Anonymous Referee #1

Overall, the article is well written and provides a useful comparison of a global model to a very large AMS data set. The agreement between fresh OA and SV-OOA and aged OA and LV-OOA is an interesting result. I recommend publication after addressing these comments.

We would like to thank the reviewer for his/her thoughtful revision. We have incorporated all his/her suggestions in the revised manuscript. Below is our response to his/her specific comments.

My main comment is to include some caveats about how comparisons based on total mass (or even 3 components of mass: POA, LV-OOA, SV-OOA) may not be able to resolve source sectors or governing pathways very well. As an example, IEPOX-SOA has been shown to be a major contributor to ambient OA around the world via more recent AMS PMF analysis (Hu et al. 2015). This type of SOA is likely formed via processing of later generation isoprene products in aqueous acidic aerosol, a process not considered in the model used here. How does neglecting this type of SOA bias comparisons? In addition, cooking aerosol (17% of total OA in Pasadena, Hayes et al. 2013) is not specifically evaluated. Is it included in your emission inventory? Should the model expect to reproduce observed OA if it is missing known pathways?

This is a valid point raised by the reviewer. We have already identified the vehicle cold starts and residential heating sources as a possible cause of model underperformance during winter since the amount of the OA emissions from these sources are probably severely underestimated in our emission inventory. As the reviewer pointed out, residential and commercial cooking is another 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). However, these emissions are not included in emission inventories and global models (Tsigaridis et al., 2014), including the one used in this study. While we have excluded this type of OA from our model evaluation (in cases where cooking OA (COA) has been resolved by the PMF analysis) in order to eliminate any model bias resulting from the omission of cooking emissions in our inventory, organic compounds emitted from cooking sources can be oxidized and form SOA. Therefore, the lack of cooking emissions can be considered as a possible source of OOA underestimation by the model over urban and urban downwind areas. Furthermore, the model does not simulate SOA formation from aqueous-phase reactions, therefore cannot produce IEPOX-SOA that has been recently resolved as an important source of SOA close to isoprene sources (Hu et al., 2015). However, IEPOX-SOA has been identified in only a few of the AMS datasets used in this study in areas strongly influenced by isoprene emissions (e.g. Duke Forest, NC). Therefore, IEPOX-SOA cannot be considered as a major source of model/measurement discrepancy in most of the areas investigated here. A discussion concerning the implications of COA and IEPOX-SOA on model performance has been added in section 5.2.2 of the revised manuscript.

Minor comments:

1. *This is a personal preference, but I encourage use of Tg C/yr instead of Tg/yr when referring to OC*

Following the reviewer's recommendation we changed the units of OC emissions to *Tg C yr*$^{-1}$.

2. *Line 64: point out that the magnitude of missing IVOCs is likely source specific*

We have added to the text that the magnitude of the missing IVOC emissions is estimated to be between 0.25 and 2.8 times POA emissions, depending on the type of the source.

3. *Line 121: Heald et al. 2005 and de Gouw et al. 2005 were performed before isoprene was even considered a source of SOA via traditional semivolatile pathways (e.g. Henze and Seinfeld 2006). These outdated references should be replaced with more recent references to assert that global models currently underestimate OA.*

We have replaced these references with Tsigaridis et al. (2014) who evaluated the current performance of 32 global models in simulating OA mass.

4. Line 257: Do Aiken et al. 2008 OM/OC values need to be updated in light of new AMS calibrations by Canagaratna et al. 2015?

Canagaratna et al. (2015) estimated an OM:OC ratio of 1.34 for HOA and 1.64 for BBOA which are very similar to the values used in our study (1.3 and 1.6 respectively). Therefore, in the revised manuscript we have replaced the reference to Aitken et al. (2008) with the Canagaratna et al. (2015).

5. Lines 310-315: was cooking OA resolved in any data sets or is it likely part of HOA and BBOA?

Cooking OA (COA) has been resolved in some datasets (e.g., Barcelona by Mohr et al., 2012; Fresno, CA by Ge et al., 2012; New York, NY by Sun et al., 2011; Pasadena, CA by Hayes et al., 2013 etc.). However, cooking emissions are not included in our emission inventory; therefore COA was not used in the model evaluation.

6. How is deposition of SOA/SOG handled?

The removal of SOA and SOG species through dry deposition is calculated within the DRYDEP submodel (Kerkweg et al., 2006a) based on the big-leaf approach and the dry deposition velocities depend on physical and chemical properties of the surface cover (e.g., the roughness length, soil pH, leaf stomatal exchange, etc.). The sedimentation of aerosols is calculated within the SEDI submodel (Kerkweg et al., 2006a) using a first-order trapezoid scheme. In cloud scavenging and rainout of gas and aerosol species are treated by the SCAV submodel (Tost et al., 2006). The effective Henry's law coefficient used for calculating the scavenging of all SOG species is equal to 10^5 M atm^{-1}. This discussion has been added in section 2.1 of the revised manuscript.

7. A table of the yield parameters for the traditional VOCs and IVOCs should be reproduced from Tsimpidi et al. 2014 in the supporting information (or in text)

Following the reviewer's recommendation we have added a table in the supporting material which summarizes the aerosol mass yields used to distribute the oxidation products of VOCs in four volatility bins. The oxidation of IVOCs results in only one product with lower saturation concentration by a factor of 10^2 and a stoichiometric coefficient of 1.15 to account for two added oxygen atoms. This information is reported in the text.

8. Line 392 and thereafter: How much does the seasonality of emissions drive the higher POA in winter? Cold temperatures favor partitioning to the particle as indicated, but higher residential wood combustion could be expected as well.

This is a good point. The wintertime emissions of fPOA used in this study are indeed 12% higher than the annual average emissions. Therefore the seasonality of the emissions can explain part of the higher POA concentrations calculated during winter (36% on average). We have included this interpretation in our discussion in 4.1.2 of the revised manuscript.

9. Paragraph starting on line 408: The annual tropospheric burden vs year plot could be made more interesting by including regional trends. Do the emissions capture the expected increase in emissions from China vs decrease in the US?

We have included the continental tropospheric burdens in Figure 6b. Indeed, the Asian anthropogenic OC emissions have increased during the simulated decade by 12%. On the other hand, anthropogenic OA emissions over North America have decreased by 15% during the same period. Over Europe, anthropogenic OA emissions increased until the year 2005 (5%) and then started to decrease reaching 4% lower emissions (compared to 2001) by the end of the decade. However, the simulated fOA tropospheric burdens over the continents (Figure 6b of the revised manuscript) do not reflect this clear trend of emissions since other factors (i.e., meteorology) play an important role on the calculated tropospheric burden as well. This information has been added to the text. We have also performed the same analysis for the simulated bbOA tropospheric burden by including the results from the three main rainforests (Amazon, Congo, SE Asia).

10. Line 428: Clarify the IVOCs from biomass burning. Line 274 indicates IVOCs from biomass burning were not added to the existing POA, yet IVOCs account for 40% of the emissions?

We have rephrased this sentence to avoid confusion. May et al. (2013) quantified the biomass burning organic emissions in the volatility range from 10^{-2} to 10^4 μg m^{-3}. We did not add any IVOC emissions in the $C^* > 10^4$ μg m^{-3} bins for open biomass burning. As a result 40% of the traditional biomass burning OA emissions are assumed to be IVOCs with C* = 10^4 μg m^{-3} (Table 2 of the manuscript). This information has been added to the revised text.

11. Section 5.2.1: How do you separately diagnose errors in the magnitude of POA emissions from the volatility profile?

We have not explicitly diagnosed any errors in POA emissions. Any reference to possible errors in the magnitude of POA emissions from specific source sectors are just hypotheses that should be considered and further explored in future studies. Errors in the POA volatility distributions can also explain parts of the discrepancy. This is explained in the revised manuscript.

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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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Global combustion sources of organic aerosols: Model comparison with 84 AMS factor analysis data sets

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Abstract

Emissions of organic compounds from biomass, biofuel and fossil fuel combustion strongly influence the global atmospheric aerosol load. Some of the organics are directly released as primary organic aerosol (POA). Most are emitted in the gas phase and undergo chemical transformations (i.e., oxidation by hydroxyl radical) and form secondary organic aerosol (SOA). In this work we use the global chemistry climate model EMAC with a computationally efficient module for the description of organic aerosol (OA) composition and evolution in the atmosphere (ORACLE). The tropospheric burden of open biomass and anthropogenic (fossil and biofuel) combustion particles is estimated to be 0.59 Tg and 0.63 Tg, respectively, accounting for about 30% and 32% of the total tropospheric OA load. About 30% of the open biomass burning and 10% of the anthropogenic combustion aerosols originate from direct particle emissions while the rest is formed in the atmosphere. A comprehensive dataset of aerosol mass spectrometer (AMS) measurements along with factor-analysis results from 84 field campaigns across the Northern Hemisphere are used to evaluate the model results. Both the AMS observations and the model results suggest that over urban areas both POA (25-40%) and SOA (60-75%) contribute substantially to the overall OA mass while further downwind and in rural areas the POA concentrations decrease substantially and SOA dominates (80-85%). EMAC does a reasonable job in reproducing POA and SOA levels during most of the year. However, it tends to underpredict POA and SOA concentrations during winter indicating that the model misses ~~a~~ wintertime sources of OA (e.g., residential biofuel use) and ~~a~~ SOA formation pathways (e.g., multiphase oxidation).

1. Introduction

Organic aerosol (OA) is a major contributor to fine particulate matter mass with potentially harmful effects on the environment and human health (Lelieveld et al., 2013; Poschl, 2005), however, ~~its~~ the sources are poorly understood (Kanakidou et al., 2005; ~~Turpin et al., 2000;~~ Goldstein and Galbally, 2007; Donahue et al., 2009; Tsigaridis et al., 2014). OA comprises primary organic aerosol (POA), i.e., directly emitted in the particulate phase, and secondary organic aerosol (SOA), formed within the atmosphere from the oxidation of gas-phase precursors. POA constitutes the particulate OA fraction~~is~~ emitted by anthropogenic combustion processes (i.e., fossil fuels, biofuels) and open biomass burning (i.e., savanna~~s,~~ and forest fires). Anthropogenic combustion emissions of particulate organic carbon (OC) are estimated at 13.9~~8.9~~ Tg C yr^{-1} ~~(2.4 Tg yr^{-1} from fossil fuel and 6.5 Tg yr^{-1} from biofuel) with an uncertainty range of 5 to 17 Tg yr^{-1}~~for the year 2005 (~~Bond~~ Clarke et al., ~~2004~~2007). OC emissions from open biomass burning ~~are approximately~~range from ~~25~~ 13.5 Tg C yr^{-1} to 21.4 Tg C yr^{-1} ~~with an uncertainty range of 13 to 57 Tg yr^{-1}~~during the decade since 2000 (Van der Werf et al., 2010). ~~This uncertainty mainly relates to the emission factors that depend on the fuel burnt and the type of combustion.~~ POA emitted from combustion sources can evaporate rapidly during atmospheric dilution depending on ambient concentrations (~~Hildemann et al., 1989; Lipsky and Robinson, 2006~~Robinson et al., 2010; Ranjan et al., 2012; May et al., 2014). The phase partitioning of the emitted POA depends on the volatility distribution of the emissions. This distribution includes low volatility (LVOC; $C^* < 0.32$ μg m^{-3}), semivolatile (SVOC; 0.32 μg m^{-3} $< C^* < 320$ μg m^{-3}), and intermediate volatility (IVOC; $3.2\text{x}10^2$ μg m^{-3} $< C^* < 3.2\text{x}10^6$ μg m^6) organic compounds. The corresponding emission factors can be measured using dilution samplers and are estimated as a function of the saturation concentration of the emitted organic compounds (Grieshop et al., 2009). Traditional emission inventories (e.g., ~~Bond~~ Clarke et al., ~~2004~~2007; Van Der Werf et al., 2010) account only for a small fraction of the emitted IVOCs since they are based on filter samples collected at aerosol concentrations up to 10^4 μg m^{-3} (Shrivastava et al., 2008; Robinson et al., 2010). The amount of IVOC emissions missing in traditional inventories is estimated to be between 0.25 and 2.8 times POA emissions, depending on the type of the source (~~Schauer et al., 1999, 2001, 2002~~Shrivastava et al., 2008; Robinson et al., 2010).

Organic emissions further downwind mix with background air, resulting in cooling and dilution and altering the~~ir~~ gas-particle partitioning. The organic compounds that remain in the gas phase can undergo chemical transformations (i.e., oxidation by hydroxyl radical), become less volatile and may be transferred into ~~the~~ SOA (Donahue et al., 2006). Therefore, in addition to direct emissions of POA, it is important to understand the potential of combustion emissions to contribute to SOA formation. Numerous studies have indicated that SOA usually exceeds POA even in urban environments with substantial primary emissions (~~Zhang et al., 2007;~~ Jimenez et al., 2009; ~~Subramanian et al., 2007;~~ Stone et al., 2009; Sun et al., 2011; Mohr et al., 2012; Hayes et al., 2013). However, the overall contribution of combustion emissions to ambient SOA and OA remains uncertain (Chirico et al., 2010; Miracolo et al., 2011; Samy and Zielinska, 2010; ~~Weitkamp et al., 2007;~~ Gentner et al., 2012; ~~Robinson et al., 2007;~~ Bahreini et al., 2012; Gordon et al., 2014). Together with the OA mass concentration, ~~its~~ the hygroscopic, chemical and optical properties continue to change because of chemical processing by gas-phase oxidants (~~Kanakidou et al., 2005~~Jimenez et al., 2009). These changes affect the OA radiative forcing ~~of OA~~ on climate ~~through both its~~by direct and indirect ~~aerosol~~ effects, the latter through cloud formation (Poschl, 2005; McFiggans et al., 2006; ~~Bond et al., 1999; Bond, 2001; Jacobson, 1999; Kirchstetter et al., 2004; Kotchenruther and Hobbs, 1998; Kotchenruther et al., 1999~~IPCC; 2013).

Mass spectrometry has been widely used in aerosol ~~analysis~~ analyses because of the universal, sensitive and rapid detection of aerosol components (~~McKeown et al., 1991;~~ Suess and Prather, 1999). The Aerosol Mass Spectrometer (AMS) (~~Jayne et al., 2000;~~ Jimenez et al., 2003) has been the most commonly used instrument in ~~the last decade~~recent years. AMS is capable of quantitatively measuring the OA mass concentrations with high time and particle size resolution (~~Allan et al., 2003;~~ Takegawa et al., 2005; Zhang et al., 2005b). Several factor analysis techniques have been employed to ~~provide~~ extract information about processes and sources of OA. These techniques include principal component analysis (CPCA; Zhang et al., 2005a), multiple component analysis (MCA; Zhang et al., 2007), hierarchical cluster analysis (Marcolli et al., 2006), the Multilinear Engine (ME-2; Lanz et al., 2008), and positive matrix factorization (PMF; Paatero and Tapper, 1994; Paatero, 1997), with the latter being the most commonly used (Lanz et al., 2007; Nemitz et al., 2008~~; Aiken et al., 2008~~; Aiken et al., 2009; Ulbrich et al., 2009; DeCarlo et al., 2010; Mohr et al., 2012; Hayes et al., 2013; Crippa et al., 2014; Carbone et al., 2014; Chen et al., 2015).

PMF allows the classification of OA into ~~different~~ several types based on different temporal and mass spectral signatures. Two major components often resolved by the analysis of the AMS measurements are hydrocarbon-like organic aerosol (HOA) and oxygenated organic aerosol (OOA) (~~Zhang et al., 2005c~~Zhang et al., 2007; Jimenez et al., 2009). Biomass burning OA (BBOA), marine-related OA (MOA) and cooking OA (COA) are other OA components that PMF may identify as important components of the observed OA (~~Zhang et al., 2007~~Lanz et al., 2010; Mohr et al., 2012; Kostenidou et al., 2013; Crippa et al., ~~2014~~2013a~~; Ng et al., 2011~~). HOA correlates with combustion tracers (e.g., CO, EC, and NO$_x$) and is considered as a surrogate for fossil fuel combustion POA (~~Zhang et al., 2005c;~~ Lanz et al., 2007; ~~Aiken et al., 2009;~~ Ulbrich et al., 2009; Crippa et al., 2014). BBOA correlates with tracers~~ originating~~ from biomass burning (e.g., acetonitrile, levoglucosan, and potassium) and is considered a surrogate of biomass burning POA (Aiken et al., 2010; ~~Ulbrich et al., 2009~~Lanz et al., 2010; Crippa et al., 2014). OOA ~~has the same temporal pattern as~~often correlates with secondary ~~PM components~~pollutants (e.g. ozone, sulfate, and nitrate) and is considered a~~s~~ surrogate for SOA (~~Zhang et al., 2007~~Jimenez et al., 2009; Ng et al., 2011; ~~Herndon et al., 2008~~). However, Crippa et al. (2014) have reported that the OOA correlation with secondary inorganic species might not be very high in at least some field campaigns. OOA can include SOA from various precursors, such as anthropogenic and biogenic VOCs, as well as SVOCs and IVOCs from fossil fuel, biofuel and open biomass burning. PMF often classifies OOA into two subtypes that differ in the degree of oxidation: a more strongly oxygenated low-volatility OOA (LV-OOA) often correlating with sulfate, and a less oxygenated semivolatile OOA (SV-OOA) usually correlating with nitrate (Jimenez et al., 2009; Crippa et al., 2014). Field campaigns in the Northern Hemisphere have shown that HOA accounts for approximately one third of the OA in urban sites ~~while~~ and OOA accounts for the remaining two thirds~~.~~, while OOA represents roughly 95% of the OA in rural/remote regions (Zhang et al., 2007).

