Anonymous Referee #2

This paper presents an analysis of a global chemical transport model's predictions of organic aerosol, through comparison with data from many AMS across the globe. Although it is encouraging to see these AMS data being used, I found the analysis in this paper disappointing, and in the end I cannot see that we learn very much. I cannot recommend this paper for publication for the following main reasons:

We thank the reviewer for his/her thorough review. The VBS approach used in this work is a promising approach for modeling OA in global scales that is only used by a rather limited number of models and global studies (Jathar et al., 2011; Tsimpidi et al., 2014; Shrivastava et al., 2015; Hodzic et al., 2015). In addition, most of the global studies have used only total OA measurements to evaluate their model performance regarding OA (Tsigaridis et al., 2014). Here we use for a first time an extensive global dataset of AMS measurements and factor-analysis results from 84 field campaigns in the Northern Hemisphere during the period of 2001-2010. Collecting this high density of AMS data allowed us to take advantage of the spatial and temporal variations of the measured HOA and OOA in order to evaluate our model's temporal and geographical variability of POA and SOA. Through this integrated effort we estimate the contribution of the fresh emitted particles and the chemically processed organic material from combustion sources to the total OA load during different seasons, types of environment and continents. In addition, we present a first attempt to simulate the SV-OOA and LV-OOA identified by the PMF analysis. We test the hypothesis that SV-OOA corresponds to simulated fresh SOA and LV-OOA corresponds to the aged-SOA. Based on this assumption, EMAC is able to reproduce the PMF results predicting a dominance of aged SOA during all seasons and environments (except North America). This is the first time that a global model tracks separately the freshly formed and aged SOA and is in line to the latest advancement in PMF analysis. For all the above reasons we do believe that the present study merits publication since it can pave the way for the improved representation of OA in global models. Please see below our point by point response to the individual comments.

*1. Much of the focus is on differences between urban, urban-downwind and rural OA components, but the horizontal resolution of the model used, 1.875 * 1.875 degrees (ca. 200 km) is wholly inappropriate for analysis down to the urban scale. Even Mexico City has dimensions of 10s of km, not 100s km. The authors admit on p18:'… the model cannot reproduce the concentrations of POA and SOA over urban locations due to its coarse resolution'. They therefore chose not to use urban results when looking at seasonal patterns. However, if the urban results are not valid for seasonal comparisons they are clearly not valid for annual comparisons either! (I also wonder why such a low resolution was used. This model has previously been run at higher resolutions. Presumably CPU time was not a big issue, since we are looking at the results of just one model run. For the results of sensitivity tests on IVOC/POA ratios and deposition parameterization we are referred to papers which are yet to be written.)*

We do agree with the reviewer that the resolution used in the current application as well as in most global model applications (see for example the use of 32 global models in Tsigaridis et al. (2014)) introduces potentially significant errors over urban areas. Our analysis and use of the corresponding urban AMS datasets should be viewed as an effort to quantify the magnitude of the errors. There have been a number of recent studies using global atmospheric chemistry models to investigate the link between premature mortality and atmospheric aerosols in urban and rural environments (Lelieveld et al., 2015). Evaluating these global models over urban locations provides therefore very useful information about their potential biases in these locations. For example, the above global health studies usually provide a lower limit of the actual contribution of atmospheric aerosols to premature mortality over large urban areas. The present study can provide an estimate of the degree of the underestimation of these global aerosol-health studies.

Please note that the resolution is not the only potential issue. For instance, global models, including ours, lack OA emissions from residential and commercial cooking activities (Tsigaridis et al., 2014). However, cooking OA is an important source of OA that can contribute significantly to measured POA (around 50%) and total OA (15%-20%) over urban areas (Sun et al., 2011; Mohr et al., 2012; Ge et al., 2012; Hayes et al., 2013). While local emissions of primary aerosols may not be captured accurately with global models, this is less critical for secondary aerosols as they are formed during transport. For example, the urban "increment" in PM2.5, comparing background with urban air, is typically small.

To avoid misunderstandings about our intensions (we do not expect a global model to reproduce urban observations), in the revised paper we discuss the urban comparisons in a different section (Section 5.1). We focus on the magnitude and direction of errors in these areas. We also discuss there the effects of missing urban aerosol sources like cooking. Urban locations are not included for the seasonal, continental (added during the revision) and total (annual) evaluation of the model and urban statistics are now provided in a separate table (Table 2). Finally, following the recommendation of the reviewer we have deleted any reference to unpublished work.

