Dear Editor:

Thank you for your feedback. We have made relevant changes to our manuscript to address all the comments from you and the two reviewers.

All of the responses have led to changes in the manuscript text and we have highlighted the specific changes in the responses below that are preceded by the corresponding comments from you and the reviewers for clarity. Changes to the manuscript text in response to the comments are indicated by "yellow" highlighted text.

Editor Decision: Reconsider after minor revisions (Editor review) (26 Nov 2014) by Jason West

Comments to the Author:

The two reviewers reviewed the paper again, and were not completely satisfied with your responses to earlier comments. Please revise to address their comments below.

In addition, I would like to follow up on one comment made earlier:

For the comparison with observations, you conclude "These studies indicate that the simulated transport patterns are able to capture the main important features of the hemispheric flows impacting Central Asia ...". It seems that there is reasonable agreement with the point observations available. But it is not clear to me that you can conclude on the basis of a few point measurements that the model simulates "transport patterns" or "hemispheric flows" correctly. Please comment and revise.

Authors' Response: *We have modified the text to leave the last part of the statement out and reflect the newly added MODIS and AERONET AOD analysis. The updated manuscript text is quoted below.*

"The evaluation of this model framework in other regions outside CA including arctic region and continental US (described earlier in Sect. 2.5) along with the comparison of regional distribution and temporal variability in simulated AOD using corresponding MODIS and AERONET measurements indicates that the predictions of aerosol mass and composition at the hemispheric scales are able to capture important aspects of horizontal gradients and variability, but have considerably higher uncertainties associated with emission estimates (in particular forest fires/biomass burning and natural dust emissions) and wet removal processes (Bates et al., 2006)."

Please find the updated response to the reviewers' comments included here.

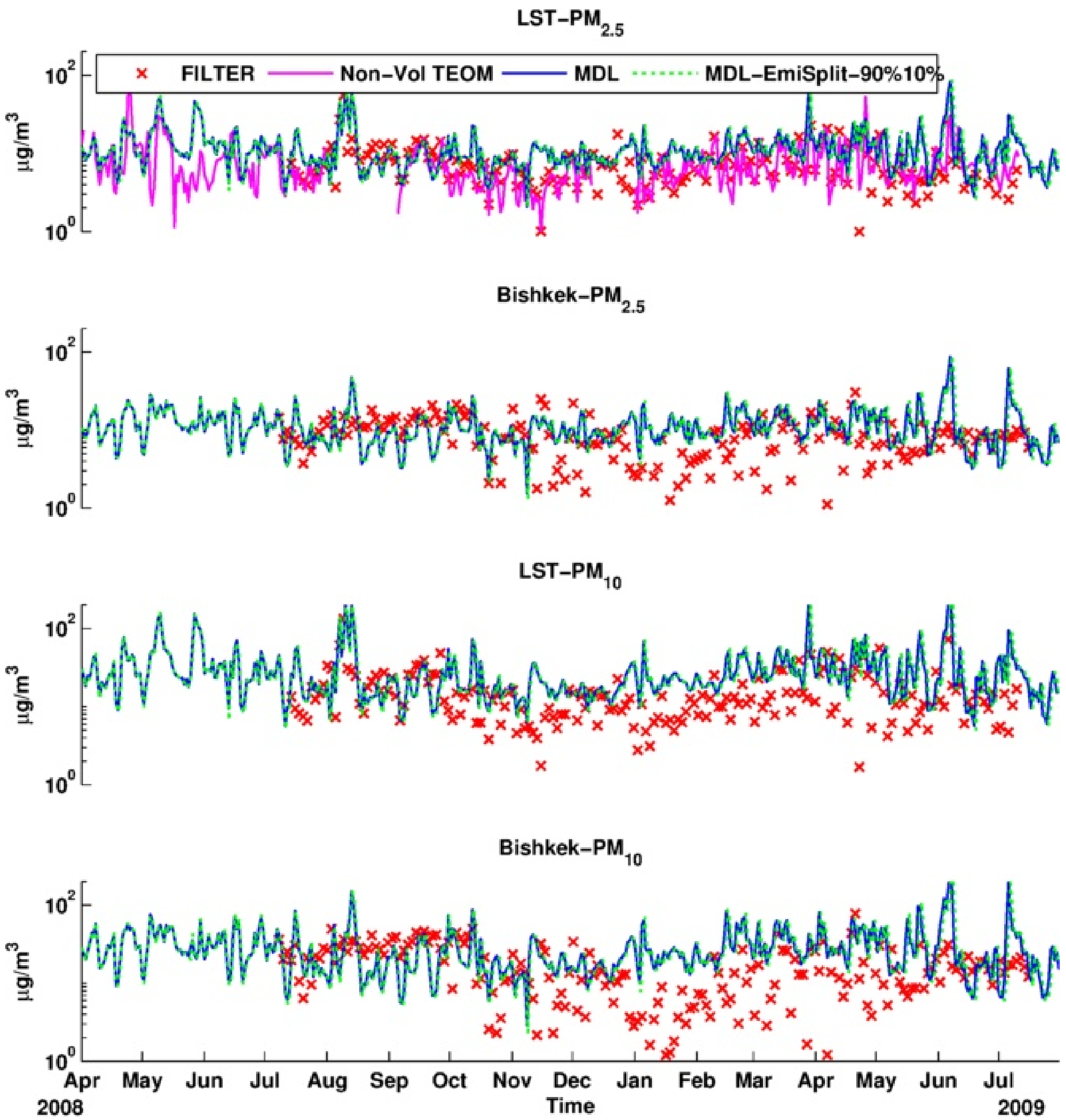
We thank the two anonymous reviewers for their careful review of our manuscript. We have addressed all the issues/concerns raised by reviewers. Below is our response that addresses the specific comments from each of the two reviewers. [Our response (in italicized font) is preceded by the reviewers' comments for clarity]. Changes in the paper text in response to reviewers' comments are indicated by "yellow" highlighted text.