Global chemistry climate and chemical transport models systematically underpredict OA levels~~ (Heald et al., 2005; de Gouw et al., 2005)~~, especially ~~during episodes of intense photochemistry~~over and downwind of anthropogenic source regions (~~Vutukuru et al., 2006; Volkamer et al., 2006~~Tsigaridis et al., 2014). At the same time, ~~traditional~~ global models tend to predict a dominance of POA at mid-latitudes in the Northern Hemisphere while measurements indicate the opposite (~~Heald et al., 2005;~~ Henze et al., 2008; Tsigaridis et al., 2014). The same models ~~predict~~ indicate that the formation of SOA from biogenic sources greatly exceeds that from anthropogenic sources. The shortcomings in ~~the traditional~~many OA models are partially due to the assumption that POA is non-volatile and nonreactive (Kanakidou et al., 2005; Jimenez et al., 2009)~~)~~. ~~In order t~~To ~~help remedy~~address these shortcomings, Donahue et al. (2006) developed the volatility basis set (VBS) framework which assumes that POA emissions are semivolatile and photochemically reactive and ~~are distributed in~~ uses logarithmically spaced volatility bins to distribute POA upon emission. Recently, several regional-scale modeling studies have accounted for the semivolatile nature and chemical aging of organic compounds demonstrating improvements in reproducing the OA concentrations and ~~its~~ chemical composition (Robinson et al., 2007; Shrivastava et al., 2008; Murphy and Pandis, 2009; Tsimpidi et al., 2010; Tsimpidi et al., 2011; Hodzic et al., 2010; Fountoukis et al., 2011; Bergstrom et al., 2012; Athanasopoulou et al., 2013; Zhang et al., 2013; Fountoukis et al., 2014). However, only few global modeling studies have yet adopted the VBS approach to simulate the SOA formation from the chemical aging of SVOC and IVOC emissions (~~Pye and Seinfeld, 2010;~~ Jathar et al., 2011; Tsimpidi et al., 2014; Shrivastava et al., 2015; Hodzic et al., 2015). ~~The model results indicate a larger share for anthropogenic SOA on a global scale.~~ According to these studies, the modeled tropospheric burden of POA is 0.~~03~~09-0.~~23~~ 94 Tg and of SOA ~~1.6~~1.8-2.8 Tg~~, with SVOCs and IVOCs contributing 0.7-1.6 Tg to the total SOA~~.

In this work we use ORACLE, a computationally efficient module for the description of organic aerosol composition and evolution in the atmosphere (Tsimpidi et al., 2014), to estimate the impact of open biomass burning and anthropogenic combustion emissions and their chemical aging on global OA budgets and distributions. An extensive global dataset of AMS measurements and factor-analysis results from 84 field campaigns in the Northern Hemisphere are used in combination with the model results during the period of 2001-2010. This integrated effort provides further insights into the temporal and geographical variability of the OA~~contribution of the emitted~~ particles, emission strengths and the chemical~~ly~~ ~~processed~~ processing of organics ~~material~~ from combustion sources ~~to the total OA load~~.

2. Model description and application

2.1 EMAC Model

The ECHAM/MESSy Atmospheric Chemistry (EMAC) model is a numerical chemistry and climate simulation system that includes sub-models describing the lower and middle atmosphere processes (Jöckel et al., 2006). EMAC includes submodels that describe gas-phase chemistry (MECCA; Sander et al., 2011), inorganic aerosol microphysics (GMXe; Pringle et al., 2010), cloud microphysics (CLOUD; Jöckel et al., 2006), aerosol optical properties (AEROPT; Lauer et al., 2007), dry deposition (DRYDEP; Kerkweg et al., 2006a), sedimentation (SEDI; Kerkweg et al., 2006a), cloud scavenging (SCAV; Tost et al., 2006), emissions (ONLEM and OFFLEM; Kerkweg et al., 2006b), and organic aerosol formation and growth (ORACLE; Tsimpidi et al., 2014).

The removal of gas and aerosol organic compounds through dry deposition is calculated with the DRYDEP submodel (Kerkweg et al., 2006a) based on the big-leaf approach, and the dry deposition velocities depend on physical and chemical properties of the surface cover (e.g., the roughness length, soil pH, leaf stomatal exchange, etc.). The sedimentation of aerosols is calculated with the SEDI submodel (Kerkweg et al., 2006a) using a first-order trapezoid scheme. In-cloud scavenging and rainout of gas and aerosol species are treated by the SCAV submodel (Tost et al., 2006). The effective Henry's law coefficient used for calculating the scavenging rates of LVOCs, SVOCs, and IVOCs is 10^5 M atm^{-1}.

The spectral resolution used in this study is T63L31, corresponding to a horizontal grid resolution of 1.875^{o}x1.875^o and 31 vertical layers extending to 10 hPa at about 25 km altitude. EMAC ~~is applied~~has been run for 11 years, covering the period 2000–2010, and the first year is used as spin-up. EMAC 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). Tsimpidi et al. (2014) performed an in depth evaluation of the EMAC calculated total OA over different continents by using measurements from the EMEP network over Europe, the IMPROVE network over North America, and several short-term field campaigns over East Asia, subtropical West Africa, the Amazon rainforest, and the Canadian boreal forest. The present work focuses on the model evaluation for the individual OA components (i.e., POA and SOA). In addition, the statistical evaluation of EMAC results for the inorganic components summarized in Tables S1-S3 in the Supplementary Material.

2.2 ORACLE Module

ORACLE is a computationally efficient submodel for the description of OA composition and evolution in the atmosphere which has been implemented into the EMAC model by Tsimpidi et al. (2014). ORACLE simulates a variety of semivolatile organic species and reaction products and separates them into groups of compounds with logarithmically spaced effective saturation concentrations.

In this study, primary organic emissions from open biomass burning and from anthropogenic sources (i.e., fossil fuel and biofuel) are simulated using separate surrogate species for each source category. They are subdivided into three groups of organic compounds: low volatility, LVOCs (10^{-2} μg m^{-3}), semi-volatile, SVOCs (10^{0} and 10^{2} μg m^{-3}) and intermediate volatility organic compounds, IVOCs (10^{4} and 10^{6} μg m^{-3}). These organic compounds are allowed to partition between the gas and aerosol phases resulting in the formation of fPOA (anthropogenic POA from fossil fuel and biofuel combustion) and bbPOA (natural POA from open biomass burning). VOCs are distinguished into anthropogenic and biogenic and their oxidation products are distributed in four volatility bins with effective saturation concentrations of 10^{0}, 10^{1}, 10^{2}, and 10^{3} μg m^{-3} at 298 K by using the aerosol mass yields (Table S4) by Tsimpidi et al., (2014). Gas-phase photochemical reactions that change the volatility of the organics are taken into account and their oxidation products (SOA-sv, SOA-iv, and SOA-v) are simulated separately in the module to keep track of their origin. The suffixes –sv, -iv and -v ~~after~~ to the term SOA ~~declare~~ define ~~the group of its~~category of precursors (SVOCs, IVOCs, and VOCs, respectively). For the current application, SOA components are divided into four groups based on their source: anthropogenic from fossil fuel and biofuel combustion sources (fSOA), natural from open biomass burning (bbSOA), SOA from anthropogenic (aSOA-v) and biogenic (bSOA-v) VOCs. This study focuses on the OA produced from primary combustion sources and discusses in detail results for the first two types of SOA (fSOA and bbSOA). The model set up for simulating the formation of aSOA-v and bSOA-v and the corresponding results can be found in Tsimpidi et al. (2014). In addition, in this work ORACLE has been modified to distinguish the formation of fresh SOA and aged SOA by adding additional tracers ~~in~~to the model. The first generation oxidation products of SVOCs, IVOCs, and VOCs are characterized as fresh while SOA produced from any additional oxidation step is grouped together and considered aged (Figure 1). LVOCs are not allowed to participate in photochemical reactions since they are in the lowest volatility bin. This assumption may introduce a small bias ~~on~~ in our results only under extremely clean conditions (OA ≤ 10^{-2} μg m^{-3}) where part of LVOC is in the gas phase. Adding another bin in the volatility distribution to accurately represent the extremely low volatility organic compounds (e.g., ELVOCs with C^* lower than 10^{-3}) would be useful only for studying new particle formation, which is outside the scope of the current work. 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). This formulation is comparable to a number of global and regional studies which assume two orders of magnitude reduction in volatility and up to 50% increase in mass per reaction (Grieshop et al. 2009; Hodzic et al., 2010; Pye and Seinfeld 2010, etc.). Shrivastava et al. (2011) even used seven orders of magnitude reduction in volatility per reaction. ~~While this representation is common for global models (e.g., Pye and Seinfeld, 2010; Tsimpidi et al., 2014), regional models use a more conservative formulation to represent the aging of SVOC and IVOC by assuming a reduction in volatility by one order of magnitude after each oxidation step (e.g., Tsimpidi et al., 2010; Bergstrom et al., 2012).~~ However, 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 (e.g., Tsimpidi et al., 2010; Jathar et al., 2011; Bergstrom et al., 2012) or two orders of magnitude, the oxidation products can be up to four orders of magnitude lower in volatility than the precursor (Kroll and Seinfeld, 2008). Furthermore, ORACLE calculates the fraction of the semivolatile organic compounds that condenses to (or evaporates from) the particle phase by assuming bulk equilibrium and that all organic compounds form a pseudo-ideal solution (Tsimpidi et al., 2014). Overall, the primary aerosol formation from the phase partitioning of the freshly emitted LVOCs and SVOCs, as well as the formation of SOA from the photo-oxidation of SVOCs and IVOCs are described by the following reactions:

$$LVOC_i \leftrightarrow POA_i \quad (R1)$$

$$SVOC_i \leftrightarrow POA_i \quad (R2)$$

$$SVOC_i + OH \rightarrow 1.15\ SOG\text{-}sv_{i-1} \quad (R3)$$

$$SOG\text{-}sv_i + OH \rightarrow 1.15\ SOG\text{-}sv_{i-1} \quad (R4)$$

$$\text{SOG-sv}_i \leftrightarrow \text{SOA-sv}_i \qquad \text{(R5)}$$

$$\text{IVOC}_i + \text{OH} \rightarrow 1.15\ \text{SOG-iv}_{i-1} \qquad \text{(R6)}$$

$$\text{SOG-iv}_i + \text{OH} \rightarrow 1.15\ \text{SOG-iv}_{i-1} \qquad \text{(R7)}$$

$$\text{SOG-iv}_i \leftrightarrow \text{SOA-iv}_i \qquad \text{(R8)}$$

where i is the original volatility bin and i-1 is the volatility bin with saturation concentration reduced by a factor of 10^2. The term SOG corresponds to secondary organic gas that is produced by at least one chemical reaction in the atmosphere. The symbol "↔" denotes the equillibrium between the gas and the aerosol phases. It is worth ~~mentioned~~ mentioning that the production of RO_2 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). The model set-up and the different aerosol types and chemical processes that are simulated by ORACLE for this study are illustrated in Figure 1. More details about ORACLE can be found in Tsimpidi et al. (2014).

2.3 Emission inventory

The CMIP5 RCP4.5 emission inventory (Clarke et al., 2007) is used for the anthropogenic POA emissions from fossil fuel and biofuel combustion sources. The open biomass burning emissions from savanna burning and forest fires are based on the Global Fire Emissions Database (GFED v3.1; van der Werf et al., 2010). These emission datasets report the mass of the OC emitted. Therefore, in order to determine the total organic matter (OM) emitted (including any additional species associated with the carbon) OM/OC values of 1.3 for anthropogenic POA and 1.6 for open biomass burning POA are used. These values are based on the OM~~/~~:OC ~~values~~ ratios ~~estimated~~ estimated by Canagaratna~~Aiken~~ et al. (2015~~08~~) for HOA and BBOA, respectively. Furthermore, the above emission datasets are monthly resolved and treat POA as non-reactive and non-volatile. However, only a fraction of this organic material is directly emitted in the aerosol phase as POA. Most of it is rapidly transferred to the gas phase where it can undergo chemical transformations and form SOA. Therefore, ~~a~~ key input for the accurate description of these compounds and their chemical aging is the~~ir~~ volatility distribution at 298 K. Figure 2 depicts the volatility distributions assumed for this study which cover a range of ~~volatilities from~~ 10^{-2} to 10^{4} μg m^{-3} for open biomass burning (May et al., 2013) and 10^{-2} to 10^{6} μg m^{-3} for fossil and biofuel combustion emissions (Robinson et al., 2007). Emission inventories are based on samples collected at aerosol concentrations up to 10^{4} μg m^{-3} (Shrivastava et al., 2008; Robinson et al., 2010). As a result IVOC emissions with $C^* > 10^{4}$ μg m^{-3} are missing from the traditional emission inventories and have to be accounted for by assigning additional emissions in this volatility range. We assume that the missing IVOC emissions from anthropogenic combustion are 1.5 times the traditional OA emissions included in the inventory (Shrivastava et al., 2008; Tsimpidi et al., 2010), therefore the sum of the emission factors is 2.5. No additional IVOC emissions are ~~added~~ assumed in the $C^* > 10^{4}$ μg m^{-3} bins for open biomass burning and therefore the sum for the biomass burning emission factors is unity. As a result, 40% of the biomass burning OA emissions represents IVOCs with $C^* = 10^{4}$ μg m^{-3} (Table 2). The sensitivity of our results to these assumptions will be discussed in a subsequent article in preparation. Overall, the decadal average global emission flux of SVOCs and IVOCs is 44 Tg yr^{-1} from anthropogenic combustion sources and 28 Tg yr^{-1} from open biomass burning sources.

3. Model evaluation methodology

3.1 Factor analysis of AMS measurements

During the period 2001-2010, 84 field campaigns were performed in the Northern Hemisphere using the AMS for measuring ambient OA concentrations in urban, urban-downwind, and rural environments (Figure 3). Information for each of these campaigns is given in Tables ~~S1~~S5-~~S3~~S7. The OA source apportionment for all sites was taken from the literature (Tables ~~S1~~S5-~~S3~~S7) and performed using factor-analysis techniques classifying OA as HOA, corresponding ~~roughly~~ to POA from fossil fuel combustion, and OOA, corresponding to SOA. Therefore, AMS HOA is compared with modeled fPOA, which is emitted and remains in the aerosol phase without undergoing ~~any~~ chemical reactions, and AMS-OOA is compared with modeled SOA (the sum of SOA-sv, SOA-iv, and SOA-v), formed from the oxidation of gas phase precursors (SVOCs, IVOCs, and VOCs). At many locations, PMF and other factor analysis techniques identified two subtypes of OOA that differ in volatility and oxidation state: semi-volatile OOA (SV-OOA) and low-volatility OOA (LV-OOA). There are different potential interpretations of SV-OOA and LV-OOA. SV-OOA often ~~correlated~~ correlates with semi-volatile species such as ammonium nitrate and ~~was~~ is less oxygenated, consistent with relatively fresh SOA (Zhang et al., 2011; Ng et al., 2011). LV-OOA ~~was~~ usually ~~correlated~~ correlates with nonvolatile secondary species such as sulfate, and ~~was~~ is highly oxygenated, consistent with regional aged OA (Zhang et al., 2011; Ng et al., 2011). Recently, Ehn et al. (2014) found a direct pathway which leads to the formation of fresh LV-OOA from the oxidation of several biogenic VOCs. Here we test the hypothesis that SV-OOA corresponds to the first generation products ~~while~~ and LV-OOA to the later generation ones. Therefore, AMS SV-OOA is compared with the fraction of SOA-sv, SOA-iv, and SOA-v from the first oxidation step of SVOCs, IVOC, and VOCs as it is tracked separately (henceforth EMAC fresh SOA). Then AMS LV-OOA is compared with the fraction of SOA-sv, SOA-iv, and SOA-v from any additional oxidation step (henceforth EMAC aged SOA). Finally, in a few ~~of the~~ field campaigns, e.g., in the Alps (Lanz et al., 2010), residential wood burning was found to be a major source of OA. However, residential wood burning is included in EMAC as fPOA. To account for this inconsistency for the AMS data sets that include BBOA we compare the sum of the simulated fPOA and bbPOA (henceforth EMAC POA) ~~against~~ to the sum of the AMS HOA and BBOA (henceforth AMS POA). In data sets where OA from cooking activities has been resolved by the PMF analysis, AMS COA has not been taken into account for the model evaluation since these emissions are not included in our emission inventory.

3.2 Evaluation metrics

The mean bias (MB), mean absolute gross error (MAGE), normalized mean bias (NMB), normalized mean error (NME), and the root mean square error (RMSE) are used to assess the model performance:

$$MAGE = \frac{1}{N}\sum_{i=1}^{N}\left|P_i - O_i\right| \ (1) \qquad MB = \frac{1}{N}\sum_{i=1}^{N}\left(P_i - O_i\right) \ (2)$$

$$NME = \frac{\sum_{i=1}^{N} |P_i - O_i|}{\sum_{i=1}^{N} O_i} \quad (3) \qquad NMB = \frac{\sum_{i=1}^{N} (P_i - O_i)}{\sum_{i=1}^{N} O_i} \quad (4)$$

$$RMSE = \left[\frac{1}{N} \sum_{i=1}^{N} (P_i - O_i)^2 \right]^{\frac{1}{2}} \quad (5)$$

where O_i is the observed campaign average value of the ith OA component, P_i is the corresponding modelled value during the same period, and N is the total number of comparisons used for the evaluation. NME (in %) and MAGE (in μg m^{-3}) provide an estimate of the overall discrepancy between predictions and observations, while NMB (in %) and MB (in μg m^{-3}) are sensitive to systematic errors. RMSE (in μg m^{-3}) is the root of the mean square error, which incorporates both the variance of the prediction and its bias. Both NME and MAGE inherently include the corresponding bias, which is the reason why their magnitude is equal or larger than NMB and MB respectively. For an unbiased prediction, NME and MAGE express the variance. When NME and NMB or MAGE and MB are close, the discrepancy is explained as a systematic bias rather than scatter. When ~~the magnitude of~~ NME/MAGE ~~is larger than~~exceeds NMB/MB, part of the discrepancy between predictions and observations is explained as scatter. ~~In order t~~To determine the effects ~~that~~ of the site type, geographical location ~~or~~and the seasonal cycle ~~has~~ on the ~~performance of the model~~model results, the evaluation metrics are calculated 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).

