Anonymous Referee #2

General comments

1. *The paper by Tsimpidi et al. describes the organic aerosol module ORACLE, coupled with the global model EMAC, which takes into account the semi-volatility of all organic aerosols, both primary and secondary, from anthropogenic and biogenic sources, including the IVOCs. The module is flexible in design, both with regard to the amount of volatility bins used, and the processes included, like aging. The paper is very clearly written and the module is thoroughly documented. I recommend publication after addressing the following minor points.*

We would like to thank the referee for the positive response and for raising important issues after thoughtful review.

Minor comments

2. *Although the model includes aerosol microphysics that take into account the aerosol hygroscopicity, no discussion is made on size-resolved and hygroscopicity results. I understand that this might be outside the scope of the paper, especially if evaluation has to come into play, but some basic discussion is missing. This includes questions like: Which mode has most of the aerosol mass? Which species condenses the most in each mode? How do organic aerosols affect the hygroscopicity of the modes? What are the assumptions made for the hygroscopicity of each volatility bin? Does hygroscopicity play a role on the size-dependence partitioning described in section 3.6? Is there any link (in the model) between hygroscopicity and volatility?*

In ORACLE, the user is allowed to use up to 3 hydrophilic modes (Aitken, accumulation, coarse) for the size distribution of all OA surrogate species. In the current application, we used only one mode (accumulation) in order to limit the computational cost of the module. During a two-year simulation that we conducted as a sensitivity test by using all 3 modes, the model predicted that 70% of the total OA existed in the accumulation mode. Currently, all OA surrogate species are assumed to have a constant hygroscopicity parameter of 0.14. The total hygroscopicity of the aerosol is estimated based on a simple mixing rule. The above information has been added to the text.

3. *Abstract, line 18: "domain-average" refers to which domain?*

The domain-average results refer to global averages. In the revised manuscript "the domain-average global surface OA concentration" has been replaced with "the global surface average OA concentration".

4. *p. 5470, l. 20-21: One year of spinup is probably not enough for semi-volatile organics that tend to accumulate in the upper layers of the troposphere where temperatures are very low, and their lifetime is higher since they are above clouds. This can be tested by looking at the organics optical depth, or upper tropospheric burden, as a function of time.*

We tested the middle-upper tropospheric burden (above 840 mb) as a function of time and we found that one year is satisfactory time for spin up (Figure 1). The tropospheric burden of OA is relatively low during the beginning of 2004 (the first year of simulation) but it increases rapidly and becomes comparable to the rest of the simulated years even before the beginning of 2005.

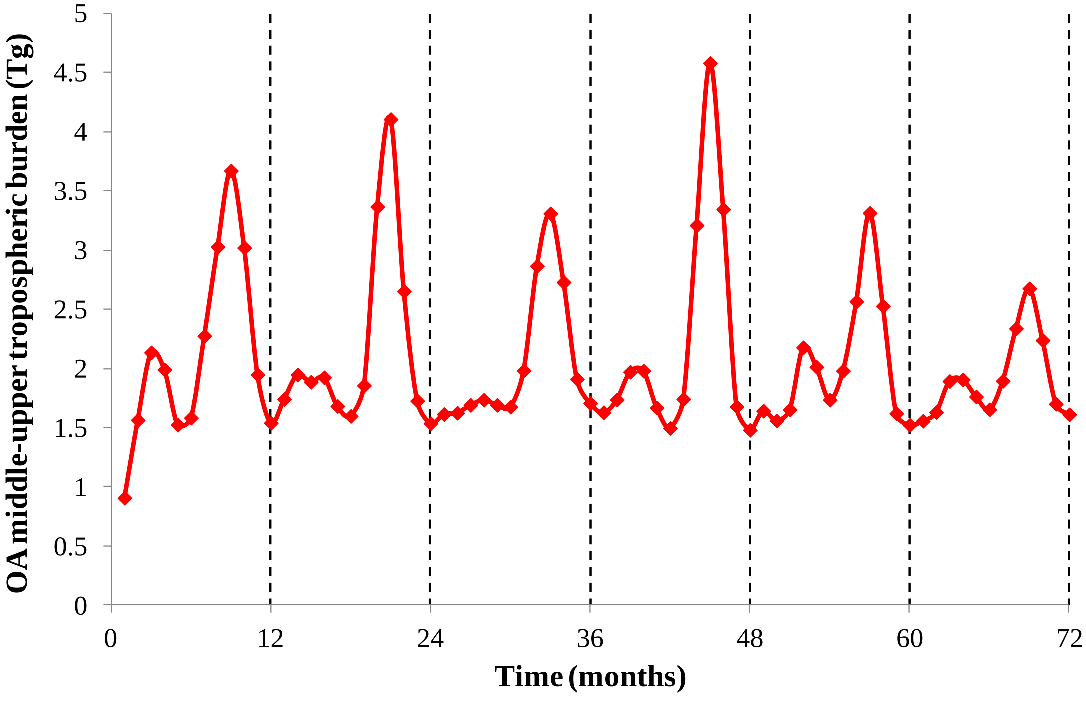

Figure 1: Predicted middle-upper tropospheric (above 840 mb) OA burden in Tg during the years of 2004-2009.