2. Essentially the whole paper builds upon global averages of OA results, and we are given no idea of how the model performs in different parts of the world. Global averaging can mask a whole slew of incorrect regional results, and with a pollutant as complex as OA this is a dangerous procedure. As can be seen in the (originally not-mentioned!) papers (Karydis et al., Pozzer et al.) where some model evaluation can be found for the inorganic components of this model, the model displays strong under and over-predictions of species such as sulfate and nitrate in different parts of the globe. I comment more below on the author's inattention to model evaluation (and their response to my original criticism of this), but although Tables S1-S3 now give the site by site comparisons, it is a daunting job for the reader to work out the spatial patters in model under or overestimation. How does the model perform in North America, Europe and Asia? I would have liked to see global bias maps (of at least total OA, ideally components) compared for the different types of stations. Ideally with separate presentation for summer and winter months, since the biomass burning signal is so large. Given that different seasons typically have very different sources of SOA, I would have liked to see maps (or mapped pie-charts or similar, as per Jimenez et al., 2009) of model bias and correlation for these different seasons.

We adopted the reviewer's suggestion and evaluated our model performance over the different continents separately. We have added the corresponding statistical analysis over Europe, North America, and Asia in Tables 3-6 of the revised manuscript. We have also added two figures (Figures 11 and 12 of the revised manuscript) showing the comparison of the model predictions against the AMS POA and OOA for each dataset over Europe, Asia, and North America. Unfortunately, we could not present this comparison on a map since our datasets are more than double the size of those used in Jimenez et al. (2009). However, we believe that the two new figures added in the manuscript clearly depict the model performance over the different continents. Discussion related to the model performance over the different continents has been added in Section 5.2.

3. I was also surprised to see no mention of, let alone comparison with, 14C data. For example, Hodzic et al. (2010) used published results from measurements to provide a comprehensive view on the fraction of fossil and biogenic carbon across the globe, and in many cases for both summer and winter periods. Such data would have provided a very good check on some of the model predictions presented here.

Unfortunately, our model cannot provide information about the fraction of fossil and non-fossil carbon on OA. The emission inventory used in this study uses only one lumped type for anthropogenic emissions from every source sector that includes both fossil (e.g., from the power sector) and non-fossil (e.g., from residential wood burning) OC. This is the reason why even for the evaluation against the AMS data we compare the sum of the simulated fPOA (anthropogenic) and bbPOA (open biomass burning) against the sum of the AMS HOA and BBOA.

4. I was not convinced by several aspects of the author's reply to my query concerning evaluation. I give here the authors comments in quote, and my replies in normal text:
"In our recent paper (Karydis et al., 2016) we have found that EMAC tends to overestimate the inorganic aerosol components over Europe while over North America and Eastern Asia, the inorganic aerosol concentrations are slightly underestimated by the model".

Well, Karydis suggests a factor 3 over-prediction of nitrate and sulfate in Europe, and a factor 2-3 under-prediction in East Asia, which is more than 'slightly' underestimated. Although such discrepancies are probably not surprising for global models, their magnitude raised serious questions about the interpretation of much more complex OA comparisons.

"Given that the performance of the model is increasingly realistic with increasing resolution, the acceptable model performance shown from all the previous model evaluations (in both higher and lower spatial resolutions than the one used in our work) should be enough to verify that EMAC can cope with the inorganic components under any resolution."

I completely disagree with this sentence. As noted above, one model setup can produce factor of three over-predictions and factor two under-predictions at the same time. It is quite likely that changing resolution might help some problem areas but exacerbate others. If for example there are problems with vertical dispersion in the model (quite likely with any model), then a change in horizontal resolution can easily cause incorrect responses. Increasing resolution can also expose other problems which lower resolutions might mask out, for example if the emissions are incorrectly distributed.

"Furthermore, there is no chemical or thermodynamic link between the inorganic (GMXe) and organic (ORACLE) aerosol modules used in this study. Therefore, we strongly believe that a comprehensive evaluation of inorganic components in our manuscript will disorientate the reader without providing him/her any useful information regarding the behavior of the OA in our model system."
Well, there are clear links between the inorganic and organic pollutants, which include meteorology (including dispersion issues, mixing heights, etc.), emissions, and atmospheric chemistry (OH, O3, NO3). If EMAC is under-predicting e.g. NO2 in an urban area, I would expect it to under-predict OA too, especially near-source POA levels.