4. Model results

4.1 OA from anthropogenic combustion

4.1.1 Geographical distribution

Figure 4 depicts the simulated, decadal average global surface concentrations of fPOA and fSOA from anthropogenic SVOC and IVOC sources (fossil and biofuel combustion). The average surface concentration of fPOA is 0.1 μg m^{-3}. Higher fPOA concentrations (up to 14 μg m^{-3}) are simulated over densely populated and highly industrialized areas (e.g., Eastern China, Northern India, Central Europe, etc.) where there are substantial anthropogenic combustion emissions. ~~Further d~~Downwind of the sources~~,~~ fPOA concentrations decrease substantially since they are diluted and a large fraction is predicted to evaporate during transport. This results in a highly inhomogeneous spatial distribution of fPOA concentrations (Figure 4a). In contrast, fSOA is more regionally distributed with high concentrations (up to 9.5 μg m^{-3}) downwind of the anthropogenic sources due to its continuous production and long-range transport from SVOCs and IVOCs (Figure 4b). This results in a continental fSOA background of 0.5-1 μg m^{-3} and in concentrations of around 1 μg m^{-3} over marine regions close to anthropogenic sources (e.g., Arabian Sea, Yellow Sea~~, etc.~~). The average surface concentration of fSOA is 0.26 μg m^{-3} with 73% of it originating from the oxidation of IVOC emissions. This result supports the hypothesis of several recent studies that IVOC emission and oxidation may be a significant SOA source that ~~is~~ has been missing from chemistry climate models (Jathar et al., 2011; Tsimpidi et al., 2014). The relatively small contribution (25%) of SVOCs to total fSOA ~~can be justified by~~follows from its low emissions compared to the IVOCs (two times lower) and by the fact that a significant fraction of SVOCs stays in the aerosol phase (as POA) without undergoing ~~any~~ chemical reactions (Tsimpidi et al., 2014).

The fraction of fossil and biofuel combustion OA (fOA) that is formed through the oxidation of gas phase species, fSOA/fOA, is consistently high with values ranging from around 20% close to the sources to 100% away ~~from them~~in remote regions with a global average of 83% at the surface (Figure 5a). This ~~result~~ suggests that over urban areas both fPOA and fSOA contribute significantly to the overall OA mass while further downwind and in rural areas ~~the~~ SOA formation dominates since POA ~~concentrations~~ decreases substantially due to dilution and evaporation. The OA due to anthropogenic combustion sources contributes significantly to total OA over the continents in the ~~northern~~ Northern Hemisphere (Figure 5b). The highest contribution is predicted over Eastern China (83%) and the lowest over the Southeastern US (23%). Over mid-latitude oceans, the contribution of fOA to total OA is also high (around 60%) due to the long-range transport of ~~secondary~~ SOA. On the other hand, fOA/OA is very low (0-10%) over the tropical and boreal forest regions ~~due~~ in contrast to the significant bbOA and bSOA-v concentrations over these areas. The eastern part of the Eurasian boreal forest is an exception since the lower emissions of bbOA together with the considerable amount of fSOA transported from Europe results in fOA/OA fractions of about 40%. Overall, the predicted global average fOA/OA is 38%. This result highlights the importance of anthropogenic emissions for global OA levels, also suggested by other recent studies (Carslaw et al., 2013; Lee et al., 2013; Spracklen et al., 2011).

4.1.2 Temporal profile

Table 1 shows the decadal average tropospheric burden of fPOA and fSOA. The decadal average tropospheric burden of total fOA is 0.63 Tg (10% fPOA and 90% fSOA). The tropospheric fSOA/fOA is higher than at the surface since SVOC and IVOC continue forming fSOA at higher altitudes (Tsimpidi et al., 2014).

The wintertime burden of fPOA is 36% higher than its annual average value (Figure 6a). This increase is partially driven by the seasonality of the emissions since anthropogenic OA emissions are 12% higher during winter compared to the annual average. Furthermore, ~~The~~ the lower temperatures that occur during winter in the Northern Hemisphere drive the gas-particle partitioning of freshly emitted SVOCs to the aerosol phase resulting in higher fPOA concentrations. ~~The wintertime burden of fPOA is 36% higher than its annual average value (Figure 6a). In~~ At the same time, less SVOCs are available in the gas phase to react with the lower wintertime OH resulting in reduced formation of fSOA. The wintertime tropospheric burden of fSOA is 16% lower than ~~its~~ the annual average value (Figure 6a) representing 87% of the fOA. During summer, the photooxidation of SVOC and IVOC is significantly enhanced; however, the increase in fSOA mass is compensated by evaporation due to the high temperatures resulting in an overall increase of only 3% compared to the annual average values. High temperatures during summer also result in a significant decrease of fPOA due to evaporation, i.e., a 27% decrease compared to the annual average tropospheric burden (Figure 6a). Overall, the tropospheric fSOA:fOA during summer ~~is increased~~ increases to 93%. The highest fSOA concentrations are predicted during spring (i.e., April) when ~~the~~ photochemistry is ~~enhanced~~ active and the moderately low temperatures favor the partitioning into the aerosol phase (Figure 6a).

Figure 6b depicts the ~~change of~~annual tropospheric fOA ~~annual tropospheric burden~~variability over the simulated years (2001 to 2010). The ~~decadal~~ variability of the model ~~predictions~~ predicted ~~for~~ fOA is very low (±4%) since anthropogenic emissions are assumed to have small differences between the simulated years (Clarke et al., 2007). The anthropogenic ~~SV~~OC ~~and IVOC~~ emissions from fossil and biofuel combustion increase~~d~~ ~~approximately~~ by ~~0.5~~1.23 Tg C yr^{-1} (10%) ~~on average~~ during the simulated decade. Over Asia and Africa, anthropogenic OC emissions have increased by 12% and 33%, respectively, during the simulated decade. On the other hand, anthropogenic OC emissions over North America have decreased by 15% during the same period. Over South America, anthropogenic OC emissions have decreased up to the year 2005 (10%) and then remained about constant until the end of the decade. Over Europe, anthropogenic OC emissions have increased up to the year 2005 (5%) and then started to decrease reaching 4% lower emissions (compared to 2001) by the end of the decade. However, the simulated fOA tropospheric burdens over the continents (Figure 6b) do not reflect this clear trend of emissions since other factors (i.e., meteorology) play an important role. Overall, ~~The~~ the lowest fOA global tropospheric burden is calculated during the years 2001 and 2003 (0.61 Tg yr^{-1}) and the highest during the year 2009 (0.66 Tg yr^{-1}).

4.2 OA from open biomass burning

4.2.1 Geographical distribution

Figure 7 depicts the simulated decadal average global surface concentrations of bbPOA and bbSOA. The average surface concentration of bbPOA is 0.11 μg m^{-3}. The highest bbPOA concentrations (up to 7.7 μg m^{-3}) are predicted over the tropical rainforests (i.e., Amazon, Congo, and Southeast Asia) and the boreal forests (i.e., Alaska, Canada, and Russia) due to substantial emissions from forest and savannah fires. Similar to fPOA, bbPOA levels rapidly decrease as the air masses ~~moves~~ ~~away~~travel from the sources due to dilution and evaporation (Figure 7a). The average surface concentration of bbSOA is 0.15 μg m^{-3}. In contrast to anthropogenic combustion emissions, IVOCs are assumed to account for only 40% of the total open biomass burning emissions. Nevertheless, the model predicts that the bbSOA formed due to the oxidation of IVOCs (46%) is similar to that from the oxidation of SVOCs (54%). This result corroborates our finding that IVOCs are a significant source of SOA. bbSOA concentrations are more spatially homogeneous compared to bbPOA reaching high levels (up to 6.4 μg m^{-3}) over a wide area covering most of South America, Central and South Africa, Southeastern Asia, ~~and~~ including Indonesia (Figure 7b). The atmosphere over the South~~ern~~ Atlantic Ocean is also strongly influenced by long-range transport of bbSOA from the Congo Basin (1-3 μg m^{-3}). Over these areas, the atmospheric conditions are favorable for the photochemical oxidation of SVOCs and IVOCs. On the other hand, over the boreal forests, the low temperatures favor the partitioning of SVOCs into the particulate phase forming bbPOA, and at the same time the photo-oxidation of IVOCs is slow. This results in moderate average bbSOA concentrations around 0.5 μg m^{-3}.

Figure 8a depicts the predicted decadal average contribution of bbSOA to total bbOA (bbSOA/bbOA) at the surface. bbSOA/bbOA is high with values ranging from around 35% over the ~~large~~ tropical and boreal forests to 85% in areas downwind ~~of them~~ and over the oceans. The global average bbSOA/bbOA at the surface is prediced to be 72%. This result indicates that even though the biomass burning emissions are distributed in relatively low volatility bins ($C^* \leq 10^4$ μg m^{-3}), bbSOA still exceeds primary biomass burning OA on a global scale. Figure 8b depicts the decadal average surface contribution of bbOA to total OA (bbOA/OA). As expected, bbOA contributes significantly to total OA over the tropical and boreal forests (around 60%) while it has a smaller impact on OA levels over the mid-latitude continents of the Northern Hemisphere. This result does not include other types of biomass combustion (e.g., for residential heating) that often contribute significantly ~~over~~ in urban areas (Chen et al., 2007; Wang et al., 2007; Lanz et al., 2010). High bbOA contributions are also predicted downwind of the boreal forests (up to 80%). Furthermore, the bbOA/OA ratio is high (50-90%) off the ~~subtropical~~ west coasts of Africa, South America and Indonesia. These high values are due to the chemical aging of biomass burning SVOCs and IVOCs in contrast to the chemical products of biogenic VOCs which are ~~assumed to have very low later generation SOA~~not allowed to participate in additional photochemical reactions (Tsimpidi et al., 2014). Overall, the global average bbOA/OA is predicted to be 26%.

4.2.2 Temporal evolution

The decadal average tropospheric burden of total bbOA is 0.59 Tg yr^{-1} (30% bbPOA, 70% bbSOA) (Table 1). The fraction of bbOA that is secondary is less than that of fOA (90%).

The interannual variability of bbPOA and bbSOA is high due to the seasonality ~~variability~~ of fires (Figure 9a). During ~~the months of~~ July to September (dry season)~~,~~ intense wildfires are reported over the tropics related to the low precipitation and high temperatures. This results in high biomass burning emissions which together with the intense photochemical activity result in bbOA tropospheric burdens of up to 1.4 Tg yr^{-1} during August (130% higher than the annual average). The lowest bbOA tropospheric burdens are estimated during the wet season (0.21 Tg yr^{-1} during April, 64% lower than the annual average). Furthermore, during the dry season OA consists mainly of bbOA over the tropical rainforests due to the intense wildfires while during the wet season OA consists mainly of biogenic SOA since biomass burning emissions are low. As a result, the bbOA/OA has a significant seasonal variability as well; during the dry season the global average bbOA/OA increases significantly (e.g., 41% during August; not shown) while during the wet season it is significantly lower (e.g., 11% during March; not shown).

The decadal variability of the model predictions is also important since open biomass burning emissions can vary significantly from year to year (Figure 9b). The years 2001 and 2009 had relatively low fire ~~emissions~~ activity (13.5 Tg C yr^{-1}) and the bbOA annual tropospheric burden was 0.47 Tg yr^{-1} (21% lower than the decadal average). During these two years tropospheric bbOA was lower over both the Amazon and the Congo basin (Figure 9b). The year of 2010 on the other hand was characterized by severe wildfires, especially ~~over~~ in the Amazon region (OC emissions were twice the decadal average) resulting in a global bbOA ~~annual~~ source~~tropospheric burden~~ of 0.72 Tg yr^{-1} (21% higher than the decadal average). Over the Congo Basin, the calculated tropospheric burden peaks during the years 2005 and 2010 (Figure 9b) while over the Amazon Basin, the highest burdens are calculated during the years 2007 and 2010. The above results~~This~~ ~~is~~ are consistent with Chen et al. (2013) who analyzed satellite data to detect the fire activity over the Amazon rainforest and reported a twofold increase in fire activity during 2010 compared to 2009.

5. Comparison with AMS data

5.1 Evaluation over urban areas

The spatial resolution used in the current application as well as in most global model applications (Tsigaridis et al., 2014) can introduce potentially significant errors over urban areas. Other issues can also add to the model/measurement discrepancy over cities. For example, global models, including EMAC, lack OA emissions from residential and commercial cooking activities (Tsigaridis et al., 2014). However, cooking can be 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). Therefore our analysis and use of the corresponding urban AMS datasets should be viewed as an effort to quantify the magnitude of these errors. In addition, 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 global models over urban locations can provide useful information about their potential biases in these locations.

AMS observations indicate that over urban areas the POA (sum of HOA and BBOA) concentration is relatively high while further downwind and in rural areas it decreases substantially due to dilution and evaporation (Figure 10a). The model is able to reproduce this trend, however, it significantly underpredicts (NMB=-65%, Table 2) the high values of POA over urban areas and especially over densely populated areas such as Beijing, Tokyo and Mexico City (Table S5; Figure 11a). This underprediction appears to be typical for global models (Tsigaridis et al., 2014) and is partly associated with the limited spatial resolution of the model (the size of a grid cell used typically exceeds the size of most urban centers) and the lack of COA emissions. The model underestimates SOA (NMB=-33%, Table 2) over densely populated areas such as Beijing and Mexico City (Table S5; Figure 12a) partially due to its limited spatial resolution. In addition, the lack of COA emissions can be considered as a possible cause of OOA underestimation by the model over urban and urban-downwind areas (see below) given that COA can be oxidized and form SOA over the urban center and further downwind. Overall, the underestimation of OA over urban locations indicates that global exposure studies (Lelieveld et al., 2015) provide a lower limit of the actual contribution of OA to premature mortality over large urban areas.

Given that the model cannot reproduce the concentrations of POA and SOA over urban locations well due to the coarse spatial resolution and lack of COA emissions, urban locations are not included for the seasonal, continental and total (annual) evaluation of the model presented below. Especially for the seasonal model evaluation, most of the urban field campaigns were conducted either during winter or summer. Therefore, including these locations in our analysis is expected to bias the model performance during winter and summer leading to a potential misinterpretation of the corresponding seasonal results.

5.~~1~~ 2 Spatial evaluation

5.~~1~~2.1 POA

~~AMS observations indicate that over urban areas POA (the sum of HOA and BBOA) has relatively high concentrations while further downwind and in rural areas it decreases substantially due to dilution and evaporation (Figure 10a). The model significantly underpredicts (NMB=-65%, Table 2) the high values of POA over urban areas and especially over densely populated areas such as Beijing, Tokyo and Mexico City (Table S1). However, this underprediction is partly associated with the limited spatial resolution of the model since the size of the grid cell used typically exceeds the size of most urban centers.~~ Over urban-downwind locations, ~~The~~ the model does a better job than urban locations in reproducing the measured POA values ~~over urban-downwind locations~~ (Table 3). This can be verified by focusing on specific field campaigns that provide data from both the urban center and urban-downwind locations over the same period of time (i.e., MILAGRO over Mexico City and MEGAPOLI over Paris). Over these areas the model captures the measured POA concentrations downwind of the urban center (Table ~~S2~~S6; Figure 11b) but it significantly underpredicts the POA concentrations measured in the urban center (Table ~~S1~~S5; Figure 11a). Overall, ~~over~~ in urban-downwind and rural areas the model captures the lower ~~values of~~ POA levels (Figure 10a; Figure 11). Over urban-downwind areas, the model slightly underpredicts POA (NMB=-15%) while over rural areas it overpredicts by 0.04 μg m^{-3} (Table ~~2~~3). However, over rural areas with high BBOA concentrations (e.g, Massognex, Payerne, etc.) the model underpredicts POA (Table ~~S3~~S7; Figure 11c) indicating that biomass burning and/or biofuel use ~~sources over~~in residential areas may be underestimated in the emission inventory.

In Europe, the model underestimates POA concentrations (NMB=-23%; Table 3). However, the comparison of simulated fPOA with AMS HOA (i.e, excluding BBOA from the comparison) suggests that the model overpredicts POA over Europe with a NMB=20% (not shown). This result underscores the underestimated emissions from residential biofuel use as a prominent cause of the model bias over Europe. The possible underestimation of biomass/biofuel burning emissions ~~over~~ in European residential areas has also been ~~recently~~ reported by ~~several~~ other studies (Bergstrom et al., 2012; Kostenidou et al., 2013; Denier van der Gon et al., ~~2014~~2015). Over North America, the model reproduces well the measured HOA (Table 3). Over Asia, the model overestimates the low values of POA measured by AMS (Table 3) mainly due to the high simulated bbPOA concentrations (45% of total POA) transported from the boreal forests of Northeast Asia.

5.~~1~~2.2 SOA

Both AMS and EMAC model results indicate that SOA (~~or~~ and OOA) is high over all ~~types of~~ environments considered (Table ~~3~~4). The highest concentrations are found over urban locations (AMS-OOA= 4.33 μg m^{-3} and EMAC-SOA= 2.97 μg m^{-3}) while further downwind SOA concentrations decrease by 37% over rural locations according to both AMS and EMAC results (Figure 10b). This indicates that the initial emissions of VOCs, IVOCs and SVOCs are photo-oxidized rapidly in the urban environment producing SOA, while their atmospheric aging and further production of SOA is offset by dilution as the air masses ~~moves away~~travel from the urban centers. EMAC does a reasonable job in reproducing SOA concentrations (Table ~~3~~4), however, a~~n~~ systematic underprediction is found in all types of environments. The best model performance is achieved over urban downwind locations (NMB=-25%) followed by urban and rural areas (NMB=-31% and -32%, respectively). ~~Similar to POA, the model underpredicts SOA over densely populated areas such as Beijing and Mexico City (Table S1) due to its limited spatial resolution. In addition, t~~The model significantly underpredicts SOA over specific urban-downwind and rural areas. In most of these cases the field campaign was short (up to 1 week) and the results were subject to specific pollution episodes which cannot be captured by our model (e.g., Puy de Dome, Table ~~S3~~S7; Figure 12c).

Over the continents, the largest SOA underestimation is found over Europe (NMB=-39%). Similar to POA, it is mostly driven by the model underperformance over sites with high biomass burning sources and biofuel use (e.g, Harkingen and Payern, CH; Figure 12). Over North America, the model simulates well the SOA formation with NMB=-15%. IEPOX-SOA, a type of SOA likely formed via processing of later generation isoprene products in aqueous acidic aerosols, has been recently suggested as an important source of SOA close to isoprene emissions (Hu et al., 2015). The model does not simulate SOA formation from aqueous-phase reactions and therefore does not produce IEPOX-SOA, which may lead to an underestimation of SOA over some sites in North America that are strongly influenced by isoprene emissions (e.g. over the Pinnacle state park, NY; Table S7; Figure 12c). Over Asia, the model slightly underestimates SOA with NMB=-22% (Table 4; Figure 12).

