Reply to Reviewer 3's Comments

The primary objective of this study is to include a detailed treatment of natural dust emissions in the CMAQ modeling system. The authors have attempted to enhance the representation of three specific processes as they pertain to airborne dust in the CMAQ modeling system: (1) inclusion of algorithms to estimate the emissions of windblown dust emissions (two approaches are discussed), (2) inclusion of representation of heterogeneous reactions on dust particle surface, and (3) updating the treatment of aerosol equilibrium thermodynamics in the model through incorporation of an updated version of the ISORROPIA module. Overall the study is of interest to the wide modeling community and represents enhancements in an aspect of PM source and composition traditionally missing in CMAQ model. In its current form, there are aspects of the manuscript which could benefit from additional work, especially in terms of (a) the description of the implementation details of the dust emission module and (b) streamlining the presentation of results so that the salient features in terms of impacts on model predictions and process characterization are readily apparent to the reader.

Reply: We thank the reviewer for positive comments. We have addressed all the comments. Please see below our point-by-point replies highlighted in bold.

The following suggestions are offered:

1. While two dust emission flux schemes (based on Westphal et al and Zender et al.) are included in the model, it is not readily apparent what the impacts are (if any) on inferences drawn from model predictions. Does one scheme systematically over/under estimate relative to the other? What are the relative impacts on model predictions of fine and coarse PM and how do they differ with the two schemes? If a user was to choose, which scheme should they use?

Reply: As we stated in Sect. 2.1, the major difference between the two schemes is that the Zender scheme splits the dust flux into two components, horizontally saltating mass flux of large particles (Q_s) and vertical mass flux of dust (F_d), whereas the Westphal calculates vertical fluxes directly. The Zender scheme is more physically-based than the Westphal scheme, however, we incorporated both approaches in CMAQ-Dust to assess the sensitivity of dust emissions and their impacts to different dust flux parameterizations. The results indeed show similar spatial patterns and very comparable total dust emissions between the

two schemes. The evaluation results also show the Zender scheme gives an overall better performance than the Westphal scheme. Also in this study, we simplified the splitting of fine and coarse PM by using a ratio of 0.1 for both schemes, due to the lack of information of dust size distributions. We would recommend the Zender scheme, because it is more physically-based and comprehensive and also performs better. We have also indicated this recommendation in Sect. 6.

2. The description of the data sets used in estimating the emissions refer to the BELD3 and the STATSGO data sets. What is the geographic coverage of these data sets – is it just North America? If so how was the relevant information derived for other parts of the modeled domain?
Reply: The BELD3 dataset is for biogenic emissions only and just over North America. The biogenic emissions over other regions were estimated separately. For soil dataset, it should be STATSGO/FAO database, which covers the whole globe. We have modified the corresponding text to reflect the correct data source.

3. What land use information is needed by the two the two dust emission flux schemes and how are these derived? Are they based on the land use scheme used in the WRF simulations – if so which one?
Reply: The land use information needed is just the dominant land use category in each grid cell. The information is taken from the USGS dataset and was gridded to the domain in this study using the WRF Preprocessing System (WPS) utility. This info has been indicated in Sect. 2.1 of the revised manuscript.

4. The fraction of erodible land is determined to be an important factor in regulating the source strength of the estimated dust emissions. While the authors term it an adjustable parameter, it is not apparent how one should go about estimating its value (between 0 and 1)? Does it vary by application? If it represents the fraction of erodible land capable of emitting dust, what is the dependency on the horizontal grid resolution employed by the model? Some additional clarification of these aspects is needed.
Reply: The EF value should be adjusted for different land conditions and episodes. For example, typically higher values should be used for land surfaces that may have a larger

potential for dust emissions. The observed emission fluxes, if available, can be used as a constraint for such an adjustment. However, due to lack of the land surface condition information, the parameter EF acts as a tuning factor and is assumed to be constant everywhere in this study, following Liu and Westphal (2001).   This limitation has been indicated in the conclusion section.

5. The authors present comparison of a number of modeled variables with measurements from a variety of model runs and have clearly put in a lot of effort. However, the presentation of the results is somewhat disjointed and in some instances difficult to follow:

a. The inclusion of the MM5 simulation results in Table 4 appears to be more of an afterthought than something central to this study – which CMAQ simulation was driven by these data?

Reply: The simulation with CMAQv4.4 was driven by MM5, which was used in our early work (see Wang et al., 2009). We included results of MM5/CMAQ v4.4 in this paper to compare them with the results from WRF/CMAQ v4.7 without and with dust.

b. What is the point of including results from the old CMAQv4.4 simulations? A number of aspects of the model appear to have changed relative to v4.7 in which the dust emissions are examined. The MM5 and CMAQv4.4 results are distracting and do not add anything to the primary focus of this study – their discussion only adds unnecessary text and length. I suggest deleting these aspects of the analysis.