5. *p. 5471: please add a sentence/reference or two regarding the aqueous formation of sulfate, and, if any, organics.*

The formation of sulfate through the aqueous phase oxidation of SO_2 is treated by the SCAV submodel (Tost et al., 2006) which calculates the aqueous phase redox reactions based on the prognostically predicted pH of clouds. As mentioned later in the manuscript (page 5481, line 21) the aqueous phase formation of SOA is not currently treated by ORACLE. This discussion has been added to the revised manuscript.

6. *p. 5473, l. 15-16: Isn't 1e^{-1} too volatile for ELVOCs?*

   Yes, this is correct. ELVOCs are considered to have saturation concentrations of the order of 10^{-4} or less. However, their accurate representation in a model is generally useful for studies related to aerosol nucleation and growth, which is out of the scope of the current work. ORACLE does not currently simulate nucleation since it is an equilibrium partitioning model focusing on particle mass and not number. From a mass perspective, which is the focus of this application, ELVOCs exist solely in the particulate phase (as SOA-elv) under all atmospheric conditions. The lowest volatility bin used in this study has saturation concentration 10^{-1}, and is used to represent all the low volatility organics (with $C^* \leq 10^{-1}$) which are mostly in the particulate phase, even in remote areas. This information has been added to the revised manuscript.

7. *Section 3.4: are there primary marine organic emissions in the model?*

   The current application does not include any POA surrogate species for marine sources. Therefore, primary marine organic emissions are not simulated. However, marine POA can have a non-negligible influence on total aerosol forcing of climate and therefore will be included in ORACLE for future applications. This information has been added in the revised manuscript.

8. *Do you have a reference for the ONLEM submodel?*

   In the revised manuscript, we have added the appropriate reference for the ONLEM submodel (Kerkweg et al., 2006).

9. *What was the impact of the inclusion of aVOCs to the model's gas-phase chemistry?*

   The gas phase chemistry was modified by including the photochemical oxidation of VOCs that are considered as SOA precursors and the photochemical aging of aSOA-v, SOA-sv, and SOA-iv. These additional reactions have resulted in a slight decrease of the oxidant levels in the troposphere. O_3 decreased by 0.3% while OH (which is the only oxidant participating in the simulated aging reactions) decreased by 6%.

10. *Why not use RCP emissions for combustion, for consistency, and use the AeroCom ones instead?*

RCP emissions were only used for the anthropogenic VOCs species that can form SOA and were missing from the gas phase chemistry submodel MECCA. All of the other emissions used in this work were already included in EMAC and have been tested and evaluated extensively in previous model applications. Their use assists in the intercomparison with previous EMAC model versions and different model set ups. For climate applications, i.e., future projections, the RCP emissions should be considered.

11. *Also, technically speaking, the RCP emissions are not IPCC, but CMIP5.*

This has been clarified in the revised manuscript.

12. *p. 5476, l. 10-15: This approach has limitations, since e.g. aVOCs are also emitted by biomass burning.*

The reviewer makes a valid point here. Indeed a fraction of the aVOC emissions included in our work is due to biomass burning sources. However, given that these emissions represent only 10% of the total, the error that is introduced by assuming that these emissions are under high NO_x conditions is rather limited. This has been clarified in the text. A more accurate representation would be to use NO_x-depended aerosol yields and this is something that will be included in future versions of ORACLE.

13. *p. 5476, l. 20: where does the 7.5% mass increase is based?*

The 7.5% mass increase is consistent with each generation of reactions adding 1 oxygen atom to a C_{15} precursor, or 2 oxygen atoms to a C_{30} precursor. This information has been added to the revised manuscript.