In most of the available datasets (~~51~~ 41 out of 84), PMF provides information for the two subtypes of OOA (LV-OOA and SV-OOA). Both PMF and EMAC results indicate that aged SOA (or LV-OOA) is higher than fresh SOA (or SV-OOA) regardless of the type of environment (Tables 5, 6). However, in North America, AMS SV-OOA is slightly higher than LV-OOA while EMAC calculations indicate the opposite (Tables 5, 6). Despite this discrepancy, the model reproduce well both the fresh SOA (NMB=-29%) and aged SOA (NMB=-20%) over North America while over Europe the underestimation is larger (Tables 5, 6) The EMAC performance is better over urban locations where it reproduces the high levels of aged SOA with NMB=-21% and NME=43% (Table ~~4~~2). Over urban-downwind and rural locations EMAC underpredicts aged SOA with NMB=-47% and -38%, respectively (Table ~~4~~5). The performance of the model for fresh SOA is better compared to aged SOA (Table ~~4~~6), with the exception of North America, indicating that the modeled OA aging parameterization may underestimate the SOA produced from ~~atmospheric aging~~chemical reactions during transport and requires improvements. Similar to aged SOA, the best performance of the model for fresh SOA is obtained over urban locations (NMB=-12%).

5.~~2~~ 3 Seasonal evaluation

~~The seasonal model performance evaluation does not include the values over the urban areas because most of the corresponding field campaigns were conducted during winter and summer. In parallel, as discussed above, the model cannot reproduce the concentrations of POA and SOA over urban locations due to its coarse resolution. Therefore including urban locations in our analysis will bias the model performance during winter and summer leading to a potential misinterpretation of the corresponding results regarding the seasonal performance of the model.~~

5.~~2~~3.1 POA

The model performs best during summer (RMSE=0.4, NMB=-3%), followed by autumn (RMSE=0.37, NMB=-15%) and spring (RMSE=0.52, NMB=21%). During winter EMAC underpredicts POA with NMB=-34% (Table ~~2~~3; Figure ~~11a~~13a). This result corroborates our hypothesis that residential biofuel emissions may be underestimated in the inventory since ~~most of the~~ residential heating ~~occurs~~ is expected during winter. Furthermore, since vehicle catalysts require a certain temperature to work to full efficiency, emissions from gasoline and diesel ~~vehicles~~ engines are significantly higher during the warm-up phase of the car (Westerholm et al., 1996). Typically, the additional emissions during the warm-up phase (or cold-start emissions) are not accounted for in emission inventories, which are based on measurements at an ambient temperature of 23 °C (Weilenmann et al., 2009). However, cold-start emissions increase considerably at lower ambient temperatures varying by more than one order of magnitude between 23 and -20 °C (Weilenmann et al., 2009), and thus significant underestimations of OA emissions from the transport sector can be expected during wintertime. Kopacz et al. (2010) provide a global estimate of CO sources by adjoint inversion of satellite datasets and reported an underestimation of CO sources during the winter season due to larger than expected CO emissions from vehicle cold starts and residential heating. Errors in the POA volatility distributions can also explain parts of the model/measurement discrepancy. An overestimation of the fresh POA volatility will favor its evaporation resulting in an underestimation of POA levels by the model. Another source of the POA underestimation ~~of POA~~ by EMAC may be the treatment of wet deposition. The sensitivity of the results to the emission and deposition parameterizations (e.g., the

Henry’s law constants for the organic vapors) will be tested in a subsequent article in preparation.

According to recent studies (Cappa and Wilson, 2012; Aumont et al., 2012; Zhang et al., 2013), not all oxidation products of SVOCs and IVOCs can be assigned to the OOA mass fraction since they are not sufficiently oxidized. Fountoukis et al. (2014) assumed that 50% of the simulated SOA-sv and SOA-iv is still considered as HOA by the AMS analysis and found significant improvements in the view of the modeled bias for POA. In this study we tested this hypothesis and we considered ~~as~~ POA to be the sum of fPOA and bbPOA and ~~the~~ 50% of the SOA-sv and SOA-iv produced from the first oxidation step of SVOCs and IVOCs, respectively. We assumed that SOA-sv and SOA-iv produced during ~~any~~ subsequent oxidation steps together with all the SOA-v are sufficiently oxidized to be considered 100% OOA. Following this hypothesis the model performance improved during winter (NME=55% µg m^{-3} and NMB=-28%) and autumn (NME=50% µg m^{-3} and NMB=1%) and ~~reduced~~ deteriorated during spring (NME=110% µg m^{-3} and NMB=49%) and summer (NME=71% µg m^{-3} and NMB=16%) when the oxidation of SVOCs and IVOCs is enhanced significantly.

5.~~2~~3.2 SOA

The best performance of the model ~~occurs~~ is found for~~during~~ spring (NME=46%, NMB=-24%) followed by the autumn (NME=52%, NMB=-25%) and summer (NME=44%, NMB=-28%) (Table ~~3~~4; Figure ~~11b~~13b). However, during winter the model strongly underpredicts OOA concentrations (NME=80%, NMB=-80%). The overall underprediction of OOA concentrations indicates that the model is missing an important source or formation pathway of SOA. Possible underestimation of residential biofuel emissions in our model, identified during the spatial and seasonal evaluation of simulated POA, can lead to an underestimation of SOA formed from the oxidation of these emissions during winter. Fountoukis et al. (2015) also reported low modeled SOA values compared to AMS OOA over the Paris region and attributed this discrepancy to the transformation of BBOA to OOA without the presence of sunlight reported by some recent studies (Bougiatioti et al., 2014; Crippa et al., 2013b). Underestimation of cold-start vehicle emissions during winter can also lead to a significant underestimation of SOA, since SOA produced from organic compounds emitted during the warm-up phase ~~are~~ can be 3-7 times higher than SOA produced when the catalyst is hot (Gordon et al., 2014). Furthermore, ORACLE assumes that the only source of SOA is the homogeneous gas-phase photochemical oxidation of SOA precursors. Therefore, the negative bias of the model during winter ~~could~~ may also be explained by its inability to simulate SOA formed from aqueous-phase and other heterogeneous reactions, including processes like oligomerization. Such processes should be taking place in all seasons. However, during the photochemically active periods (e.g., summer) 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. Finally, the underprediction of SOA by the model during winter may be also associated with an overestimation of atmospheric removal.

PMF and EMAC results indicate that aged SOA levels exceed those of fresh SOA during all seasons. The EMAC performance for aged SOA appears to be better during spring (NMB=-33%), summer (NMB=-36%), and autumn (NMB=-32%), and much worse during winter (NMB=-91%) (Table ~~4~~5; Figure ~~11c~~13c). The overall performance of the model for fresh SOA (NME=60%, NMB=-30%) (Table ~~5~~6, Figure ~~11d~~13d) appears to be better than aged SOA (NME=71%, NMB=-40%) which supports our conclusions from the spatial model evaluation that the atmospheric aging of SOA may be underestimated by EMAC. However, this apparent discrepancy may be partially due to our assumption that LV-OOA corresponds only to multiple generational SOA. This ~~hypothesis~~ is not consistent with recent studies that reported formation of LV-OOA from the first oxidation step of biogenic VOCs (Ehn et al., 2014). During winter, EMAC also underestimates the fresh SOA levels (NMB=-79%). This underprediction of both fresh and aged SOA during winter suggests that one or more important wintertime SOA formation pathways are missing ~~from~~ in our model.

5.~~3~~ 4 OA composition

According to PMF results, the OOA/OA ratio increases downwind of the urban centers and in rural areas (from 61% over urban environments to 86% over remote areas; Figure ~~12a~~14a). This is ~~in~~ generally consistent with the EMAC predictions. The predicted SOA/OA fraction increases downwind of the urban centers (from 76% over urban locations to 80% over rural areas). This change is lower than the PMF estimates but could be explained by the uncertainty of the PMF analyses (Figure ~~12a~~14a). Alternatively, this may indicate that EMAC tends to underpredict the aging rate of OA. OOA/OA is consistently high during all seasons (around 80%) with the highest ratio predicted in summer (90%) and the lowest in winter (74%) (Figure ~~12b~~14b). The model predicts high SOA/OA during all seasons except winter (Figure ~~12b~~14b). The highest SOA/OA ratio is predicted during summer (87%) when the photo-oxidation of SOA is enhanced. The low SOA/OA during winter (47%) once again shows the inability of EMAC model to reproduce the observed SOA levels during that season.

Both PMF and EMAC indicate that aged SOA is higher than fresh SOA ~~over~~ in all types of environment and seasons (Figure ~~13~~15). PMF results suggest that LV-OOA/OOA is higher over urban-downwind environments (69%), while EMAC aged SOA/SOA is similar over all types of locations (59%) (Figure ~~13a~~15a). The high fresh SOA fraction estimated over rural areas by both PMF and EMAC (around 40%) indicates that fresh SOA production occurs even ~~far away~~remote from the sources. The composition of OOA exhibits a seasonal cycle as well since AMS results indicate that LV-OOA/OOA is higher during winter (73%) and lower during summer (57%) (Figure ~~13b~~15b). EMAC predicts the highest aged SOA/SOA fraction during spring (68%) and the lowest during winter (53%) without any clear seasonal pattern (Figure ~~13b~~15b).

6. Conclusions

This study estimates the impact of open biomass burning and anthropogenic combustion emissions (from fossil and biofuels) of SVOCs and IVOCs to global OA budgets and distributions. The EMAC simulations indicate that the tropospheric burden of OA consists of 32% fOA and 30% bbOA. Furthermore, 90% of fOA and 70% of bbOA is predicted to be secondary. These results support recent findings from global studies that have also reported strong contributions of SOA from anthropogenic sources to global OA concentrations (Spracklen et al., 2011; Carslaw et al., 2013; Lee et al., 2013; Tsimpidi et al., 2014).

The tropospheric burdens of fOA and bbOA follow a clear seasonal pattern. fOA is higher during the boreal summer (0.63 Tg) and lower during winter (0.57 Tg), while bbOA is higher during the dry season in the tropics (1.15 Tg during August) and lower during the wet season (0.17 Tg during April). The simulated spatial distribution of fOA and bbOA is driven by the sources of their precursors and ~~the range of their~~ atmospheric transport. Higher fPOA concentrations occur over densely populated and highly industrialized areas of the Northern Hemisphere while further downwind fPOA decreases substantially due to dilution and evaporation. On the other hand, fSOA ~~remains at high~~maintains similar levels downwind of the anthropogenic sources due to the continued chemical transformations. bbPOA concentrations peak over the tropical and the boreal forests while bbSOA has high concentrations over a wide area covering most of South America, Central and South Africa, Southeastern Asia, including Indonesia and even parts of the Southern Atlantic Ocean.

AMS results from 84 field campaigns performed at continental locations in the Northern Hemisphere during the examined period (2001-2010) ~~are also~~have been used to provide further insights into the composition of OA in three different types of environments: urban, urban-downwind and rural areas, during ~~the~~ four seasons ~~of the year~~. The spatial analysis of AMS and EMAC results indicate that over urban areas POA is highest while further downwind and in rural areas decreases substantially due to dilution and evaporation. On the other hand, SOA is found to be high over all types of environments. This results in an increase of the SOA/OA ratio downwind of the urban centers. The seasonal analysis of the results ~~performed in this study~~ does not include the ~~values over the~~ urban areas since the model cannot reproduce the high OA concentrations over urban environments due to its limited spatial resolution. The seasonal evaluation of the model results against the AMS ~~identified~~ measurements showed a major weakness of the model associated with ~~its failure to reproduce the~~calculated POA and SOA concentration levels during winter. This indicates that the model is probably missing both an important source and a formation pathway of OA, which becomes increasingly important during boreal winter. Possible causes include the underestimation of residential biofuel emissions during winter, the underestimation of vehicle cold-start emissions, the neglect of aqueous-phase and heterogeneous oxidation reactions in the model, and the overestimation of the atmospheric removal of POA and freshly formed SOA.

AMS results indicate that OA consists of 15% HOA and 85% OOA on average during all ~~four~~ seasons. EMAC is able to reproduce this dominance of OOA and its results suggest that SOA accounts for 80% of total OA. At many locations, PMF analysis identified two subtypes of OOA that differ in volatility and oxidation state (LV-OOA and SV-OOA). PMF results indicate that LV-OOA is higher than SV-OOA regardless of the season or the type of environment. The overall LV-OOA/OOA fraction during the four seasons ~~of the year~~ is 63% according to AMS measurement analysis. Assuming that SV-OOA corresponds to fresh SOA (first generation oxidation products) and LV-OOA corresponds to aged SOA (later generation oxidation products), EMAC is able to reproduce the PMF results predicting a dominance of aged SOA during all seasons (59% of the total SOA on average).

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Table 1. Predicted tropospheric burden in Tg of organic aerosol components during the decade 2001-2010.

OA component	**Tropospheric burden** (Tg)	**Monthly Standard Deviation** (σ)
fPOA	0.06	0.01
fSOA	0.57	0.06
bbPOA	0.18	0.13
bbSOA	0.42	0.27
aSOA	0.44	0.08
bSOA	0.31	0.10
OA	1.98	0.54

Table 2. Statistical evaluation of EMAC results against AMS measurements over urban locations of the Northern Hemisphere during 2001-2010.

EMAC Element	**AMS Element**	**Number of datasets**	**Mean Observed** ($\mu g\ m^{-3}$)	**Mean Predicted** ($\mu g\ m^{-3}$)	**MAGE** ($\mu g\ m^{-3}$)	**MB** ($\mu g\ m^{-3}$)	**NME** (%)	**NMB** (%)	**RMSE** ($\mu g\ m^{-3}$)
POA*	HOA+BBOA	23	2.70	0.98	1.73	-1.72	64	-64	2.58
SOA	OOA	23	4.25	2.85	1.97	-1.40	46	-33	2.50
Aged SOA	LV-OOA	10	3.43	2.72	1.47	-0.72	43	-21	2.04
Fresh SOA	SV-OOA	10	2.14	1.88	0.69	-0.26	32	-12	0.81

* Sum of fPOA and bbPOA

Table ~~2~~3. Statistical evaluation of EMAC POA (sum of fPOA and bbPOA) against AMS POA (sum of HOA and BBOA) ~~from 84 data sets worldwide~~ in the Northern Hemisphere during 2001-2010.

Site Type[a]	**Number of datasets**	**Mean Observed** (μg m^{-3})	**Mean Predicted** (μg m^{-3})	**MAGE** (μg m^{-3})	**MB** (μg m^{-3})	**NME** (%)	**NMB** (%)	**RMSE** (μg m^{-3})
~~Urban~~	~~22~~	~~2.74~~	~~0.96~~	~~1.79~~	~~-1.79~~	~~65~~	~~-65~~	~~2.64~~
Urban Downwind	~~14~~15	0.~~69~~82	0.~~59~~64	0.~~35~~38	-0.~~10~~18	~~51~~47	-~~15~~22	0.~~45~~50
Rural/Remote	46	0.43	0.47	0.37	0.04	87	9	0.5
Continent[b]								
Europe	42	0.61	0.47	0.36	-0.14	59	-23	0.47
N. America	10	0.51	0.50	0.29	-0.01	57	-3	0.37
Asia	9	0.15	0.69	0.54	0.54	363	363	0.72
~~**Season**~~[~~b~~]**Season**[c]								
Winter	~~5~~6	~~0.93~~1.18	0.~~61~~74	0.~~51~~60	-0.~~32~~44	~~55~~51	-~~34~~37	0.~~69~~76
Spring	~~31~~30	0.~~44~~42	0.53	0.41	0.~~09~~11	~~94~~97	~~21~~26	0.52
Summer	~~13~~14	0.~~45~~50	0.44	0.30	-0.~~01~~06	~~68~~59	-13	0.~~40~~39
Autumn	11	0.49	0.42	0.27	-0.07	54	-15	0.37
Total	~~60~~61	0.~~49~~53	0.~~50~~51	0.~~37~~38	-0.~~01~~02	~~75~~71	~~1~~-3	0.~~49~~50

[a] Statistics are calculated for a specific site type during all ~~four~~ seasons

[b] Statistics are calculated for a specific continent excluding the values from urban areas

[~~b~~c] Statistics are calculated for a specific season excluding the values from urban areas

Table ~~3~~4. Statistical evaluation of EMAC SOA against AMS OOA ~~from 84 data sets worldwide~~ in the Northern Hemisphere during 2001-2010.

Site Type[a]	**Number of datasets**	**Mean Observed** (μg m^{-3})	**Mean Predicted** (μg m^{-3})	**MAGE** (μg m^{-3})	**MB** (μg m^{-3})	**NME** (%)	**NMB** (%)	**RMSE** (μg m^{-3})
~~Urban~~	~~22~~	~~4.33~~	~~2.97~~	~~1.96~~	~~-1.36~~	~~45~~	~~-31~~	~~2.51~~
Urban Downwind	~~14~~15	2.~~95~~98	2.~~20~~07	1.~~07~~20	-0.~~75~~91	~~.36~~40	-~~25~~30	1.~~64~~77
Rural/Remote	46	2.72	1.86	1.45	-0.86	54	-32	2.09
Continent[b]								
Europe	42	2.47	1.49	1.59	-0.98	64	-39	2.28
N. America	10	3.29	2.78	0.91	-0.51	28	-15	1.37
Asia	9	3.68	2.89	1.00	-0.79	27	-22	1.11
~~**Season**[b]~~**Season**[c]								
Winter	~~5~~6	2.~~72~~81	0.~~55~~50	2.~~17~~31	-2.~~17~~31	~~80~~82	-~~80~~82	2.~~56~~65
Spring	~~31~~30	2.~~38~~22	1.~~80~~79	~~1~~0.~~10~~97	-0.~~58~~43	~~46~~44	-~~24~~20	1.~~47~~18
Summer	~~13~~14	4.~~07~~30	2.~~94~~89	~~1~~2.~~81~~04	-1.~~13~~41	~~44~~47	-~~28~~33	3.~~01~~20
Autumn	11	2.35	1.78	1.22	-0.57	52	-25	1.39
Total	~~60~~61	2.~~77~~78	1.~~94~~91	1.~~36~~39	-0.~~83~~87	~~49~~50	-~~30~~31	2.~~00~~02

[a] Statistics are calculated for a specific site type during all four seasons

[b] Statistics are calculated for a specific continent excluding the values from urban areas

~~[b]~~[c] Statistics are calculated for a specific season excluding the values from urban areas

Table ~~4~~5. Statistical evaluation of EMAC aged SOA against AMS LV-OOA ~~from 51 data sets worldwide~~ in the Northern Hemisphere during 2001-2010.