In respect to the reviewer's comment we want to clarify that our initial response not to include the inorganic evaluation in this manuscript was not a matter of "inattention" but it was based on the fact that the model performance with regards to inorganic aerosol components and the major gas-phase species was thoroughly documented in Karydis et al. (2016) and Pozzer et al. (2012a; 2012b). Although we do agree with the reviewer that this comparison can help someone to identify errors in meteorology or transport that can affect the OA performance as well, at the same time it can be confusing given that other uncertainties regarding the emission of these species and their chemistry (which may not be the same with the organics) can drive their performance. However, following the reviewer's concern we have included the evaluation of the inorganic components of our model in the supplementary material.

5. The discussions in e.g. 5.2 explain any discrepancies solely in terms of SOA mechanisms or rather particular emissions problems (e.g. cold start). There are many reasons why modeled and measured OA components don't match, including for example dispersion or deposition processes, which is why I don't think we learn very much from rather broad-brush OA comparisons alone.

The EMAC model has been extensively described and evaluated against in situ observations and satellite measurements that include filter-based particulate matter concentrations, aerosol optical depth, acid deposition, gas-phase mixing ratios, and meteorological parameters (Jöckel et al., 2006; Pozzer et al., 2012a; pozzer et al., 2012b; Karydis et al., 2016). Therefore, we expect that processes like dispersion and deposition are well simulated by the model. Furthermore, these processes affect the other aerosol species as well. However, e.g., over Europe, the model underestimates both POA and SOA. In contrast, the model overestimates inorganic components over Europe (e.g., NO_2 and nitrate which are expected to correlate with POA and SOA, respectively; Table S2). Therefore, the modelled underestimation of SOA and POA should be attributed to processes that affect only OA formation. In Section 5.3 of the revised paper we now discuss possible discrepancies and omissions in chemistry and emissions that should be considered and further explored in future studies

6. This paper focuses on the primary emissions, with presumably other papers to come addressing biogenic and such like. One of the biggest assumptions with POA is the factor 1.5 used to get IVOC emissions. This factor is very uncertain, and I would have expected a paper concerned with POA to address this uncertainty directly. Instead, we read on p.10 (L277-278) that this issue will be addressed in another paper which is in preparation. This is surprising, and again makes me wonder what this paper is actually showing, apart from largely unverified model predictions.

The VBS framework used in this study, is considered a state-of-the-art approach for modeling OA, however, there are several uncertainties in the parameters used by the corresponding models. These parameters include the emissions of LVOCs, SVOCs and especially IVOCs, the aging chemistry, etc. which all impact the predicted OA concentration and composition. Given the wide range of uncertain parameters a detailed sensitivity study is required to quantify the impact of these uncertainties on the simulated OA and to better constrain the model using the available observations. Such a study is outside the scope of the present work but it is something that we are currently working on. To avoid confusion we have deleted any reference to it in the revised manuscript.

7. There is always the danger with SOA modeling studies that a lot of colorful maps are presented, and various numbers given, but the reader never knows if these results are meaningful. We have seen global modeling papers for OA for about two decades now doing this, often with claims that 'this time' the results are reasonable. With time such results change markedly though as new understanding becomes available, often with dramatic results. For example, the recent paper by Hodzic et al. (ACPD, 2015) is suggesting that SOA processing and losses should be much faster than previously assumed. With AMS data one has the possibility to try to pin down model performance in a much more robust way that was previously done with filter measurements. By ignoring the spatial and temporal variations seen in these AMS data the current paper missed a real chance to show how well their model performed, and thus how seriously we should take the results.

We have done our best to take advantage of the spatial and temporal coverage as well as the chemical resolution of the AMS observations. In order to determine the effect that the site type, the site geographical location (added in the revised manuscript), and the seasonal cycle has on the performance of the model, we evaluated our model performance separately for urban, urban-downwind, and rural sites; European, North American, and Asian sites; and for the four seasons of the year (winter, spring, summer, and autumn). In addition, we present a first attempt to simulate the SV-OOA and LV-OOA identified by the PMF analysis. We test the hypothesis that SV-OOA corresponds to simulated fresh-SOA and LV-OOA to the aged-SOA. To date, global studies use total OA measurements to evaluate their model performance (Tsigaridis et al., 2014). Here we use for a first time an extensive global dataset of AMS measurements and factor-analysis results from 84 field campaigns in the Northern Hemisphere during the period of 2001-2010. We have concluded that EMAC is able to reproduce the observed dominance of aged SOA during all seasons and environments (with the exception of North America).