14. *p. 5477, l. 1: why this is not the case for all other OA?*

Functionalization tends to decrease volatility while fragmentation tends to increase it. Unfortunately, isolating the rates at which they occur in smog-chamber experiments is extremely difficult. Therefore, the relative importance of these two processes cannot yet be quantified. However, while smog-chamber results indicate a net average decrease in volatility (and increase in SOA production) for anthropogenic SOA after their multigenerational aging (Hildebrandt et al., 2009), it appears there is not an important net average change in volatility (and SOA mass) of biogenic SOA (Ng et al., 2006; Donahue et al., 2012). Murphy et al. (2012) attributed this to a balancing of fragmentation and functionalization effects during the photochemical aging of bSOA-v. More precisely, they developed a detailed functionalization-fragmentation parameterization and predicted bulk OA concentrations similar to their base-case model configuration, which employs an organic aging module consistent with our study and excludes the aging of bSOA-v. The above are now discussed in section 3.6.

15. *The aging parameterization appears inconsistent to me. bSOA do not age, aSOA age by steps of a factor of 10, and all others age by steps of a factor of 100. Are there chemical (or other) evidence for that? The enthalpy of vaporization has a similar inconsistency, why not use an enthalpy of vaporization as a function of the volatility for all species?*

As discussed above, there is evidence that the oxidation of bSOA-v does not lead to any significant change on the corresponding mass concentration due to a rough balance between functionalization and fragmentation (Murphy et al., 2012; Wang et al., in preparation). Furthermore, we have distributed aSOA-v in 4 volatility bins with C^* equal to 1, 10, 10^2, and 10^3; and SOA-sv/SOA-iv in 4 volatility bins with C^* equal to 10^{-1}, 10^1, 10^3, and 10^5 in order to minimize the computational cost by covering their volatility range (10^{-2} to 10^6) with a low number of surrogate species. To express the decrease of volatility with aging, the products of aging reactions are shifted down one volatility bin which in the case of SOA-sv/SOA-iv is a factor of 10^2 reduction in C^* while in the case of aSOA-v is a factor of 10. Lastly, the enthalpy of vaporization for SOA-sv/SOA-iv is based on data for large saturated species commonly found in primary emissions (Donahue et al., 2006) while the effective value used for SOA-v is based on data for a-pinene and is roughly 30 kJ mol^{-1} (Pathak et al., 2007). This has been clarified in the text.

16. *What molecular weights have you used for each volatility bin?*

Based on Tsimpidi et al. (2010), the molecular weight of POA, SOA-sv, and SOA-iv is 250 g mol^{-1}. The molecular weight of bSOA-v and aSOA-v is 180 g mol^{-1} and 150 g mol^{-1}, respectively. This information has been now added in section 3.6.

17. *Results: Frequently Congo is mentioned, when the maximum is much wider than the country of Congo.*

We refer to the Congo Basin Rainforest across the Democratic Republic of Congo, most of the Republic of Congo, the southeast of Cameroon, southern Central African Republic, Gabon, and Equatorial Guinea. In the revised manuscript we use the "Congo Basin Rainforest" and not simply "Congo" to avoid confusion.

18. *In addition there is a strong seasonal cycle, from Sahel to southern Africa, which is not clearly visible in the annual mean, but needs to be mentioned.*

Indeed over the subtropical Africa there are two main seasons, the dry season (October-March) and the wet season (April-September). The dry season is characterized by intense agricultural fires in the sub-Sahelian region and forest fires in the Congo Basin rainforest. During this period, the predicted total OA average concentration is 17.4 $\mu g\ m^{-3}$, mainly due to high biomass burning emissions over the area. The wet season is characterized by low biomass burning emissions, therefore, OA consists mainly of biogenic SOA and the predicted average total OA concentration is 7.8 $\mu g\ m^{-3}$. While part of this discussion already exists in the evaluation section (4.2), it has been also added to the discussion of model predictions as well (section 4.1).

19. *p. 5481, l. 21-22: In my opinion, missing processes add primarily to the model bias, not the model uncertainty, since there is a missing source or sink. The uncertainty of the missing parameterization is second order.*

Following the reviewers recommendation we have replaced the phrase "adds to uncertainty" with "adds to the model bias".

20. *Why not exclude Ispra completely from the analysis, since the model is not able to capture the unique characteristics of the station? In any case, even if Ispra stays in the analysis, how do the statistics change in case Ispra is dropped?*

In the revised manuscript the measurements from the Ispra site have been omitted from the statistical analysis.

21. *Sections 4.3-4.7: when mentioning global average surface concentrations, it would be useful to also mention the mean over land only, since most of the aerosols are there. You can also add these numbers in table 7.*

    Following the reviewer's recommendation, we have added the average surface values over land in Table 7 and in the discussion of the results in sections 4.3-4.6.