Site Type[a]	**Number of datasets**	**Mean Observed** ($\mu g\ m^{-3}$)	**Mean Predicted** ($\mu g\ m^{-3}$)	**MAGE** ($\mu g\ m^{-3}$)	**MB** ($\mu g\ m^{-3}$)	**NME** (%)	**NMB** (%)	**RMSE** ($\mu g\ m^{-3}$)
~~Urban~~	~~10~~	~~3.43~~	~~2.72~~	~~1.47~~	~~-0.72~~	~~43~~	~~-21~~	~~2.04~~
Urban Downwind	8	1.77	0.94	1.28	-0.83	72	-47	1.55
Rural/Remote	33	1.65	1.02	1.17	-0.63	71	-38	1.69
Continent[b]								
Europe	35	1.71	0.98	1.24	-0.73	73	-43	0.47
N. America	6	1.45	1.17	0.87	-0.28	60	-20	1.00
Asia	-	-	-	-	-	-	-	-
~~Season~~~~[b]~~**Season**[c]								
Winter	3	2.36	0.20	2.16	-2.16	91	-91	2.36
Spring	~~19~~18	1.~~25~~06	0.~~84~~81	0.~~94~~82	-0.~~41~~25	~~76~~77	-~~33~~24	1.~~25~~03
Summer	~~10~~11	2.~~45~~64	1.~~57~~55	1.~~64~~79	-1.0~~.88~~9	~~67~~68	-~~36~~41	2.~~38~~47
Autumn	9	1.49	1.01	0.89	-0.48	59	-32	1.10
Total	41	1.68	1.01	1.19	-0.67	71	-40	1.67

[a] Statistics are calculated for a specific site type during all four seasons

[b] Statistics are calculated for a specific continent excluding the values from urban areas

[~~b~~c] Statistics are calculated for a specific season excluding the values from urban areas

Table ~~5~~6. Statistical evaluation of EMAC fresh SOA against AMS SV-OOA ~~from 51 data sets worldwide~~ in the Northern Hemisphere during 2001-2010.

Site Type[a]	Number of datasets	Mean Observed (μg m^{-3})	Mean Predicted (μg m^{-3})	MAGE (μg m^{-3})	MB (μg m^{-3})	NME (%)	NMB (%)	RMSE (μg m^{-3})
~~Urban~~	~~10~~	~~2.14~~	~~1.88~~	~~0.69~~	~~-0.26~~	~~32~~	~~-12~~	~~0.81~~
Urban Downwind	8	0.81	0.64	0.41	-0.17	51	-21	0.76
Rural/Remote	33	1.03	0.70	0.64	-0.33	62	-32	0.85
Continent[b]								
Europe	35	0.90	0.63	0.56	-0.27	62	-30	0.81
N. America	6	1.51	1.07	0.80	-0.44	53	-29	0.96
Asia	-	-	-	-	-	-	-	-
~~Season~~[b]Season[c]								
Winter	3	0.87	0.18	0.69	-0.69	79	-79	0.76
Spring	~~19~~18	0.~~65~~54	0.~~40~~37	0.~~53~~46	-0.~~25~~17	~~81~~86	-~~39~~31	0.~~71~~60
Summer	~~10~~11	1.~~81~~89	1.~~25~~22	0.~~88~~96	-0.~~56~~67	~~48~~51	-~~31~~36	1.~~21~~27
Autumn	9	0.83	0.86	0.39	0.03	47	4	0.52
Total	41	0.99	0.69	0.60	-0.30	60	-30	0.83

[a] Statistics are calculated for a specific site type during all four seasons

[b] Statistics are calculated for a specific continent excluding the values from urban areas

[~~b~~c] Statistics are calculated for a specific season excluding the values from urban areas

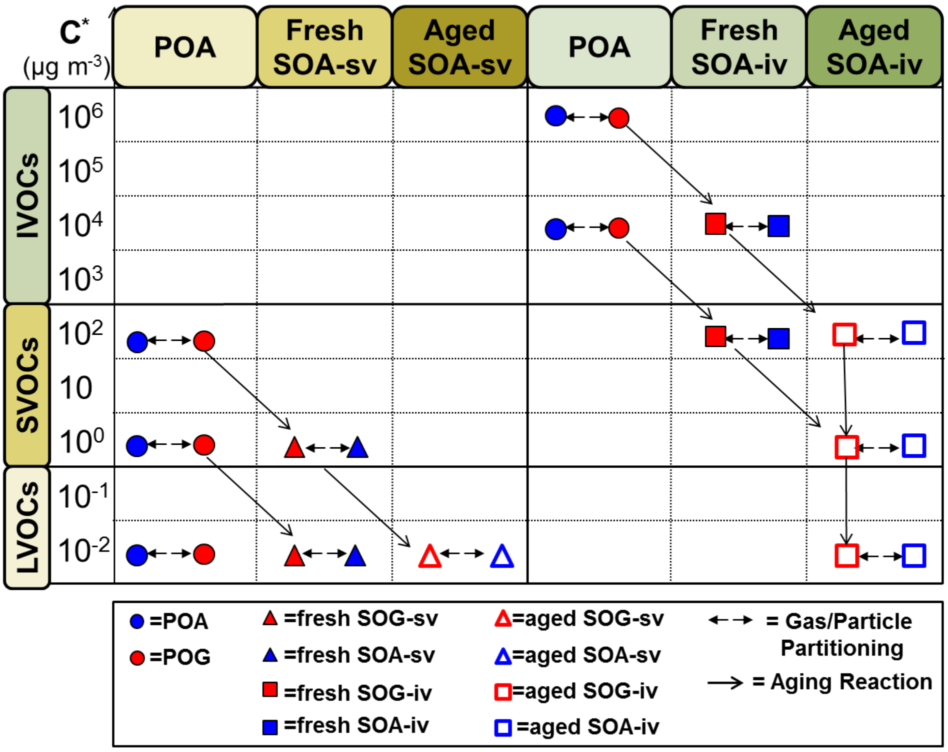

Figure 1: Schematic of the VBS resolution and the formation of SOA from SVOC and IVOC emissions. Red indicates the vapor phase and blue the particulate phase. The circles represent primary organic material that can be emitted either in the gas or in the aerosol phase. Filled triangles and squares indicate the formation of SOA from SVOCs and IVOCs, respectively, by fuel combustion and biomass burning sources from the first oxidation step (fresh SOA). Open triangles and squares represent SOA formed in additional oxidation steps (aged SOA) from SVOCs and IVOCs by the same sources. The partitioning processes, the aging reactions of the organic compounds, and the names of the species used to track all compounds are also shown.

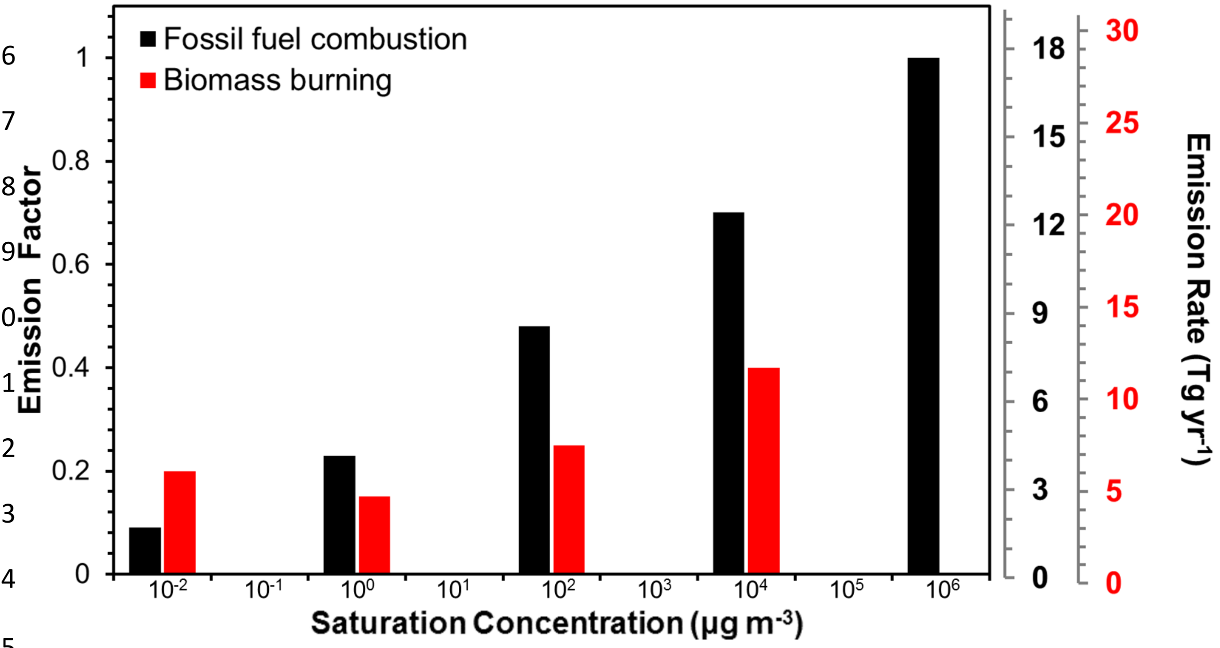

Figure 2. Volatility distribution for fuel combustion (in black) and biomass burning (in red) organic emissions. The emission factors for fuel combustion emissions are derived from Robinson et al. (2007) while for biomass burning POA emissions are from May et al. (2013) (shown in the primary y-axis). The corresponding emission rates are also shown in the secondary y-axis.

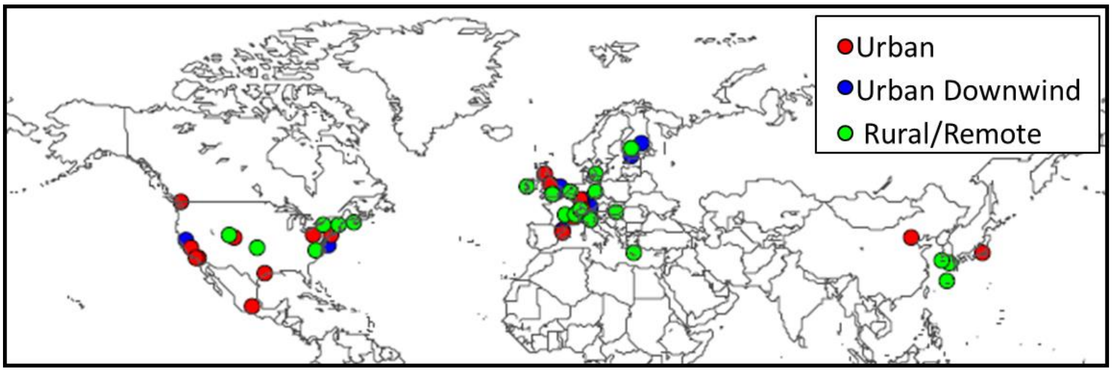

Figure 3: Location of the field measurement campaigns used for evaluating the model during 2001-2010. Urban, urban downwind and rural/remote areas are represented by red, blue, and green colors respectively.

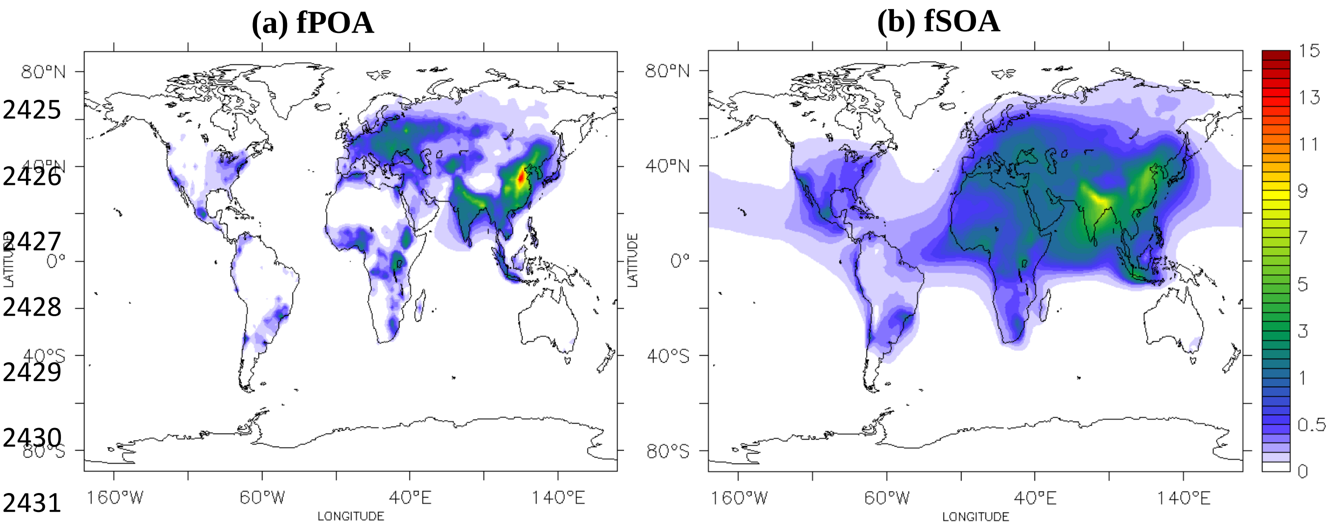

Figure 4: Predicted average surface concentrations (in μg m^{-3}) of: **(a)** POA from fuel combustion sources (fPOA) and **(b)** SOA from the oxidation of SVOCs and IVOCs from fuel combustion sources (fSOA) during the years 2001-2010.

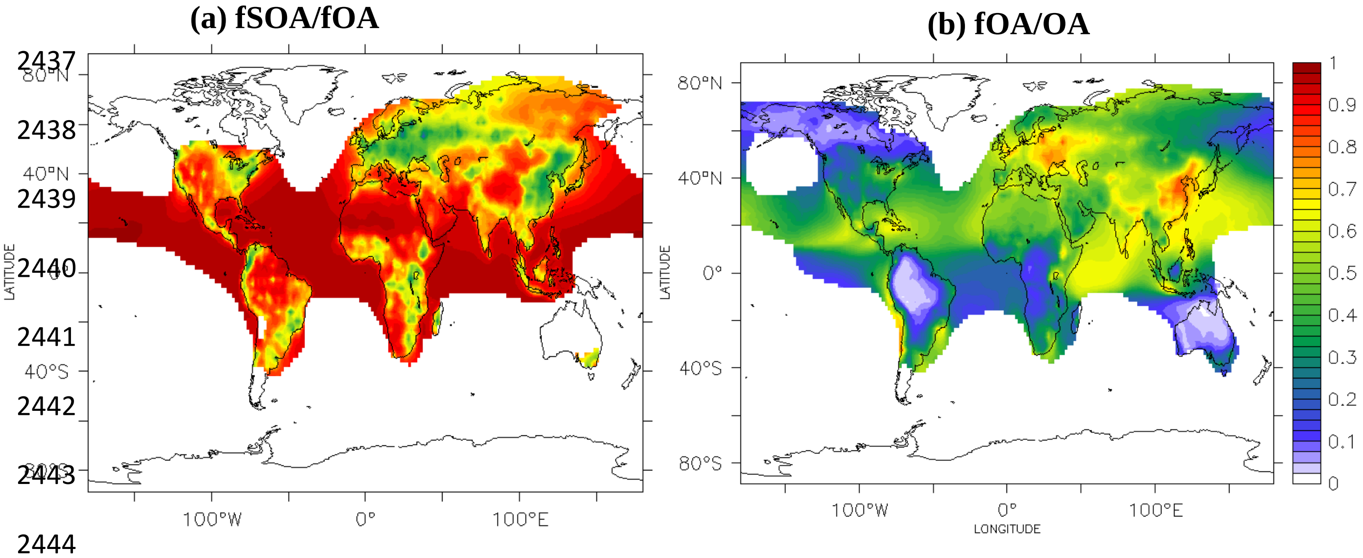

Figure 5: Predicted ratio of **(a)** fuel combustion SOA (fSOA) to total fuel combustion OA (sum of fPOA and fSOA) and **(b)** fuel combustion OA to total OA (sum of fOA, bbOA, aSOA, and bSOA) during the years 2001-2010.

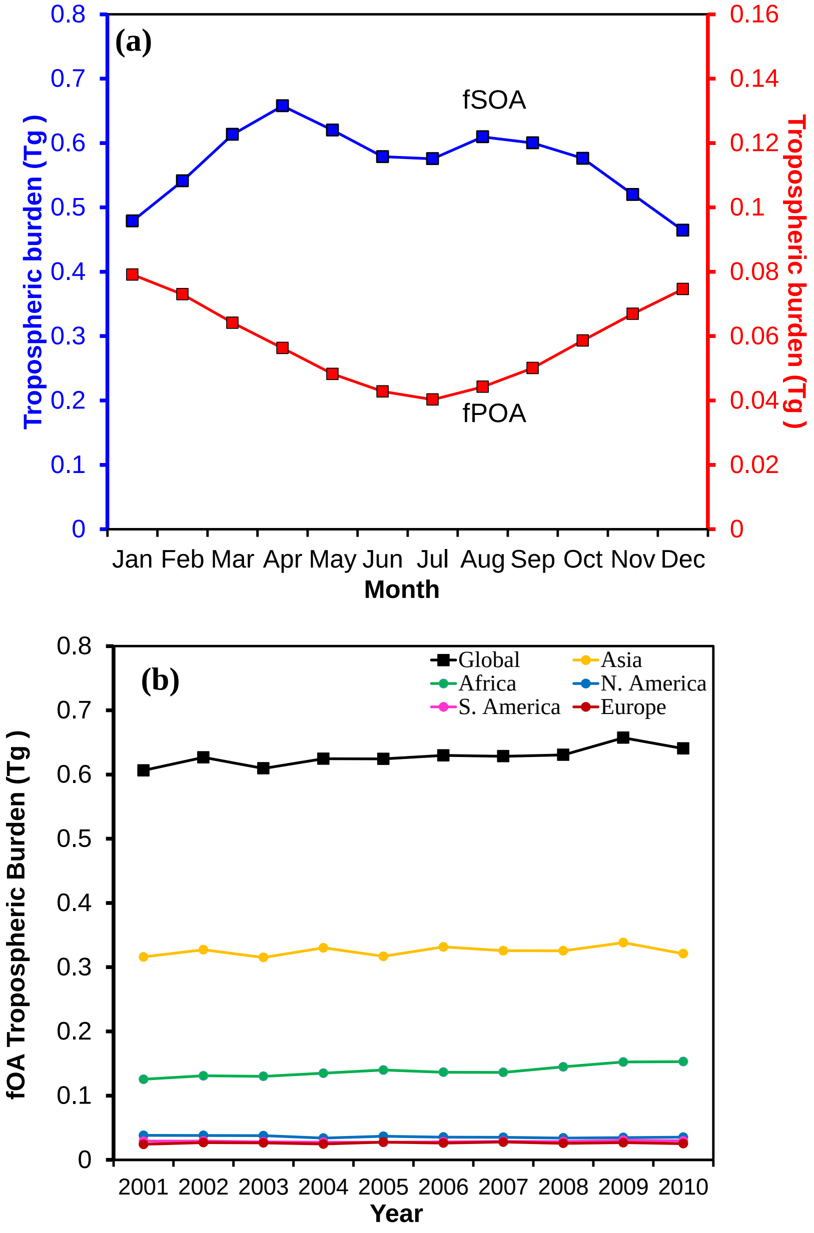

Figure 6: (a) Average predicted tropospheric burden (Tg) of fSOA (in blue, primary y-axis) and fPOA (in red, secondary y-axis) ~~during the year~~ and **(b)** annually averaged tropospheric burden of total fuel combustion OA (fOA) during 2001-2010.