Other points:
1. General: The paper makes use a large fraction of older references. Although I like to see citation of original or ground-breaking papers, I also expect to see that the authors know about and have incorporated recent advances in their work. The cited references throughout the paper need to checked and updated where possible. (I give some examples below, but in general I worry when literature is more than 10 years old in this field.)

Following the reviewer's recommendation we have updated the citations used throughout the revised manuscript whenever necessary.

2. Although the focus of this paper is on anthropogenic SOA, the treatment of biogenic VOC is rather confusing. On p9 for example we read that the production of RO2 after oxidation of SVOC and IVOC has been omitted because the OH-sink is so low. What about forested regions though, where there are plenty of papers showing that the OH sink from BVOC can be very significant indeed (Kurpius & Goldstein, 2003, Mogensen et al., 2011)? The Tsimpidi et al. (2014) paper which introduced ORACLE has reaction R3 which also seem to ignore RO2.

For both anthropogenic and biogenic VOCs the gas-phase chemistry mechanism MECCA (Sander et al., 2011) was used which includes the production of RO_2 after their oxidation. However, SVOCs and IVOCs are not part of this chemical mechanism and similar to other large scale models (e.g., Robinson et al., 2007; Grieshop et al., 2009) we used a more simplified chemistry omitting the production of RO_2.

3. p2, L42-43: Technically, aerosol is a mixture of gas and particle, and some authors use the term POA to include both phases, so it is good if the authors state clearly at the beginning that for them POA means the particulate fraction of primary OA.

We have added this information in the revised manuscript.

4. p3, L46. Please be explicit. You mean particulate OC, not gaseous VOC species.

Yes, we refer to particulate OC.

5. p3, L46-50. Again, old literature (>10 years) is used. These numbers need to be updated, e.g. with those from Tsigaridis et al. 2014.

Tsigaridis et al. (2014) did not provide any information on the amount of OC emissions from anthropogenic and open biomass burning sources. Instead, we have updated our literature by using the values reported by the CMIP5 (for anthropogenic sources) and GFED3 (for open biomass burning) emission inventories. These emissions are adopted by most of the available OA global models as seen in Tsigaridis et al. (2014).

6. p3, L50. What is the reference for the 13-57 Tg/yr?

It is Bond et al. (2004). However, following the previous comment of the reviewer, these values have been replaced by the values reported in Van der Werf et al. (2010).

7. p3, L53: May et al., 2014, and/or Ranjan et al. (2012) could also be cited here for a more recent evaluation.

We have updated the reference to "(Robinson et al., 2010; Ranjan et al., 2012; May et al., 2014)"

8. p3, L564 Again, only old references are used (>14 years!). There has been a lot of work and progress (even 'rethinking') since these Schauer evaluations.

We have updated our reference to "(Shrivastava et al., 2008; Robinson et al., 2010).

9. p4, L109. It is not true that OOA has the same diurnal pattern as other secondary organic PM components. Indeed, not even sulfate and nitrate have the same pattern as each other. The authors even mention later in the manuscript that one can usually observe different diurnal variations for SV-OOA and LV-OOA.

We agree with the reviewer that OOA does not always have the same diurnal profile as other secondary PM components (Crippa et al., 2014). However, OOA often correlates with some (not all) of the inorganic secondary species (e.g., sulfate) which allow us to interpret it as SOA (Jimenez et al., 2009). For instance, LV-OOA is expected to correlate with sulfate and SV-OOA with nitrate (Ng et al., 2011). We have updated the text to make clear that OOA correlation with secondary inorganic species might not be always very high. We also mention that SV-OOA and LV-OOA are expected to correlate with nitrate and sulfate, respectively.

10. p5, L120-122. These statements need to be updated, e.g. Tsigaridis et al. 2014 show that this commonly repeated statement that CTMs underestimate OA is not always true.

We have updated the text following the findings of Tsigaridis et al. (2014) and we now state that global models systematically underpredict OA over and downwind of the urban centers.

11. p5, L131-132. Donahue didn't 'assume' that emissions were distributed in bins. The bins are a modeling approach applied to what he knew was a continuous distribution of volatilities.

