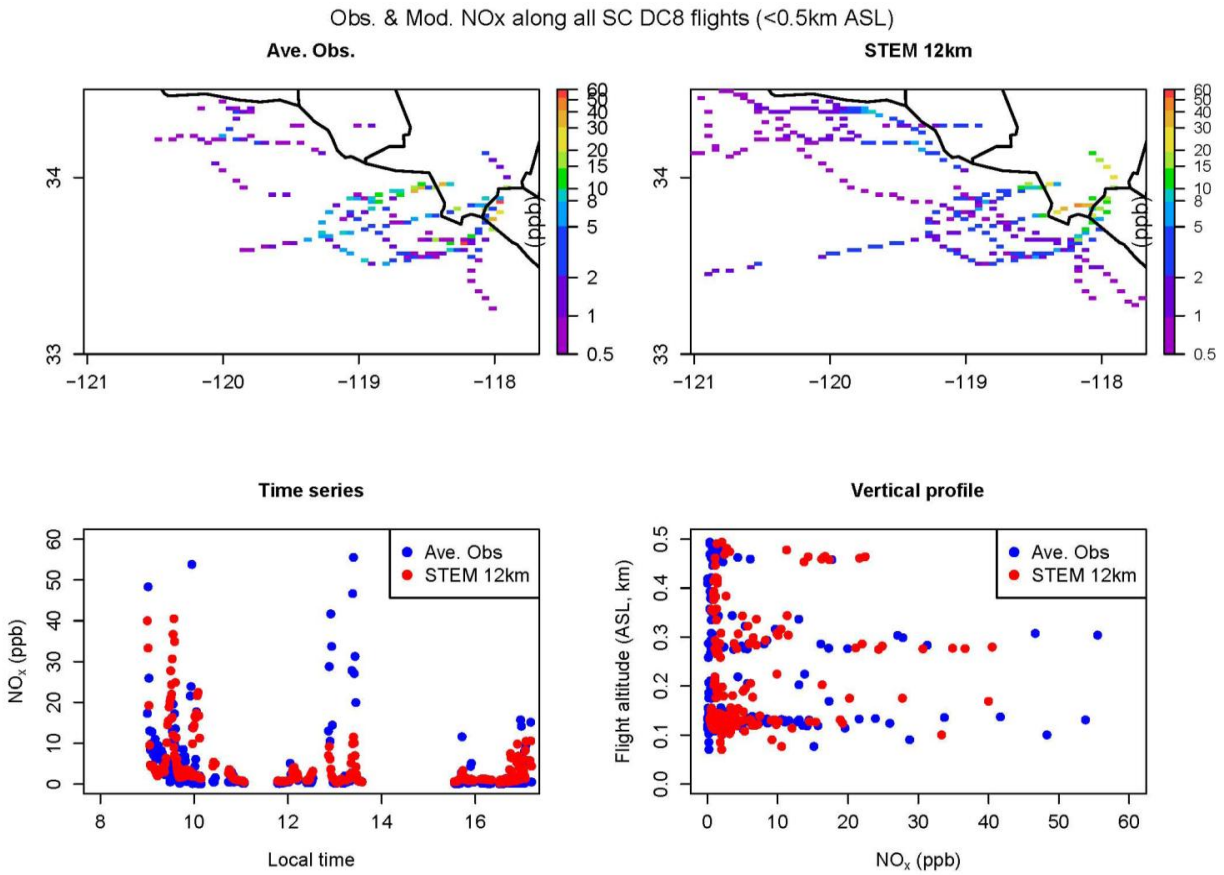
negative biases by comparing with observations. This motivated the 12 km simulations designed at post-analysis stage to assess the impacts of model resolution, EI and other factors on model prediction skills, using the best available input.

4. For section 3.1, a table to show the comparison between ozone observations and simulations as table 3 is preferred.
We have expanded O_3 discussions in section 3.1 and included statistics. Complete comparison for surface O_3 and other NO_y species along DC8 flights will be contained in a forthcoming manuscript. (Huang et al., 2011, in preparation). We also added a paragraph to describe the validation of NO_y and O_3/NO_y in section 3.7.

5. What's the problem for ozone simulation at night (Fig 3c)? The NOx titration issue in the model? In case that NOx is important to your ozone simulation (you also discussed the NOx-regime versus VOC-regime in later section), it is worth to include an evaluation of modeled tropospheric NO_2 column with satellite measurements.
First of all, we added descriptions about the chemical mechanism used (SAPRC 99) in section 2.2.

To answer the question of NO_x validation:
1) The comparison of NO_x along all DC8 SC flight tracks during ARCTAS-CARB. There are two NO_2 measurement teams (NCAR and UC-Berkeley). We calculated mean values of their measurements and added them to the NCAR-measured NO concentrations.