Report #1

Suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)

I still do not understand why 10% residential and transportation emissions were injected into layer 2 and not 100% into layer 1. Presumably, there are no cars/houses sitting around at 100m. So, what specific residential/transportation sources exist at that height? Or what other reasons are there for putting emissions there?

Authors' Response: *We thank the reviewer for raising this point. The residential, transportation and shipping emissions were treated as area source emissions. The shipping sector emissions and elevated residential emissions (i.e. high rise buildings) are often emitted at heights above the surface layer. The height of the layer 1 and layer 2 are set at ~ 30 and 100 m for the modeling domain used in this study. Our intent in putting a fraction of these area source emissions in layer 2 was to recognize that area sources are not just emitted at the surface. To address the reviewer's concern, we conducted a new sensitivity base case model simulation that placed 100% of the residential and transportation sector emissions into layer 1. The comparison of* $PM_{2.5}$ *and* PM_{10} *time series with observed values from this sensitivity simulation at the two Central Asia sites discussed in the manuscript is shown in the following figure. It can be seen that the temporal variability and magnitude of the PM species essentially remain unchanged. Similar behavior was also seen for BC time series, altitude time cross sections at the sites and spatial distribution of the PM species for the domain of interest in this study (not shown here). This suggests that the results are not very sensitive to the exact split for these sectors when using fairly coarse horizontal grids and distributing emissions in the lowest layers. However, the same is not true for power plant plumes and large fires that can emit into higher elevations, for which near source surface concentrations may be more sensitive to the choice of the emission height.*



Report #2

Submitted on 26 Nov 2014

Anonymous Referee #2

Suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)

The authors' effects to address my comments and concerns are greatly appreciated. I still have one concern regarding the issue of proper evaluation of the model. It appears to me that the newly-added discussion of model evaluation is not directly relevant to the domain of interest in this manuscript. The black-dotted domain in Figure 1 clearly indicates the model domain used in the analysis, which is centered in Central Asia and includes almost all of Europe, North Africa, Middle East, China, and parts of India. This domain is where the model evaluation should be focused on, but the revised text does not focus on this domain. For example, the authors' new analysis of evaluating the model with IMPROVE BC observations over the US is not relevant here; their discussion of previous modeling evaluation against the ARCTAS field campaign, although useful, is not directly relevant either. I suggest the authors to focus their discussion of the model's pros and cons on the model domain of interest, which they clearly indicate in Figure 1.

<u>Authors' Response:</u> *We thank the reviewer for this comment. We have moved the discussion of previous modeling evaluation against the ARCTAS field campaign to section 2 on data and methods.*

We have added a model evaluation section pertinent to the model domain of interest in this study in section 3.2 that discusses in detail the comparison of predicted values with observations from the AErosol RObotic NETwork (AERONET) and MODIS AOD along with PM_{10} measurements from the European Monitoring and Evaluation Programme (EMEP) and the Acid Deposition Monitoring Network in East Asia (EANET) surface site networks. Further details of this analysis are added in the response to the reviewer's specific recommendations below.

My specific recommendations are:

1) Given the lack of in situ PM observations in many regions such as China and Middle East, the authors may consider conducting a more in-depth, quantitative comparison of simulated AOD vs MODS AOD over the focused domain. MODIS AOD is used in their analysis (Fig 3), but for reasons not clear to me the authors chose not to provide a detailed comparison of their model AOD with MODIS. They only provide a vague statement that the model captures the 'main observed features' of MODIS (line 389-390, pg 19). Given the satellite-derived AOD has been valuable data to study long-range transport, the comparison can be more quantitative in terms of spatial correlation, regional-specific bias, and seasonality.