Reply: As mentioned above, we included results of MM5/CMAQ v4.4 in this paper to compare them with the results from WRF/CMAQ v4.7 without and with dust (see Tables 6-8). We also compared the enhancement factors shown in Fig. 11 from CMAQ-Dust to those by MM5/CMAQ v4.4 of Wang et al. (2009). Such a comparison can show clearly the improvement of default CMAQ v4.7 against default CMAQ v4.4 due to changes that are not related to dust and additional improvements due to the addition of dust treatments in CMAQ-DUST as compared with default CMAQ 4.7.   For example, compared with MM5/CMAQv4.4, the biases in the predictions of O_3, $PM_{2.5}$, and AOD are generally reduced, which are due to updates in CMAQ v4.7 and the use of a different meteorological model. Adding dust treatments, CMAQ_DUST further improves AOD and did not affect O_3 performance, although $PM_{2.5}$ predictions are worse at some networks. The inclusion of

such comparison would (1) help separate the impact of dust-related processes from the impact of non-dust related processes in CMAQ_DUST, as compared with default versions of CMAQ v4.7 and CMAQ v4.4; (2) record the model performance to reflect the history of the model improvement, which would benefit the scientific community. In particular, despite the release of newer versions of CMAQ, there remain some users for MM5/CMAQ v4.4, who would like to see the performance comparison between WRF/CMAQ v4.7 and MM5/CMAQ v4.4.

c. What impact did the inclusion of K, Ca, and Mg (via ISORROPIA II) have on the partitioning of oxidized and reduced nitrogen between the gas and the aerosol phase? The CRUST_ONLY simulation appears to have been designed to examine this issue –however the discussion of results does not clearly demonstrate the effects.

Reply: The impacts of including crustal species have been discussed mainly in Sect. 5.1. The effects include decreasing fine-mode NH_4^+ throughout the domain, increasing fine-mode NO_3^- over dust source regions and decreasing it over downwind heavily-polluted areas, and shifting NO_3^- from fine-mode to coarse-mode. These effects are also summarized in Sect. 6 in the revised manuscript.

d. Section 4.2.2 presents a lot of information on model performance statistics but little interpretation from a model process standpoint – there is useful information in this section as it pertains to the dust model and its impacts on model predictions but is obscured in the current presentation of the results. The authors should consider restructuring this section by presenting only the statistical comparisons for the relevant species and simulations. Given the model process enhancements included in the simulations, did the authors realistically expect to see differences in predicted CO and TOR between the various simulations? Would global statistics such as NMB capture any such possible changes?

Reply: We have revised the presentation of material in Sect. 4.2.2 to make it more readable. We also added more evaluation results of the two dust schemes by using the measurements of surface dust concentrations and also daily-average AOD near the dust source regions from AERONET. Regarding the column evaluation of CO and TOR, we did expect some differences for TOR, due to the surface uptake of O_3 by dust particles. For CO, it has been

widely used as a tracer species for long range transport study. We think that the evaluation of column CO will facilitate the discussion of Sect. 5.4 in our manuscript, since an accurate simulation of CO typically indicates a realistic long-range transport pattern simulated by the model, and thus more accurate impacts of long-range transport. We have also shown the spatial distribution of those species in the supplementary materials to complement the domain-wide stats such as NMBs in the table.

e. Page 13480, line 15: what does gas phase NO3- represent? Do the authors imply nitrate radical?
Reply: It was a typo and should be gas-phase NH_3.

f. Page 13481, lines 10-15: it is difficult to follow this discussion without seeing the figure (not included) that the authors are referring to. Line 18: "and replaces NO3- as ions: : :" is awkward – the discussion needs to be reworded.
Reply: The figure for HNO_3 has been added in the supplementary material. An example is given in the sentence in the first paragraph of Sect. 5.2.

g. Page 13484: lines 5-10: Are the reported increases in O3 and CO of 3.6% and 2.1%, respectively in "background" levels or total simulated levels – the authors need to be careful in the terminology.
Reply: It should be total simulated levels, which has been indicated in the revised manuscript.

h. Page 13484: lines 11-15: what results in the "negative contribution" of Asian NOx emissions to NOx levels in the US? From a process stand point how can this be attributed to a possible negligible import? Is the suggestion that this is a numerical effect?
Reply: The conclusion is based on a previous study (Wang et al., 2009), which found that the direct long-range transport of NO_x to the US is negligible. So the negative change of NO_x as a result of the removal of Asian anthropogenic emissions is not due to the transport itself but to the differences in the rates of chemical destruction between the simulations with and without Asian anthropogenic emissions. This point has been added in Sect. 5.4.