We have rephrased the text to "…Donahue et al. (2006) developed the volatility basis set (VBS) framework which assumes that POA emissions are semivolatile and photochemically reactive and uses logarithmically spaced volatility bins to distribute POA upon emission."

12. p5, L141 on. Again, the Tsigaridis paper is the current best source of information on global SOA models.

In this part of the text we refer to global models that have adopted the VBS approach. Tsigaridis et al. (2014) model intercomparison included only one model with the VBS approach (Jathar et al., 2011). Here we cite three more global models (Tsimpidi et al., 2014; Shrivastava et al., 2015; Hodzic et al., 2015) that have been published after the Tsigaridis et al. (2014) paper and use the VBS approach. Note that the Shrivastava et al. (2015) and Hodzic et al. (2015) studies are discussed in the revised version of the manuscript.

13. p6, Sect. 2.1. As mentioned in my earlier ACPD comment, I would have expected a model description to point readers towards works that could prove the validity (or *at least document performance) of any model. This manuscript is remarkably free of the kind of information on model performance (also for inorganic components) that I expect to see in any model study.*

EMAC has been extensively described and evaluated against ground-based and satellite observations (Pozzer et al., 2012a; Pozzer et al., 2012b; Karydis et al., 2016). This information has been added to the text. Furthermore, we have performed the evaluation of our model results for the inorganic components against ground based measurements over N. America, Europe, and Asia. The results of this evaluation have been added to our supplementary material.

14. p7 - L204. The sentence about LVOC not being allowed to participate in photochemical reactions begs the questions of how SVOC or indeed any semi-volatile compounds are allowed to participate, and how. Do SVOC and IVOC deplete oxidant concentrations, and if so, what happens to products and e.g. RO2 formation?

The formation of SOA from the photo-oxidation of SVOCs and IVOCs are described by the reactions R3-R8 of the manuscript. The volatilities of SVOCs and IVOCs are reduced by a factor of 10^2 as a result of the OH reaction with a rate constant of 2×10^{-11} cm^3 $molecule^{-1}$ s^{-1} and a 15% increase in mass is assumed to account for two added oxygen atoms (Tsimpidi et al., 2014). The production of RO2 as an intermediate after the oxidation of SVOC and IVOC has been omitted since it would be essential only in cases where these reactions are a potentially significant sink of OH (i.e. in concentrated smoke plumes) (Alvarado et al., 2015). All the above information is now present in the revised manuscript.

15. p8, L211-216. The authors claim that a reduction in volatility by a factor of 100 is common in global models, but cite just their own code and one other paper (Pye and Seinfeld, 2010). Can the authors justify the word 'common'? In fact, this reduction of a factor of 100 for SVOCs and IVOCs upon oxidation is rather extreme. Although I know this has been dealt with to some extent in the 2014 paper, another sentence or so of explanation would help the reader of this paper.

We have rephrased this sentence to avoid misunderstandings. A few global models include the oxidation of SVOCs/IVOCs (Pye and Seinfeld, 2010; Jathar et al., 2011; Tsimpidi et al., 2014; Shrivastava et al., 2015; Hodzic et al., 2015), therefore, the word "common" cannot be justified. Other global and regional studies which assume two orders of magnitude reduction in volatility include those by Grieshop et al. (2009), Hodzic et al. (2010) and Pye and Seinfeld (2010). Shrivastava et al. (2011) even used seven orders of magnitude reduction in volatility per reaction. However, as we state in the manuscript, despite the fact that most of the studies assume that each oxidation reaction of SVOC and IVOC reduces the volatility of the precursor by one to two orders of magnitude, the particle-phase oxidation products can be up to four orders of magnitude lower in volatility than the precursor (Kroll and Seinfeld, 2008).