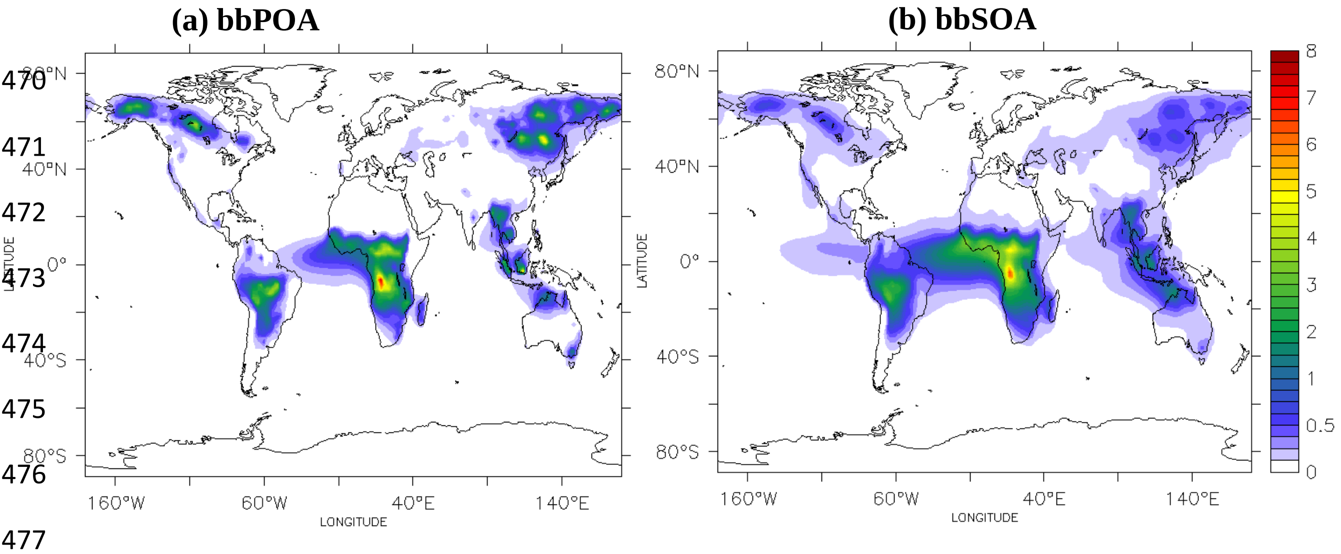

Figure 7: Predicted average surface concentrations (in µg m^{-3}) of: **(a)** POA from biomass burning sources (bbPOA) and **(b)** SOA from the oxidation of SVOCs and IVOCs from biomass burning sources (bbSOA) during the years 2001-2010.

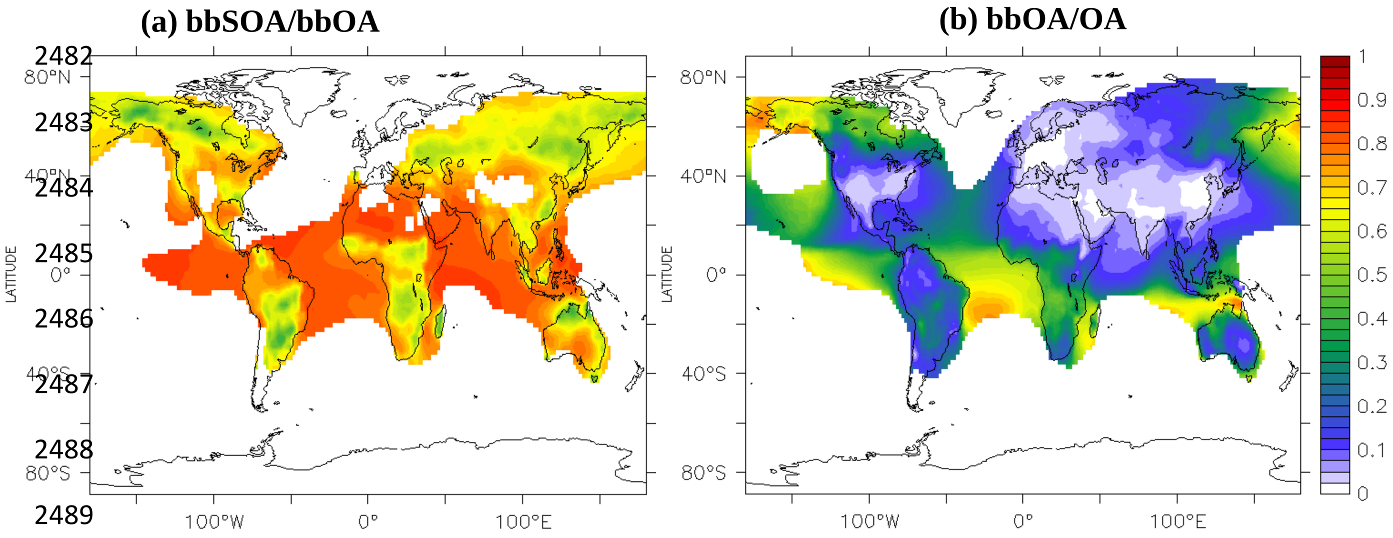

Figure 8: Predicted ratio of **(a)** biomass burning SOA (bbSOA) to total biomass burning OA (sum of bbPOA and bbSOA) and **(b)** biomass burning OA to total OA (sum of fOA, bbOA, aSOA, and bSOA) during the years 2001-2010.

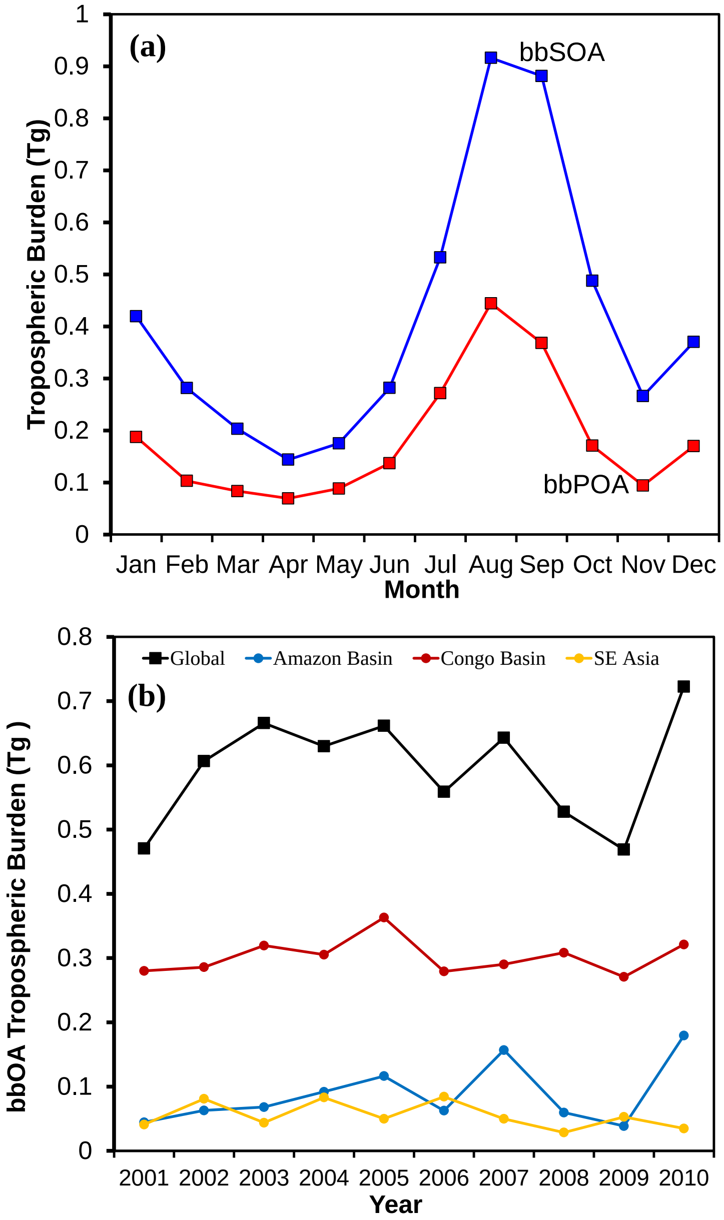

Figure 9: (a) Monthly averaged predicted tropospheric burden (Tg) of bbSOA (in blue) and bbPOA (in red) and **(b)** Annual average tropospheric burden of total biomass burning OA (bbOA) during 2001-2010.

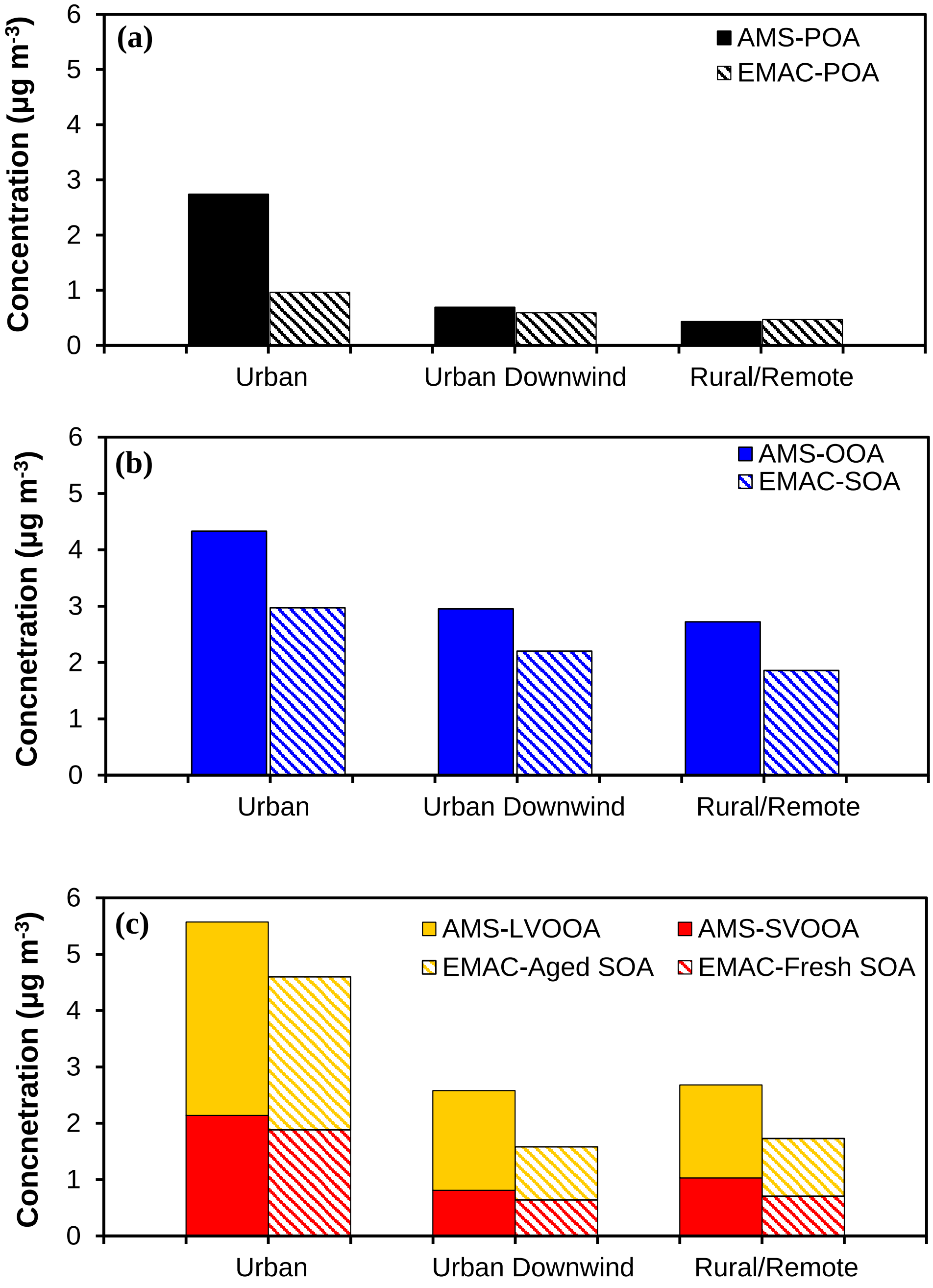

Figure 10: Comparison of average **(a)** EMAC predicted POA ~~against~~ to AMS-POA (sum of AMS-HOA and AMS-BBOA) **(b)** EMAC predicted SOA ~~against~~ to AMS-OOA, and, **(c)** EMAC predicted fresh ~~SOA~~ and aged SOA ~~against~~ to AMS-SVOOA and AMS-LVOOA from 84 data sets ~~worldwide~~ over urban, urban downwind and rural/remote areas during 2001-2010.

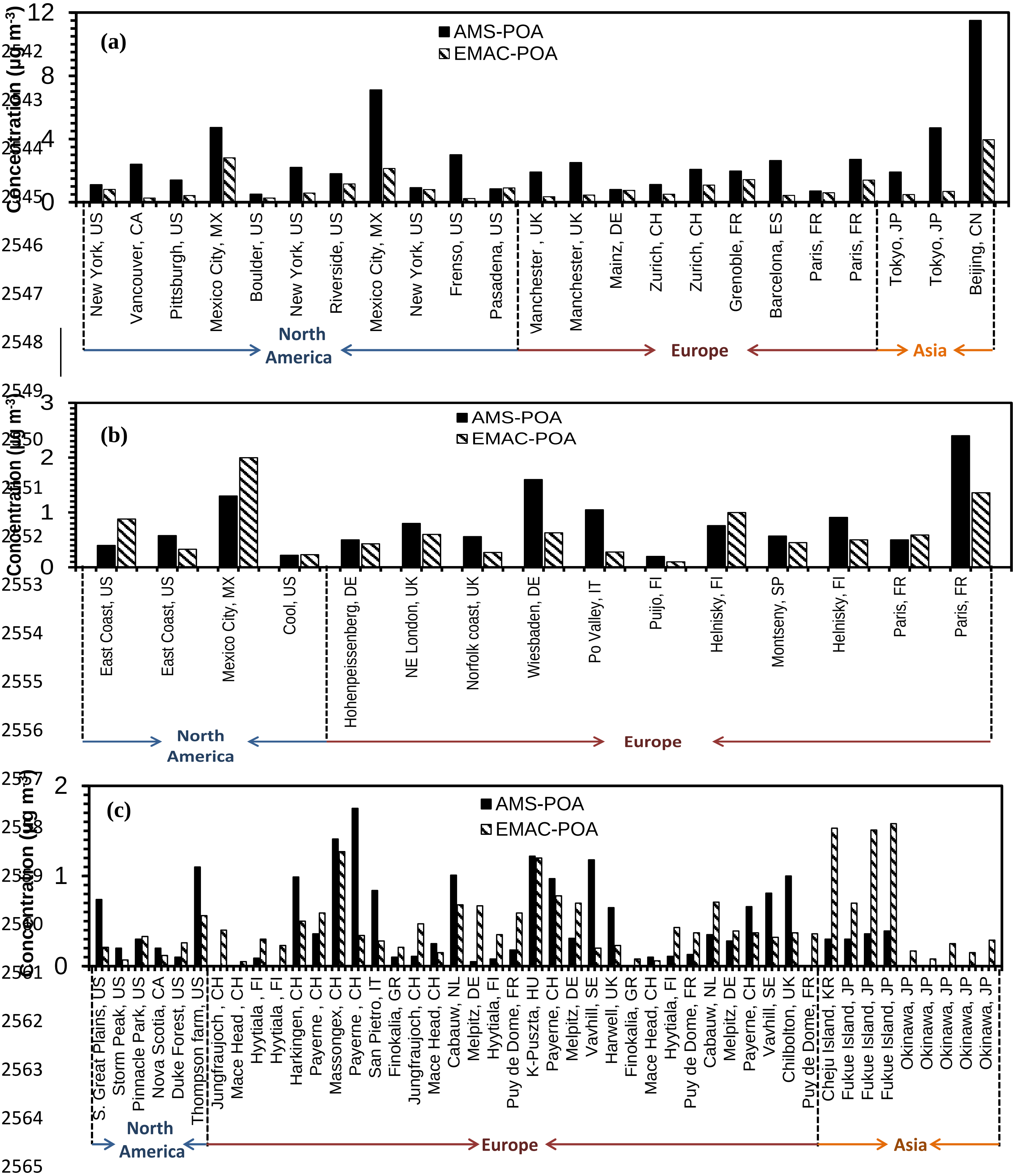

Figure 11: Comparison of EMAC POA (sum of fPOA and bbPOA) to AMS POA (sum of HOA and BBOA) from 84 data sets worldwide over **(a)** urban, **(b)** urban downwind and **(c)** rural/remote areas during 2001-2010.