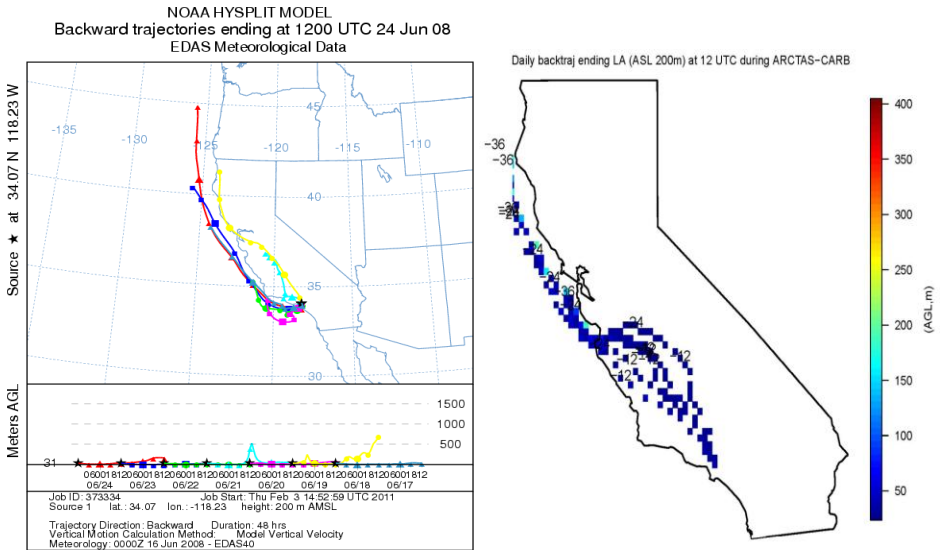
The model shows similar spatial and temporal pattern as the observations below 0.5 km ASL. The model overall shows highest positive biases at 1-2km.

2) The comparison of mean NO_2 diurnal circles with the six SC ground sites was also conducted: The model underprediction on NO_2 during nighttime and early morning are associated with the overprediction of O_3.

To answer the question of reasons for overprediction of O_3 during nighttime and early morning:

1) We cannot confirm the 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.

6. In page 27793, you calculate the factors for nighttime. But it's not very clear to me why don't you just use the nighttime surface observations to calculate the factors?
We removed the scaling part.

7. In section 3.4, how to calculate the age of VOC and the China contribution? Need more description here.
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."

anthropogenic China CO%= anthropogenic China CO in ppb/(sum of all eight regions anthropogenic CO in ppb)×100%, the anthropogenic CO for each region are calculated by the tracer model.

8. In line 25-28 of page 27796, the authors argued to scale SO_2 and SO_4 in order to correct the uncertainties from CARB EI, but how could author prove that the model bias of sulfur is not from model itself (e.g., chemical production and loss processes of sulfur and also the deposition of SO_4)? At least, author should show the comparison of modeled SO_2 to aerosol SO_4 ratios with observations. In this way, we can know if the major atmospheric oxidation processes leading to aerosol sulfate formation are

captured correctly in the model.
We compared the observed and modeled $SO_2\%=SO_2\times100\%/(SO_2+SO_4)$ in Fig 7, and mentioned OH comparisons in text (r=0.47 and 0.41 for 12km and 60km, respectively).

The model, designed specifically for sulfate chemical transport, has been repeatedly evaluated, and has been shown to accurately simulate the atmospheric oxidation processes leading to aerosol sulfate formation (Carmichael et al., 2002; Guttikunda et al., 2003).

9. In line 7 of 27799, what's 2008 emission? The paper only talked about 2001 and 2005 inventories. And also, how could authors exclude that some of the simulation underestimate may be from model bias by itself?
We meant the CARB 2005 EI, developed for the 2008 ARCTAS-CARB campaign. We reworded.

Technical Comments
1. Figure is too small to read. It's very hard to get the information from figures themselves.
Modified.

2. In abstract, line 5 of page 27779, change "simulations with the STEM …" to "simulations using the STEM …".
Done.

3. Line 6 of page 27779, change "used to assess" to "conducted to assess".
Done.

4. What is the unit of ug/sm3 in line 12 of page 27779?
It meant m^3 under standard temperature and pressure. We changed into ppb.

5. Line 5 of page 27780, "troposphere ozone" to "tropospheric ozone".
Done.

6. Line 4 of page 27782, "in on-shore SO_2 concentrations" to "of on-shore SO_2 concentrations".
Done.

7. Line 9 of page 27782, "impacts of O_3" to "impacts on O_3"
Done.

8. Line 12 of page 27782, "model resolution" to "model resolutions".
Done.

9. Line 24 of 27783, why Fig. 2b is mentioned before Fig. 1?
We switched the order, showing site locations and flight paths in Figure 1.

10. Line 8 of 27789, add "for" after "Eq. (1)".

Done.

11. In line 32 of page 27793, Fig. 7 is shown here without explanation of the case "12 km TR", which should be at least described in the caption of the figure.
We have modified the figure with clearer legend.

12. Section 3.5, I feel that the long-range transport events should be moved to the end of the results part.
We concluded that local emissions had major impacts on surface SO_x levels after discussing the source identifications and long-range transport episodes.

13. Section 3.7 also discuss the maritime NOx emissions on ozone, but not mentioned in the title.
We modified the title.

14. In line 16-17 of page 27799, the sentence should be re-worded.
Removed.

References

G. R. Carmichael, D. Streets, G. Calori, M. Amann, M. Jacobson, J. Hansen, and H. Ueda, Changing Trends in Sulfur Emissions in Asia: Implications for Acid Deposition, Air Pollution, and Climate, Environ. Science & Tech., 36:4707-4113, 2002.

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.

Sarath K. Guttikunda, Gregory R. Carmichael, Giuseppe Calori, Christina Eck, Jung-Hun Woo, The Contribution of Megacities to Regional Sulfur Pollution in Aisa, Atmospheric Environment, 37(1), 11-22, 2003.

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.