16. p8, L215. The statement about regional models and the following sentence miss an important point. The regional models have usually followed the suggestions of the groups that developed the VBS ideas (Donahue, Robinson, etc.), and who worked directly with smog-chamber data. As noted in Donahue et al. (2009), the phase *partitioning of a complex mixture can be accurately described by lumping material into volatility bins separated by an order of magnitude (at 298 K). This is not only convenient; it is a nearly optimal distribution. The cruder methodology (bins spaced by two orders of magnitude in C*) being suggested in the ORACLE scheme may well be sufficient for global models given the large uncertainties of any SOA scheme, but is not optimal. Indeed, a lot of atmospheric 'action' may be expected to take place over quite a narrow range of concentrations between say 1 and 10 ug/m3, a range which isn't resolved by the factor 100 spacing of ORACLE.*

The discussion at page 8 is related to the SOA aging reactions and to the assumed reduction in SVOC/IVOC volatility after each photochemical oxidation. This is not necessarily related to the volatility resolution used. For instance, Grieshop et al. (2009) assumed two orders of magnitude reduction in the SVOC/IVOC volatility after their oxidation even if they used "one order of magnitude" volatility resolution in their model. Regarding the volatility resolution used in our study, we use 3 volatility bins to describe the atmospherically relevant conditions: $C^*=10^{-2}$ μg m^{-3} for clean areas; $C^*=1$ μg m^{-3} for typical polluted regions; and $C^*=100$ μg m^{-3} for much polluted cases. While we acknowledge that this resolution can introduce some errors in our results, we believe that it is a reasonable compromise for a global model given the spatial resolution used (average OA concentrations over a wide area) and that the uncertainty introduced by other parameters (i.e., the emission factors) are more important. Other studies have even used saturation concentrations that are not logarithmically spaced. For instance, Pye and Seinfeld (2010) used C*=20 and 1646 μg m^{-3} to describe the SVOC and C*=0.2 and 16.46 μg m^{-3} to describe their oxidation products. We now stress in the revised manuscript that the bin resolution used in ORACLE is a compromise between accuracy and computational efficiency.

17. p13, L381. This statement, that the fOA/OA fraction is 0-10% over boreal regions is incorrect. Values seem to be 0.3-0.5 over much of the Eurasian boreal forest. Which is rather surprising, isn't it?

Indeed the east part of the Eurasian boreal forest seems to be an exception since the lower emissions of bbOA together with the considerable amount of fSOA transported from Europe resulted in fOA/OA values of ~40%. This information has been added in the revised text.

18. p17, L517. The Denier van der Gon 2014 ACPD paper made it to ACP in 2015.

We have updated the citation in the revised manuscript.

19. p23. Why would aqueous-phase and heterogeneous oxidation be important in winter but not summer? Provide some citation to back up such statements. I would also suspect that the factor 100 spacing between volatility bins would have some implications for summer/winter differences in model performance.

Such processes should be taking place in both seasons. However, during the photochemically active periods there are other chemical pathways (e.g., reactions with

OH and ozone) to convert the organic precursors to SOA. Adding to this the increased presence of lower-level clouds during winter and early spring compared to summer in North Hemisphere mid-latitudes (Stubenrauch et al., 2006), one would expect a higher importance of heterogeneous oxidation in winter. This is still speculative though so we have rephrased the corresponding sentence making this clear.

20. p34, Table 1. Add BSOA also.

We have added in Table 1 the tropospheric burdens of anthropogenic (aSOA) and biogenic (bSOA) SOA from the oxidation of VOCs.

21. p35, Table 2. The caption mentions 84 stations, but the table lists 82 in the upper rows and 60 in the lower. Explain.

We thank the reviewer for pointing this out. We had omitted to update the tables after the inclusion of the two final AMS datasets. In the revised manuscript, we updated the number of datasets and their statistics in tables since we used 23 urban datasets, 15 urban-downwind, and 46 rural (total of 84 datasets). The lower rows sum to 61 since we have not included the urban datasets in the seasonal and total (annual) evaluation of our model. In the revised manuscript we provide the urban statistics in a separate table so as to avoid any confusion since urban locations are excluded from the seasonal and continental (added during the revision) evaluations.

22. Figs: The quality of the Figures should be improved. In Fig. 4 the color scale leaves almost the whole map as different hues of blue. Also, although acroread shows Figs. 1 and 2 fine, my print-out has black-backgrounds and is almost unreadable. I suggest a change of Figure format for this.

Since the initial submission of the manuscript in ACPD, we have updated the color scale used in the global-map figures using a more detailed color scheme for the low values. However, SOA concentrations are uniformly low over a large part of the continents; therefore, we cannot avoid using hues of blue over these areas. We have also changed the format of Figures 1 and 2. They are clearly printed in our experience. Even if this is not the case for the reviewer, during the file submission for production of the manuscript the figures will be submitted individually as .eps files and not using the MS Word format that probably cause the problem here.

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