22. *Section 4.3, tPOA: The discussion for cities is laid in a way that might give the wrong impression that you can actually resolve them. Use something like "the greater Beijing area" or "the gridbox that includes Beijing" or similar when it comes to large urban centers.*

    We have adopted the reviewer's recommendation in the discussion.

23. *p. 5486, l. 1-2: This is very interesting, are there any measurements that support it?*

    Several field campaigns over Megacities have shown that hydrocarbon-like organic aerosol (HOA; a surrogate species for POA) consists of primary combustion particles and decreases with distance from the urban source areas, due to evaporation and deposition, remaining at low levels in surrounding areas. Oxygenated organic aerosol (OOA; a surrogate species for SOA) on the other hand, consists of more oxygenated and photo-chemically processed organics which remain at high concentrations in suburban and rural sites (Aiken et al., 2009; Morgan et al., 2010; Hildebrandt et al., 2010). A short discussion about the findings of field campaigns on the matter has been added to the text.

24. *p. 5489, l. 25-27: There are studies that have challenged this statement in the past, e.g. the work of Spracklen et al.*

    That is correct; in section 4.7 we had cited other studies that highlight the importance of anthropogenic SOA for the total global budget. In the same section we have now added the work of Spracklen et al. (2011) as recommended by the reviewer. Also in the conclusion section we have rephrased the sentence "Such high anthropogenic OA fractions challenge some previous results suggesting that anthropogenic sources do not strongly contribute to global OA concentrations.” as follows “Such high anthropogenic OA fractions challenge the results of the traditional CCMs and add to the most recent findings that suggest a strong contribution of anthropogenic sources to global OA concentrations.”

25. *Figure 2: The aging arrows for SOA-v only apply to aVOCs, not bVOCs, it should be clearly mentioned. The legend should say “Gas/Particle Partitioning” not “Partition”. “Rhombi”, although literally correct, is better to be “diamonds”, since this is the symbol we are interested in the figure, not the geometrical shape. You should explain what the circles are, and probably add arrows showing which species can be emitted. Lastly, the footnote with the star should also be added at the bottom of p. 5473.*

This figure corresponds to a schematic overview of the capabilities of the ORACLE module. The user can decide to include aging reactions for bSOA-v. However, in this specific application we assume that bSOA-v does not participate in aging reactions, which is mentioned in the footnote of the figure. “Gas/Particle Partition” has been replaced by “Gas/Particle Partitioning”. “Rhombi” has been replaced by “diamonds”. Circles indicate primary organic material that can be emitted either in the gas or in the aerosol phase; this has been added to the figure caption. Lastly, section 3.3 describes only the volatility basis set theory which assumes the distribution of organic compounds into groups with logarithmically spaced effective saturation concentrations. The photochemical aging reactions are described in section 3.5 and the information that bSOA-v are assumed not to participate in aging reactions for this application already exist in this section.

Technical corrections

26. *p. 5467, l. 12: “aerosol related” needs a hyphen.*

Corrected

27. *p. 5468, l. 11: “logarithmically spaced” needs a hyphen.*

Corrected

28. *p. 5469, l. 8: “two product” needs a hyphen.*

Corrected

29. *p. 5469, l. 12: "most cases" should be "some cases". In the next line, "etc." is not needed: Pye lacks aging, Farina lacks semivolatiles; is there anything else included in "etc."?*

    That is correct; the text has been modified accordingly.

30. *p. 5470, l. 24: NO2 is not an oxidant, maybe you mean NO3 radical?*

    Yes, "NO_2" has been replaced by "NO_3".

31. *p. 5471, l. 9: It appears there is something missing here: "same size range" with their hydrophilic counterparts?*

    The sentence has been rephrased into: "The 3 hydrophobic modes have the same size range with the hydrophilic modes apart from the nucleation mode."

32. *p. 5472, l. 4: Please add "as described in section 3.6" (or something like that) after "size modes".*

    We have added in parenthesis the section number in which each process simulated by ORACLE is discussed.

33. *p. 5472, l. 8: "high number": please add the exact number under the present configuration.*

    We have added the "(i.e., 48 species are used in the current configuration)" after the "total atmospheric OA"

34. *p. 5473, l. 13: Please change "groups with" with "groups, each with".*

    The proposed change has been implemented.