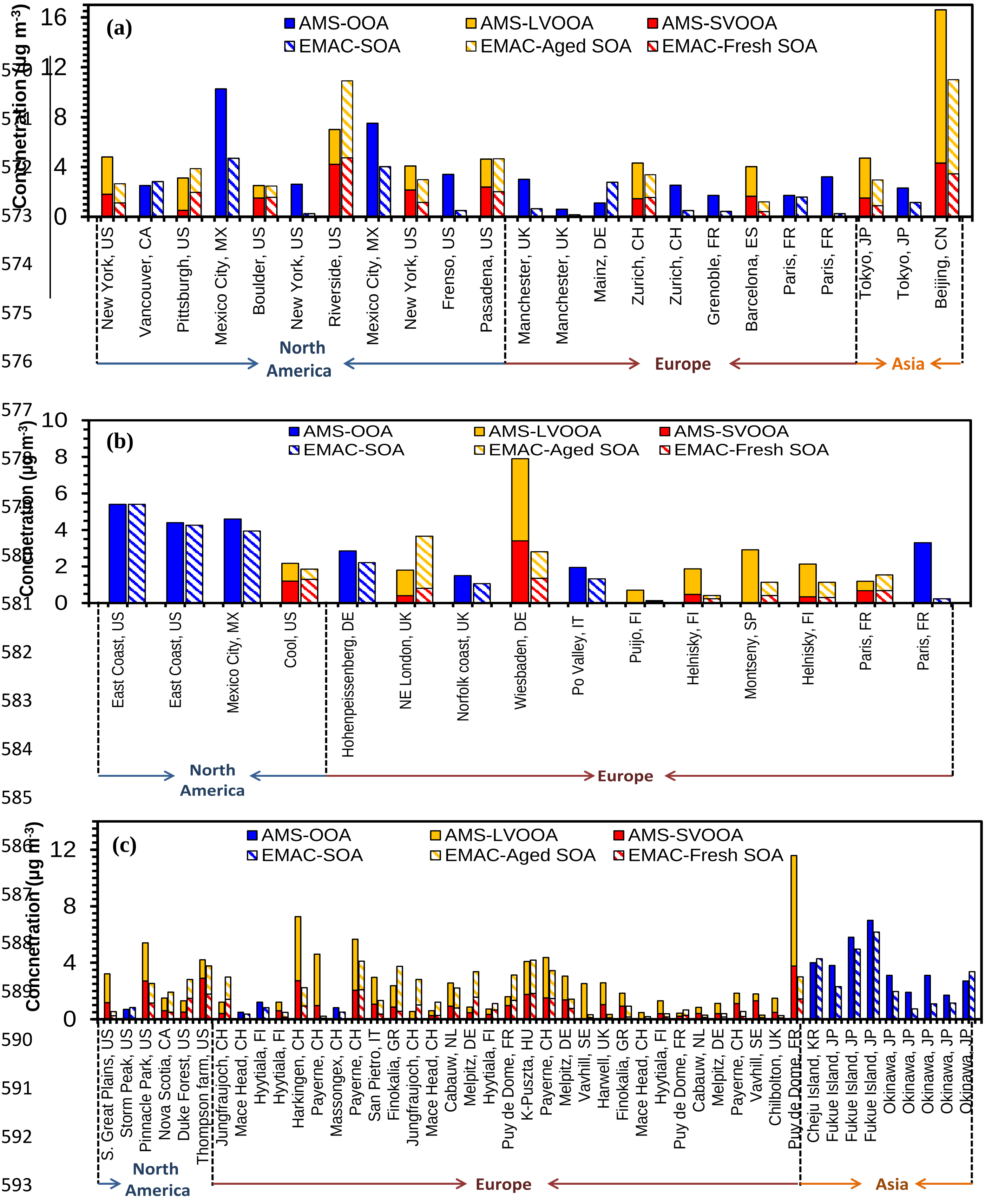

Figure 12: Comparison of EMAC SOA (fresh SOA and aged SOA) to AMS OOA (SV-OOA and LV-OOA) from 84 data sets worldwide over **(a)** urban, **(b)** urban downwind and **(c)** rural/remote areas during 2001-2010.

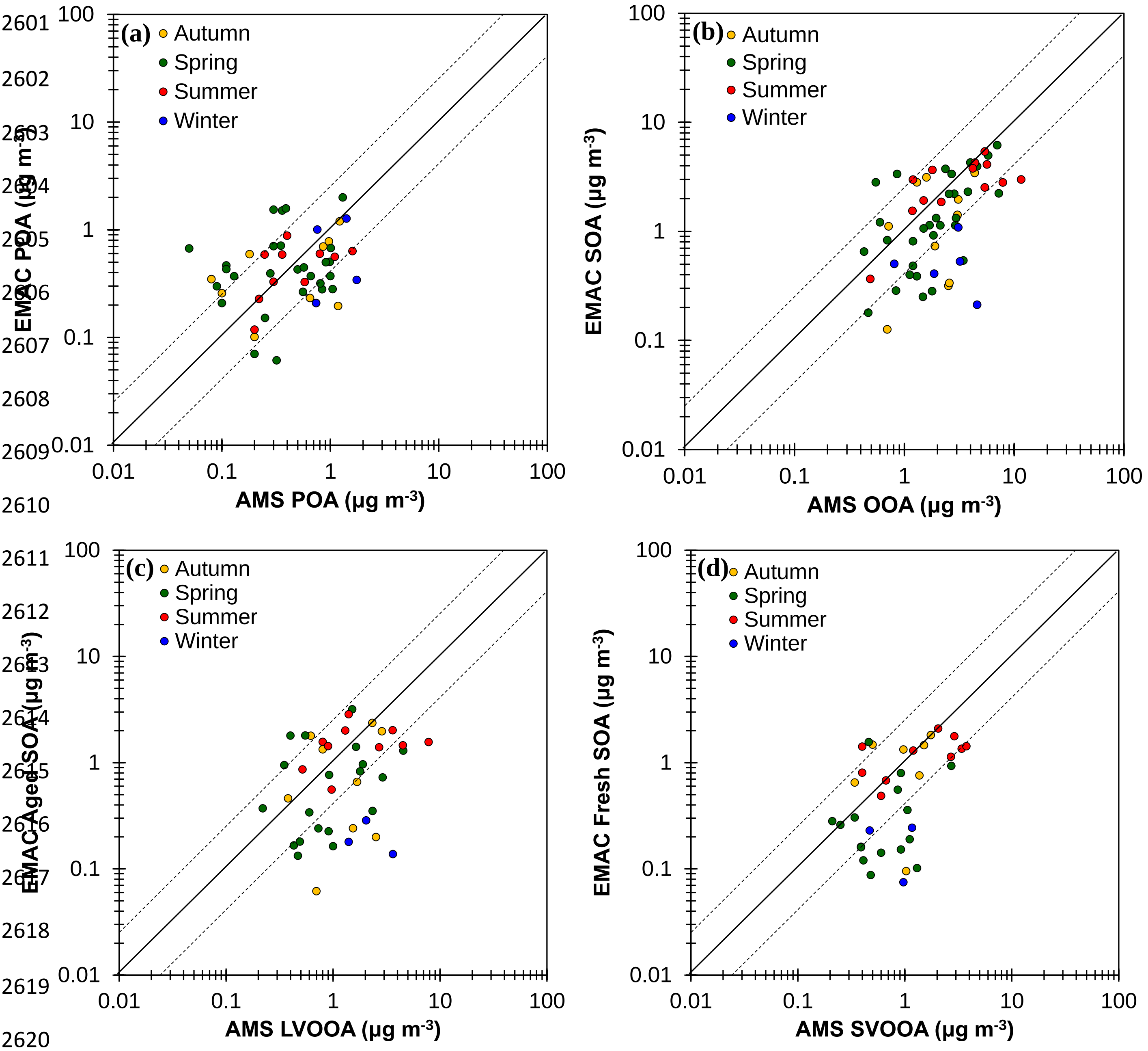

Figure ~~11~~13: Scatterplots comparing model results ~~against~~ to AMS for: **(a)** POA, **(b)** OOA, **(c)** LV-OOA, and **(d)** SV-OOA concentrations (in µg m^{-3}) ~~derived from 84 data sets over~~ in the Northern Hemisphere during 2001-2010. Each point represents the data set average value and is colored based on the season of the field campaign ~~took place~~. Also shown are the 1:1, 2:1, and 1:2 lines.

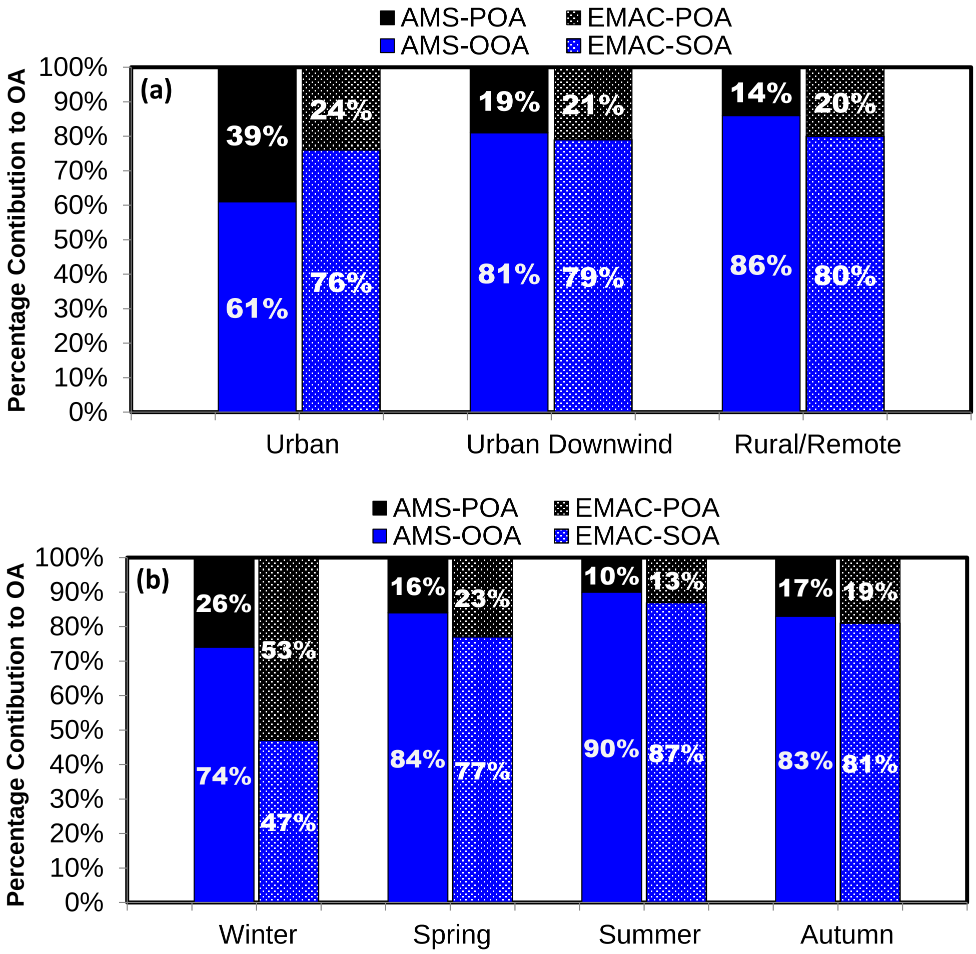

Figure ~~12~~14: (a) Spatial and **(b)** seasonal composition of total OA mass calculated from EMAC and AMS results in the Northern Hemisphere~~for 84 data sets worldwide~~ during 2001-2010.

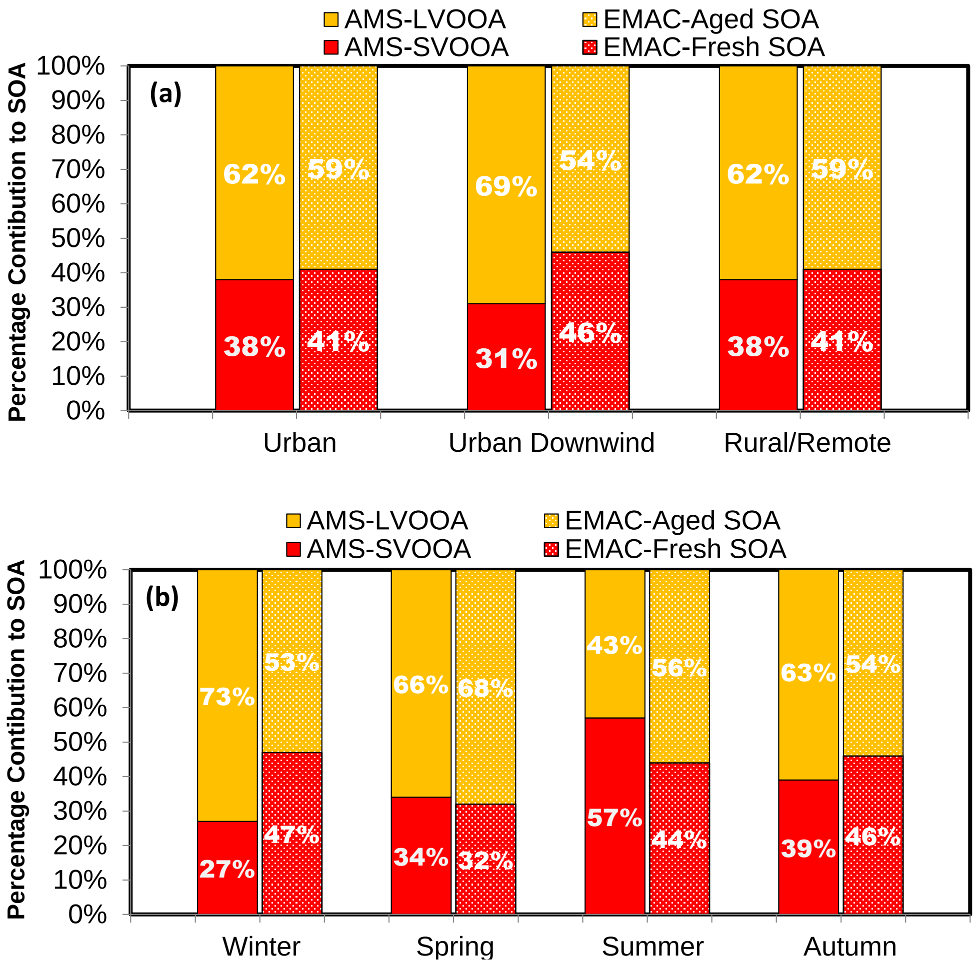

Figure ~~13~~15: (a) Spatial and **(b)** seasonal composition of SOA and OOA mass calculated from EMAC and AMS results, respectively, in the Northern Hemisphere~~for 51 data sets worldwide~~ during 2001-2010.

Supplement of

Global combustion sources of organic aerosols: Model comparison with 84 AMS factor analysis data sets

Tsimpidi et al.

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Table S1. Statistical evaluation of EMAC simulated inorganic gas and aerosol concentrations against monthly average observations from North America during 2001–2010. The units are in ppb for gas and in μg m^{-3} for aerosol species.

EPA Network for gas-phase species and IMPROVE Network for aerosols

Metric	**SO_2**	**NO_2**	**SO_4^{2-}**	**NO_3^-**	**NH_4^+**
Observed	3.35	12.07	1.50	0.71	1.12
Simulated	1.89	4.22	0.83	0.52	0.82
MAGE	2.03	8.07	1.12	0.50	0.54
MB	-1.46	-7.85	-0.67	-0.19	-0.30
NME (%)	61	67	75	70	49
NMB (%)	-44	-65	-44	-26	-27
RMSE	4.27	10.36	1.76	1.06	0.65
Number of comparisons	53094	46250	17129	39056	481

Table S2. Statistical evaluation of EMAC simulated inorganic gas and aerosol concentrations against monthly average observations from Europe during 2001–2010. The units are in µg m^{-3} for both gas and aerosol species.

EMEP Network

Metric	**SO_2**	**NO_2**	**SO_4^{2-}**	**NO_3^-**	**NH_4^+**
Observed	1.54	6.98	1.35	1.12	0.83
Simulated	2.93	7.28	1.00	1.34	0.76
MAGE	1.90	3.58	0.82	1.17	0.48
MB	1.39	0.3	-0.35	0.22	-0.07
NME (%)	123	51	61	105	58
NMB (%)	90	4	-25	19	-8
RMSE	2.97	7.18	1.27	2.14	0.74
Number of comparisons	9130	8626	16707	9307	7787

Table S3. Statistical evaluation of EMAC simulated inorganic gas and aerosol concentrations against monthly average observations from Eastern Asia during 2001–2010. The units are in ppb for gas and in µg m^{-3} for aerosol species.

EANET Network

Metric	**SO_2**	**NO_2**	**SO_4^{2-}**	**NO_3^-**	**NH_4^+**
Observed	2.80	14.40	3.78	1.13	1.05
Simulated	1.74	2.07	1.07	0.75	0.67
MAGE	1.90	12.59	2.84	0.93	0.68
MB	-1.06	-12.33	-2.71	-0.38	-0.38
NME (%)	68	87	75	82	64
NMB (%)	-38	-86	-72	-33	-36
RMSE	3.39	16.94	5.17	2.01	1.74
Number of comparisons	7076	926	2965	2862	2897

Table S4. Secondary organic aerosol mass yield[1] parameters

	C in μg m^{-3} at 298K*			
VOC Species	1	10	10^2	10^3
Benzene, Toluene	0.003	0.165	0.300	0.435
Trimetyl Benzenes, Xylene, and other Aromatics	0.002	0.195	0.300	0.435
Pentanes	0.000	0.038	0.000	0.000
Hexanes, other higher Alkenes	0.000	0.150	0.000	0.000
Propene	0.001	0.005	0.038	0.150
Other Alkenes	0.003	0.026	0.083	0.270
Isoprene	0.009	0.030	0.015	0.000
Monoterpenes	0.107	0.092	0.359	0.600

[1] The SOA yields are based on an assumed particle density of 1.5 g cm^{-3}.

Table ~~S1~~S5. AMS and EMAC average concentrations for OA species over urban areas. In all cases the averages are over all the available data for each campaign. The period and the exact location of the each site are also listed.

