## **Response #2**

We appreciate the reviewer's comments and have made efforts to address the concerns listed. The most significant changes involved tightening up the results section and making it easier to follow.

## **Main Comments:**

Page 29783, lines 7-20: This is the fragile point of the method proposed, and it is nice to see it discussed in this version of the manuscript. Since GOCART defines the regions where a given aerosol type is expected to dominate, results are likely to be GOCART-dependent. The question is how much so? The authors have yet to prove that using another model would yield similar regions (line 17). Since daily data from other models may not be readily available, I would suggest trying to vary the threshold used to define the regions, and assess the impact on results.

We agree that employing these methods on additional aerosol speciation models would have been a very useful endeavor. Unfortunately, the required model output could not be acquired and processed prior to the primary author's departure from UAH. We did study the effect of varying the thresholds on the GOCART model and noted some interesting results. The figure below shows MODIS FMF as a function of threshold in percent. As the threshold increases, the average FMF trends closer to expected values for each aerosol type. Ideally, a threshold of greater than 90% would be used, but sample size at this level was too small. Thus, the 80% threshold was used by this research. The effect of this difference is noted in the text.



Page 29789, line 21: Since the study is limited to oceans, it is expected than all regions have a sea-salt contribution to both the AOD and FMF, as acknowledged by the authors. It is thus unlikely that any region will ever correspond to a "pure" aerosol type (assuming such a thing

exists outside of numerical models). So the authors try too hard in explaining differences between the different datasets: they are simply looking at different aerosols, where one type may dominate if GOCART-defined regions can be relied upon. In the end, the FMF values given by the study in Table 1 are unlikely to be "more correct" than those in previous studies shown in the same table - they are just different. And a future study using other models and satellite retrievals will end up with yet another set of different results, which will not be wrong either... In addition, GOCART-defined regions are small: at such small scales, the variability is expected to be large.

We now note that the differences observed are also in part due to the changes in spatial and temporal sampling between data sets. We agree that our results are not necessary more or less "correct" that those presented previously, and this is noted. However, we also believe that some of these differences fall beyond that which can be explained by simple changes in spatial or temporal sampling. This was one of the reasons behind performing this analysis using data from Terra and Aqua at two different resolutions.

Section 4.2 brings more information and using the AI seems to be improving the sampling of SU and CC aerosol types. I would therefore have organized the paper differently, presenting the most powerful method (using the AI) and its results first, then making a sensibility study where the AI is not used.

Yes, we agree that OMI-AI proved to be a very useful tool in this research. However, we decided to make MODIS FMF the focus given that we were interested in direct comparisons with previous MODIS-based research. Given this focus, we elected to discuss the OMI data in terms of value-added information over that available from MODIS. Whether one or the other method is more powerful can be debated. For example, OMI-AI cannot readily distinguish between absorbing BC and DU without taking into account geographical or other ancillary information. Thus, we believe showing a combined approach is best.

Section 4.3 on vertical distribution is purely based on unvalidated GOCART vertical profiles, and there are no satellite data to discuss. It brings nothing to the manuscript and should be removed.

Based on your and another reviewer's recommendation, this section and references to it have been removed.

## **Minor Comments:**

Page 29764, line 17: "the use of daily GOCART simulations improves our confidence in the results compared to monthly analysis". It remains to demonstrate that GOCART is able to get correct daily aerosol distributions. Is the validation of GOCART AODs made on a daily basis, or a monthly basis?

The validation we are aware of was performed using daily data. However, we elected to remove this statement from the manuscript as part of the revisions

Page 29776, lines 6-8: Writing that "Their results clearly show anthropogenic aerosols are mostly fine mode" is incorrect. This is not a result of these studies, this is an assumption: by assuming that anthropogenic aerosols have low FMF, studies manage to identify aerosols as being anthropogenic. Not the other way around.

This statement has been corrected.

Page 29780, line 22 and Page 29782, line 26: If OC has a natural component due to natural biomass burning events, then BC must also have a natural component from the same sources, since the two species are co-emitted.

We agree with the reviewer in principle; however, GOCART assumes that the natural component of BC = 0 no matter what the natural OC component is. We have now explicitly stated this in the next.

Page 29781, line 22: While it is true that DU and SS aerosols exhibit more diversity in the AeroCom intercomparison than for other species, it remains that the overall diversity is large. As shown in Figure 3b of Textor et al. (2006), diversity on total dry mass is of the order of 30species.

We have made revisions to note that variations among anthropogenic aerosol species from model-to-model remain significant.

Page 29789, line 5: "Carbonaceous aerosols" would be a better term than "carbonate", as carbonate is not necessarily dominant in the chemical composition. Change made

Page 29793, line 7: "increase in biomass burning in Central Africa". Increase compared to what reference?

This statement has been removed.

Page 29793, line 13: "Note that GOCART considers DU as surface source". The authors make it sound like a simplification. Obviously mineral dust aerosols originate from the surface.

This is true, we were pretty much restating the obvious. This statement has been removed.