<u>Authors' Response:</u> *We have made relevant changes to reflect the comparison with satellite-derived MODIS AOD as sect 3.2.1 in the revised manuscript. We have strengthened this analysis further by including the comparison of the MODIS and simulated AOD values with AERONET measurements, which is quoted below.*

"3.2.1 Comparison with MODIS and AERONET AOD

The predicted period mean AOD spatial distribution is shown in Fig. 3b (using only data from grid cells where MODIS AOD was available. The white colored areas denote regions *where MODIS AOD data was not available). The simulated AOD values capture the main observed spatial features including enhanced AOD over desert regions of Asia including East Asia, West Asia and along the western border of India, Eastern China, Northern India covering the Indo Gangetic Plain and Southeast Asia that are known to have large impacts of anthropogenic and wildfire emissions. However, the period mean AOD values are biased low relative to MODIS AOD over the regions of Northern India and Eastern China. This could be in part related to uncertainty associated with anthropogenic emissions over these regions and the 60km model resolution used in this study. The simulated AOD values are overpredicted relative to MODIS AOD over regions surrounding CA including parts of Eastern Europe, Russia, Northern China, Western Asia and Africa. These biases could be partly attributed to the uncertainty associated with regional transport of dust shown in spatial patterns of simulated dust and* $PM_{2.5}/PM_{10}$ *ratio (Fig. 4b and d).*

The model prediction skills in simulating the temporal and spatial patterns in AOD was evaluated by comparing the predicted daily AOD with the corresponding measured values at 142 sites from the AERONET program (See Table S1 for AERONET site details) located within the domain and for the time period used in this study. We have also compared the AERONET AOD with MODIS on a daily time scale by extracting the daily MODIS retrieved AOD corresponding to the AERONET site locations for the simulation time period. The comparison of the predicted daily AOD with the available MODIS retrievals (n = 29680 using MODIS and simulated AOD extracted at the AERONET site locations, which are paired in space and time) is shown in Fig 3c. The model values show a similar mean value (average simulated and MODIS AOD are 0.24 and 0.31 respectively), with a negative bias and an underprediction in the variability. The comparison of model predictions with respect to AERONET AOD data (n = 22875) shows much closer agreement with mean modeled and AERONET values of 0.21 and 0.23 respectively; (note the comparison is based on paired data for the times only when AERONET data was available so the means are different than the MODIS/model comparison). Also shown is the comparison between MODIS and AERONET (for times/locations with paired AERONET measurements; n = 12719) with AERONET and MODIS mean values of 0.24 and 0.29 respectively. The comparison results of AERONET with respect to MODIS are similar to corresponding values of the MODIS/MODEL comparison, indicating that MODIS retrievals are biased high in the study domain. A more detailed analysis of the AOD comparison by region and season is included in the Supplemental Materials (Table S2 and Figs. S5 and S6).

2) There should be in situ observations of BC and PM over Europe (which is part of the study domain) that the authors can use to evaluate their model, e.g. data shown in Koch et al. 2009 that the authors referred to. The BC evaluation for US should be replaced by an evaluation of BC over Europe.

Authors' Response: *We thank the reviewer for this comment and have made the changes suggested by the reviewer. We have added new analysis that discusses the comparison of simulated PM_{10} with corresponding measurements from the European Monitoring and Evaluation Programme (EMEP available at http://www.nilu.no/projects/ccc/emepdata.html) and the Acid Deposition Monitoring Network in East Asia (EANET available at http://www.eanet.asia/product/index.html) surface site networks.*

We have not included the BC analysis due to very limited amount of BC data available over the study domain and time period.

The PM_{10} analysis discussion included in the text is quoted below.

"3.2.2 Comparison with PM_{10} observations from EMEP and EANET network

We also evaluated the simulated PM_{10} values with monthly mean observed data for the simulation time period from the EANET network over Asia (see Fig. S7). The modeled values are underpredicted as evident from the mean observed and modeled PM_{10} values of 32.2 and 22 μg m^{-3} (n = 314). The normalized mean bias and error are ~ -32% and 44 % respectively. This could be partly attributed to the uncertainty associated with dust emissions that have a significant impact on the EANET site locations (See Table S1 for PM_{10} site locations). We also evaluated the simulated PM_{10} values over Europe using the available monthly mean observations from EMEP for the 2002-2003 time period (See Table S1 for EMEP PM_{10} site locations). The mean observed and modeled values of PM_{10} are 23.9 and 22.2 μg m^{-3} (n = 130). The normalized mean bias and error for the EMEP PM_{10} are -7 and 43 % respectively suggesting an overall underprediction by the model.

The evaluation of this model framework in other regions outside CA including arctic region and continental US (described earlier in Sect. 2.5) along with the comparison of regional distribution and temporal variability in simulated AOD using corresponding MODIS and AERONET measurements indicates that the predictions of aerosol mass and composition at the hemispheric scales are able to capture important aspects of horizontal gradients and variability, but have considerably higher uncertainties associated with emission estimates (in particular forest fires/biomass burning and natural dust emissions) and wet removal processes (Bates et al., 2006)."