Location	Coordinates		Time	AMS					EMAC				Reference
URBAN	**Lat**	**Lon**	**Period**	**HOA**	**BBOA**	**OOA**	**SVOOA**	**LVOOA**	**POA**	**SOA**	**Fresh SOA**	**Aged SOA**	
Manchester , UK	53.5	-2.22	14/06/01-25/06/01	1.9	-	3	-	-	0.35	0.64	-	-	Allan et al., 2003 ; Jimenez et al., 2009
New York , US	40.74	-73.92	30/06/01-5/08/01	1.1	-	4.8	1.8	3	0.81	2.64	1.11	1.53	Drewnick et al. 2004; Jimenez et al., 2009
Vancouver, CA	49.25	-123.13	11/08/01-24/08/01	2.4	-	2.5	-	-	0.26	2.81	-	-	Boudries et al., 2004; Jimenez et al., 2009
Manchester , UK	53.5	-2.22	17/01/02-28/01/02	1	1.5	0.6	-	-	0.45	0.15	-	-	Allan et al., 2003; Jimenez et al., 2009
Pittsburgh, US	40.44	-79.94	06/09/02-22/09/02	1.4	-	3.1	0.5	2.6	0.42	3.86	1.96	1.89	Zhang et al., 2004; Jimenez et al., 2009
Mexico City , MX	19.48	-99.15	13/04/03-16/04/03	3.24	1.48	10.26	-	-	2.81	4.69	-	-	Salcedo et sl., 2006; Tsimpidi et al., 2010
Boulder, US	40.02	-105.27	07/06/03-20/06/03	0.5	-	2.5	1.5	1	0.26	2.46	1.56	0.90	Nemitz et al., 2008; Jimenez et al., 2009
Tokyo , JP	35.67	139.75	23/07/03-14/08/03	1.9	-	4.7	1.5	3.2	0.48	2.95	0.89	2.06	Takegawa et al., 2005; Jimenez et al., 2009
New York , US	40.74	-73.92	07/01/04-06/02/04	2.2	-	2.6	-	-	0.58	0.24	-	-	Drewnick et al. 2004; Jimenez et al., 2009
Tokyo , JP	35.67	139.75	20/01/04-10/02/04	3.7	1	2.3	-	-	0.68	1.14	-	-	Takegawa et al., 2006; Jimenez et al., 2009
Mainz, DE	49.98	8.23	16/09/04-01/10/04	0.8	-	1.1	-	-	0.74	2.76	-	-	Vester et al. 2007; Jimenez et al., 2009

Table ~~S1~~S5. Continued

~~Location~~	Coordinates		Time	AMS					EMAC				Reference
URBAN	**Lat**	**Lon**	**Period**	**HOA**	**BBOA**	**OOA**	**SVOOA**	**LVOOA**	**POA**	**SOA**	**Fresh SOA**	**Aged SOA**	
Riverside, US	33.95	-117.4	14/07/05-13/08/05	1.2	0.6	7	4.2	2.8	1.16	10.9	4.73	6.17	De Carlo et al., 2006; Jimenez et al., 2009
Zurich , CH	47.4	8.5	14/07/05-04/08/05	0.46	0.65	4..31	1.44	2.87	0.5	3.37	1.55	1.82	Lanz et al. 2007; Lanz et al. 2010
Zurich , CH	47.4	8.5	06/01/06-25/01/06	0.32	1.75	2.53	-	-	1.08	0.49	-	-	Lanz et al. 2008; Lanz et al. 2010
Mexico City , MX	19.48	-99.15	10/03/06-30/03/06	4.6	2.5	7.5	-	-	2.13	4.01	-	-	Aiken et al., 2008; Aiken et al., 2009
Beijing, CN	40.0	116.0	09/07/06-21/07/06	11.5	-	16.6	4.3	12.3	3.95	11	3.45	7.54	Sun et al., 2009; Jimenez et al., 2009
Grenoble, FR	45.18	5.73	14/01/09-30/01/09	0.4	1.56	1.7	-	-	1.43	0.43	-	-	Favez et al., 2010; Lanz et al. 2010
Barcelona, ES	41.39	2.12	25/02/09-26/03/09	1.97	0.66	4.02	1.64	2.38	0.43	1.19	0.41	0.78	Mohr et al., 2012; Crippa et al., 2014
New York , US	40.74	-73.92	13/07/09-03/08/09	0.91	-	4.07	2.14	1.93	0.8	2.98	1.15	1.82	Sun et al., 2011
Paris, FR	48.83	2.36	01/07/09-31/07/09	0.7	-	1.7	-	-	0.6	1.58			Crippa et al., 2013a; Fountoukis et al., 2015
Paris, FR	48.83	2.36	12/01/10-16/02/10	2.7	-	3.2	-	-	1.4	0.24	-	-	Crippa et al., 2013b; Fountoukis et al., 2015
Fresno, US	36.81	-119.78	09/01/10-23/01/10	1.74	1.26	3.4	-	-	0.23	0.49	-	-	Ge et al. 2012
Pasadena, US	34.14	-118.12	15/05/10-16/06/10	0.84	-	4.62	2.38	2.24	0.9	4.64	2.00	2.64	Hayes et al. 2013

Table ~~S2~~S6. AMS and EMAC average concentrations for OA species over urban downwind areas. In all cases the averages are over all the available data for each campaign. The period and the exact location of the each site are also listed.

Location	**Coordinates**		**Time period**	**AMS**					**EMAC**				**Reference**
Urban Downwind	**Lat**	**Lon**		**HOA**	**BBOA**	**OOA**	**SVOOA**	**LVOOA**	**POA**	**SOA**	**fresh SOA**	**aged SOA**	
Hohenpeissenberg, DE	47.8	11.0	19/05/02-31/05/02	0.5	-	2.85	-	-	0.43	2.21	-	-	Hock et al., 2008; Lanz et al., 2010
East coast 1, US	37.95	-74.3	18/07/02-26/07/02	0.4	-	5.4	-	-	0.88	5.40	-	-	DeGouw et al., 2005; Zhang et al., 2007
East coast 2, US	37.95	-74.3	29/07/02-10/08/02	0.58	-	4.4	-	-	0.33	4.26	-	-	DeGouw et al., 2005; Zhang et al., 2007
NE London, UK	51.7	0.4	29/07/03-31/08/03	0.8	-	1.8	0.4	1.4	0.60	3.66	0.80	2.85	Cubison et al., 2006; Jimenez et al., 2009
Norfolk coast, UK	53.0	1.1	25/04/04-26/05/04	0.56	-	1.5	-	-	0.27	1.06	-	-	Gysel et al., 2007; Zhang et al., 2007
Wiesbaden, DE	50.22	8.45	14/07/04-04/08/04	1.6	-	7.9	3.4	4.5	0.63	2.8	1.35	1.46	Dusek et al., 2006; Jimenez et al., 2009
Mexico City, MX	19.7	-98.98	10/03/06-30/03/06	1.3	-	4.6	-	-	2	3.94	-	-	Aiken et al., 2008; Tsimpidi et al., 2011
Po Valley, IT	44.65	11.62	30/03/08-20/04/08	0.79	0.26	1.95	-	-	0.28	1.32	-	-	Saarikoski et al. 2012
Puijo, FI	62.90	27.65	27/09/08-21/10/08	0.2	-	0.7	0	0.7	0.10	0.13	0.07	0.06	Crippa et al., 2014
Helsinki 1, FI	60.2	24.95	09/01/09-13/03/09	0.41	0.35	1.87	0.47	1.4	1.00	0.41	0.23	0.18	Carbone et al., 2014
Montseny, SP	41.76	2.4	21/02/09-27/03/09	0.25	0.32	2.91	0	2.91	0.45	1.13	0.41	0.73	Crippa et al., 2014
Helsinki 2, FI	60.2	24.95	09/04/09-08/05/09	0.34	0.57	2.13	0.34	1.79	0.5	1.13	0.30	0.83	Timonen et al., 2012
Paris, FR	48.72	2.21	01/07/09-31/07/09	0.5	-	1.19	0.67	0.52	0.59	1.54	0.68	0.86	Crippa et al., 2013a
Paris, FR	48.72	2.21	12/01/10-16/02/10	2.4	-	3.3	-	-	1.36	0.23	-	-	Crippa et al., 2013b; Fountoukis et al., 2015
Cool, US	38.88	-121.0	02/06/10-28/06/10	0.22	-	2.17	1.2	0.97	0.23	1.86	1.30	0.56	Setyan et al., 2012

Table ~~S3~~S7. AMS and EMAC average concentrations for OA species over rural/remote areas. In all cases, the averages are over all the available data for each campaign. The period and the exact location of the each site are also listed.

Location	**Coordinates**		**Time Period**	**AMS**					**EMAC**				**Reference**
Rural/Remote	**Lat**	**Lon**		**HOA**	**BBOA**	**OOA**	**SVOOA**	**LVOOA**	**POA**	**SOA**	**Fresh SOA**	**Aged SOA**	
Cheju Island, KR	33.51	126.50	11/04/01-30/04/01	0.3	-	4	-	-	1.53	4.27	-	-	Topping et al. 2004; Jimenez et al.,2009
Jungfraujoch , CH	46.3	7.6	28/06/02-17/07/02	0	-	1.2	0.4	0.8	0.40	3	1.41	1.57	Jimenez et al.,2009
Mace Head , CH	53.3	-9.8	01/08/02-31/08/02	0	-	0.49	-	-	0.05	0.37	-	-	Alfarra et al., 2004; Zhang et al., 2007
Fukue Island , JP	32.69	128.84	18/03/03-03/04/03	0.3	-	3.8	-	-	0.7	2.30	-	-	Takami et al., 2005; Zhang et al., 2007
Fukue Island , JP	32.69	128.84	03/04/03-16/04/03	0.36	-	5.8	-	-	~~0~~1.51	4.96	-	-	Takami et al., 2005; Zhang et al., 2007
Hyytiala , FI	61.8	24.3	19/03/03-13/04/03	0.09	-	1.2	-	-	0.3	0.81	-	-	Zhang et al., 2007
Fukue Island , JP	32.69	128.84	10/05/03-03/06/03	0.39	-	7	-	-	~~0~~1.58	6.17	-	-	Takami et al., 2005; Zhang et al., 2007
Okinawa , JP	26.87	128.25	03/10/03-28/10/03	0	-	3.1	-	-	0.17	1.96	-	-	Takami et al., 2005; Zhang et al., 2007
Okinawa , JP	26.87	128.25	28/10/03-02/12/03	0	-	1.9	-	-	0.08	0.73	-	-	Takami et al., 2005; Zhang et al., 2007
Okinawa , JP	26.87	128.25	02/12/03-24/12/03	0	-	3.1	-	-	0.25	1.09	-	-	Takami et al., 2005; Zhang et al., 2007
Okinawa , JP	26.87	128.25	13/03/04-08/04/04	0	-	1.7	-	-	0.15	1.14	-	-	Takami et al., 2005; Zhang et al., 2007
Okinawa , JP	26.87	128.25	08/04/04-27/04/04	0	-	2.7	-	-	0.29	3.36	-	-	Takami et al., 2005; Zhang et al., 2007

Table ~~S3~~S7. Continued

Location	Coordinates		Time period	AMS					EMAC				Reference
Rural/Remote	**Lat**	**Lon**		**HOA**	**BBOA**	**OOA**	**SVOOA**	**LVOOA**	**POA**	**SOA**	**Fresh SOA**	**Aged SOA**	
Storm Peak, US	40.45	-106.7	04/04/04-02/05/04	0.2	-	0.7	-	-	0.07	0.83	-	-	Richardson et al.,2007; Jimenez et al., 2009
Pinnacle Park, US	43.0	-76	14/07/04-05/08/04	0.3	-	5.4	2.7	2.7	0.33	2.53	1.13	1.39	Bae et al. 2007; Jimenez et al., 2009
Nova Scotia, CN	43.76	-66.1	07/07/04-14/08/04	0.2	-	1.5	0.6	0.9	0.12	1.92	0.49	1.43	Holzinger et al., 2007; Jimenez et al., 2009
Duke Forest, US	35.97	-79.1	13/09/04-21/09/04	0.1	-	1.3	0.5	0.8	0.26	2.81	1.48	1.33	Jimenez et al.,2009
Hyytiala , FI	61.8	24.3	31/03/05-15/04/05	0	-	1.2	0.6	0.6	0.23	0.48	0.14	0.34	Allan et al. 2006; Jimenez et al., 2009
Harkingen, CH	40.74	7.82	12/05/05-30/05/05	0.99	-	7.26	2.72	4.54	0.5	2.23	0.93	1.30	Lanz et al., 2010
Thompson, US	43.11	-70.95	09/07/05-15/08/05	1.1	-	4.2	2.9	1.3	0.56	3.78	1.77	2.01	Cottrell et al., 2008; Jimenez et al., 2009
Payerne , CH	46.8	6.95	31/05/06-03/07/06	0.36	-	4.6	0.97	3.63	0.59	0.21	0.07	0.14	Lanz et al. 2010
Massongex, CH	46.24	6.14	23/11/06-17/12/06	0.31	1.1	0.81	-	-	1.27	0.50	-	-	Lanz et al. 2010
Payerne , CH	46.8	6.95	12/01/07-17/02/07	0.65	1.1	5.66	2.05	3.61	0.34	4.1	2.09	2.02	Lanz et al. 2010
San Pietro, IT	44.65	11.65	31/03/08-21/04/08	0.38	0.46	2.96	1.06	1.9	0.28	1.32	0.36	0.96	Saarikoski et al. 2012; Crippa et al., 2014

Table ~~S3~~S7. Continued

Location	Coordinates		Time period	AMS					EMAC				Reference
Rural/Remote	**Lat**	**Lon**		**HOA**	**BBOA**	**OOA**	**SVOOA**	**LVOOA**	**POA**	**SOA**	**Fresh SOA**	**Aged SOA**	
Finokalia , GR	35.33	25.66	09/05/08-05/06/08	0.1	-	2.37	0.86	1.51	0.21	3.74	0.56	3.18	Hildebrandt et al. 2010; Crippa et al.,2014
Jungfraujoch , CH	46.3	7.6	01/05/08-29/05/08	0.04	0.07	0.55	0	0.55	0.47	2.81	1.01	1.80	Crippa et al.,2014
Mace Head , CH	53.3	-9.8	16/05/08-13/06/08	0.11	0.14	0.6	0.25	0.35	0.15	1.21	0.26	0.95	Dall'Osto et al., 2010; Crippa et al.,2014
Cabauw , NL	51.97	4.93	29/04/08-30/05/08	0.59	0.42	2.56	0.92	1.64	0.68	2.2	0.80	1.41	Mensah et al., 2012 ; Crippa et al.,2014
Melpitz , DE	51.54	12.93	24/05/08-09/06/08	0.05	-	0.86	0.46	0.4	0.67	3.36	1.56	1.80	Poulain et al., 2011; Crippa et al.,2014
Hyytiala , FI	61.8	24.3	11/09/08-15/10/08	0.05	0.03	0.72	0.34	0.38	0.35	1.12	0.65	0.46	Crippa et al. 2014
Puy de Dome , FR	45.77	2.97	14/09/08-18/10/08	0.02	0.16	1.59	0.97	0.62	0.59	3.12	1.33	1.79	Freney et al, 2011; Crippa et al.,2014
K-Puszta, HU	46.96	19.58	17/09/08-15/10/08	0.64	0.58	4.08	1.75	2.33	1.2	4.18	1.82	2.37	Crippa et al., 2014
Payerne , CH	46.8	6.95	18/09/08-16/10/08	0.32	0.65	4.37	1.51	2.86	0.78	3.43	1.46	1.97	Crippa et al. 2014
Melpitz , DE	51.54	12.93	17/09/08-03/11/08	0.31	0.55	3.05	1.37	1.68	0.7	1.42	0.76	0.66	Poulain et al. 2011; Crippa et al.,2014
Vavihill , SE	56.02	13.15	01/10/08-07/11/08	0.74	0.44	2.52	0	2.52	0.2	0.32	0.12	0.20	Crippa et al., 2014

Table ~~S3~~S7. Continued

Location	Coordinates		Time period	AMS					EMAC				Reference
Rural/Remote	**Lat**	**Lon**		**HOA**	**BBOA**	**OOA**	**SVOOA**	**LVOOA**	**POA**	**SOA**	**Fresh SOA**	**Aged SOA**	
Harwell, UK	51.15	-1.44	20/09/08-22/10/08	0.23	0.42	2.57	1.03	1.54	0.23	0.34	0.10	0.24	Crippa et al., 2014
Finokalia , GR	35.33	25.66	27/02/09-26/03/09	0	-	1.84	0.92	0.92	0.08	0.92	0.15	0.77	Hildebrandt et al. 2011; Crippa et al., 2014
Mace Head , CH	53.3	-9.8	27/02/09-27/03/09	0.1	0.22	0.47	0	0.47	0.06	0.18	0.05	0.13	Dall'Osto et al., 2010; Crippa et al., 2014
Hyytiala , FI	61.8	24.3	04/03/09-01/04/09	0.04	0.07	1.3	0.39	0.91	0.43	0.4	0.16	0.23	Crippa et al., 2014
Puy de Dome , FR	45.77	2.97	23/02/09-10/03/09	0.03	0.1	0.43	0.21	0.22	0.37	0.65	0.28	0.37	Freney et al, 2011; Crippa et al., 2014
Cabauw , NL	51.97	4.93	25/02/09-25/03/09	0.23	0.12	0.84	0.41	0.43	0.71	0.29	0.12	0.17	Crippa et al., 2014
Melpitz , DE	51.54	12.93	10/03/09-27/03/09	0.13	0.15	1.12	0.39	0.73	0.39	0.4	0.16	0.24	Poulain et al. 2011; Crippa et al., 2014
Payerne , CH	46.8	6.95	01/03/09-24/03/09	0.29	0.37	1.84	1.11	0.73	0.37	0.54	0.19	0.35	Crippa et al., 2014
Vavihill , SE	56.02	13.15	26/02/09-02/04/09	0.55	0.26	1.79	1.3	0.49	0.32	0.28	0.10	0.18	Crippa et al., 2014
Chilbolton, UK	51.1	-1.4	25/02/09-26/03/09	0.5	0.5	1.48	0.48	1	0.37	0.25	0.09	0.16	Crippa et al., 2014
Puy de Dome , FR	45.77	2.97	22/06/10-29/06/10	0	-	11.58	3.76	7.82	0.36	2.99	1.43	1.56	Freney et al, 2011
S. Great Plains, US	36.61	-97.49	01/12/10-31/12/10	0	0.74	3.21	1.17	2.04	0.21	0.53	0.24	0.29	Parworth et al., 2015

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