35. *p. 5473, l. 19: "exist exclusively" should be "exist almost exclusively".*

    Corrected

36. *p. 5474, l. 27: The range from 0.01 to 100 is elsewhere mentioned as 0.1-10, including Fig. 1. Same for the 1e3-1e6 range 2 lines later, which is mentioned 1e3-e5 elsewhere (including Fig. 1).*

    In principle, SVOCs have saturation concentrations ranging from 10^{-1} to 10^{2} μg m^{-3} and IVOCs have saturation concentrations ranging from 10^{3} to 10^{6} μg m^{-3}. In the present application we have selected two surrogate species for SVOCs with $C^{*} = 0.1$ and 10 μg m^{-3} and two for IVOCs with $C^{*} = 10^{3}$ and 10^{5} μg m^{-3} to cover the range reported above. So, in this part we refer to the theoretical range of SVOCs' and IVOCs' saturation concentrations while later on we refer to the exact volatility bins used in this study. This has been clarified in the revised manuscript. Similarly, on the left side of Fig. 2 the theoretical range of SVOCs' and IVOCs' saturation concentrations is reported, while the symbols are placed on the exact volatility bins used by the current application. We assume that the reviewer means Fig. 2 since Fig. 1 does not include any reference to saturation concentrations

37. *p. 5475, l. 25: "monoterpene" should be "monoterpenes".*

    Corrected

38. *Eq. R8/10/12/14: The indices should be i-1, or the indices in R6 should be i. However,none of these reactions are needed here, since they show the partitioning, which is described in the following section. In any case, they are rather trivial reactions which are nicely described in the text, so they can be omitted. Line 3 from the following page also has a similar issue, it should be i-1 produces i-2, unless you change reaction R6. Lastly, this sentence (in line 3, p. 5478) should end by "until they reach the lowest volatility bin".*

    We agree with the reviewer that these reactions are simple and are part of the process described in the next section but we still believe that their addition here makes clear to the reader that the photo-oxidation of aSOG, fSOG, and bbSOG can further increase the production of SOA significantly. Therefore, following the reviewers recommendation, we have changed the indices of reactions *R8/10/12/14* to *i-1*. Furthermore, we also believe that the "until they reach the lowest volatility bin" is a nice addition to the last sentence of the section and we have adopted this change as well.

39. *The first sentence of section 4.3 is repetitive and is not needed.*

    This sentence summarizes which organic material is considered as POA by ORACLE and will help the reader to understand the behavior of POA reported on the results. Therefore we prefer not to make the suggested change.

40. *p. 5485, l. 17: "more chemically processed" compared to which?*

    They are more chemically processed than their precursor. To avoid potential ambiguity we have rephrased the sentence to: "IVOCs are emitted in the gas phase where they react with OH, becoming less volatile and more chemically processed, and condense to the aerosol phase to produce secondary organic aerosols (SOA-iv)."

41. *p. 5485, l. 19-20: "4 times higher than of SVOC (Table 4)": I don't see that in the table, but it would be good to have it there, please add it.*

    This information exists in Table 4 in the emission factors used for SVOCs and IVOCs. The sum of the emission factors for SVOCs is 0.5 while the sum of the emission factors for IVOCs is 2 (4 times higher than 0.5). However, in order to point out the difference between the SVOC and IVOC emissions we have now split the last column into the individual contributions of SVOCs and IVOCs.

42. *p. 5485, l. 26: "if" should be "though".*

    Corrected

43. *p. 5488, l. 27: Where exactly is the "higher up" that has 92% of SOA?*

    It is in the free troposphere. The sentence has been rephrased to "This results in a higher fraction of SOA in total OA in the free troposphere than at the surface (92% compared to 82% at the surface)." to avoid confusion.

44. *Table 1: POG comes from direct emissions, or only from evaporation of emitted POA?*

POG can come either from direct emissions or from the evaporation of POA. SVOCs are assumed to be emitted as POA while IVOCs are emitted as POG. Then, they are allowed to partition between the gas and particle phase which can result in some POG from the evaporation of POA on the SVOC volatility bins (10^{-1} and 10^{1}). This information has been now added to section 3.4.

45. *Table 4: Is the factor 2.5 already applied in the numbers in the last two columns, or it is applied on these numbers? Also, can you split the last column into the individual contributions of the S/IVOC?*

Yes, these factors are already applied in the last column. That is why they are 2.5 times higher than the non-volatile POA emissions reported in the table. Also, following the reviewer's recommendation, we have split the last column into the individual contributions of the SVOC and IVOC emissions.

46. *Table 6: The RMSE is not discussed at all in the manuscript. Either mention it, or drop it, don't simply include the numbers in the table.*

We would like to thank the reviewer for noticing this. RMSE is an important error metric since it incorporates both the variance of the prediction and its bias. Therefore we have added it to the discussion in section 4.2.

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