Responses to the Comments of Referee #1

1. The paper titled "Simulation of the effects of low volatility organic compounds" describes additions to the secondary organic aerosol treatment in the advanced chemistry aerosol model PMCAMx-UF. The paper includes a quite extensive comparison with measurements during a period of measurement campaigns including high frequency output from a Zeppelin. Creating a good dataset for comparison is hard and takes time. The paper also includes a general overview of the model in particular on the aerosol physics. The summary is helpful but lacks clear information on which part of the model system is new compared to older papers. Based on the summary I presume that the new parts are the ELVOC and IVOC as described by the experiments although this should be made more clear in the description or perhaps even in the introduction. This being unclear is the main reason why I placed the paper in the major revision category.
We do appreciate the positive assessment of our manuscript. We have made several changes in the revised text (including the abstract, the introduction and the model description) to improve it and make the description clearer. These changes are described below (in regular font) following each comment of the reviewer (in italics).

2. The sensitivity tests that show the impact of the additional parameterisations show is an important part, but the set-up of this experiments is unclear and should be presented earlier in the paper. Even including the small supplementary table in the main paper will help the understanding. The case definition may also be used more throughout the paper in particular in tables and figures.
We have followed the suggestion of the reviewer and moved Table S1 from the supplementary material to the main manuscript. We also discuss the various simulations earlier in the paper and repeat the definitions where necessary in the paper including tables and figures.

3. Personally I also find the structure in which a sensitivity excluding a parameter change but describing the impact as a change towards the baseline simulation is non-intuitive but again as long as the cases are clearly described this should be understandable. The equation describing this (1) is only defined for ELVOC. I think you should add the same for IVOCs.
We understand the point of the reviewer. We have chosen to start with a base case that contained both ELVOCs and IVOCs, so that a direct comparison of the model predictions to the field measurements could be made early in the manuscript. We clearly describe each case in the revised paper to avoid confusion. We have added the corresponding equation for IVOCs.

4. With respect to the comparison with measurements. Probably beyond the scope of the article but any possibility to discuss whether a change is significant or not?
This is clearly a major challenge given the issues affecting these comparisons. These issues are not limited to the experimental uncertainty, but also include problems related to the grid resolution of the model and the temporal/spatial resolution of the Zeppelin measurements.

Specific comments

5. Line 20. Presumably size = diameter everywhere?
We have replaced size with diameter at this point and later in the text where appropriate.

6. Line 25. Was this a hypothesis or the output of the model?
This is a statement that is based on the results of the model. This is now clarified in the revised paper.

7. Line 27: decreases --> decreased.
The typo has been corrected.

8. Line 43 and 48: An increase in CCN does not imply an increase in CDNC. Depending on the size of the original particles and available water vapor one may even see the inverse result. Can --> May ?
We have rephrased these two sentences to account for the potential nonlinearities in the response of CDNC to CCN changes.

9. Line 64-79 As far as I understand the yield factors from VOC precursors have been relatively constant with time. The addition of ELVOC does not change the total amount of SOA but give a different distribution of solubility; i.e. is the ELVOC a modification of the traditional treatment or and additional source?
This is a good point. The addition of the ELVOCs in our model actually increases the SOA yields, so it assumed to be, for all practical purposes, an additional source of SOA especially at low OA levels. At the same time, the addition of this extra material results in a change in the volatility distribution. This is now clearly explained in the revised manuscript.

10. Line 90. How do you decide best treatment for comparison with 2 different estimates? Sum of relative differences?
This statement refers to the recent work of Sengupta et al. (2021). The authors used different values of the ELVOC yield and compared their predictions with observations of OA mass concentration as well as N_3 and N_{50} number concentrations. A number of metrics were used for model evaluation, but the analysis was based on the Taylor model skill score. This information has been added to the paper. Additional details can of course be found in the cited publication.

11. Line 93-95. "Use" or "Extend" --> as discussed in the beginning of this review.
The word “extend” is more appropriate at this point as the simulation of ELVOCs is added to the previous version of PMCAMx-UF.

12. Line 100. The general model discussions on the previous page use N3 and N50. Can this be connected to the N10 and N100 discussed here?

Different studies have used different cutoffs. The N_3 and N_{50} concentrations are used in Gordon et al. (2016), whereas Fountoukis et al. (2012) have used N_{10} and N_{100}. In general, N_3 and N_{10} are connected to some extent and the same applies to the N_{50} and N_{100} pair. We prefer to use the N_{10} because more reliable measurements are available in more sites and the N_{100} is often closer to the CCN sizes at moderate cloud supersaturations and also for continuity given that they have been used in previous PMCAMx-UF evaluations. The use of N_3 and N_{50} is of course another good alternative. A brief explanation of this point has been added to the paper.

13. Line 113-115: This is the same as base case?

Yes, this is the base case. We have added this information at this point.

14. Line 150. I presume negligible effects of gravitational settling refers to the impact on coagulation not the overall deposition?

This refers only to the coagulation impact of the gravitational settling. We have rephrased this sentence to make this point clear.

15. Line 218. The boundary conditions are identical to Patoulias et al (2018)

We now explain in the main text that the boundary conditions are identical to those used by Patoulias et al. (2018).

16. Line 225. References e.g. version of Global Forecast System

We have added the corresponding reference and version of GFS.

17. Line 265. Organic carbon refers to the measurements? Model calculate organic mass.

The organic carbon refers to the measurements. These have been converted to organic mass for comparison with the model predictions. We have rephrased this sentence to clarify this point.

18. Line 303: What are the limit for no bias? (+- 0.5 % is quite strict if that is the limit)

The absolute bias in this station was less than 0.1%. We have changed "no bias" to "practically zero bias (less than 0.1%)".

19. Line 345-351: Any mass observations available for the PEGASOS flights?

We have followed the suggestion of the reviewer and added a comparison of the predictions of PMCAMx-UF for the aerosol mass concentration with the Zeppelin measurements. A figure comparing the average vertical profiles was also added in the Supplementary Information. Overall the model performance aloft was quite similar with that at the ground level. For example, for the 9 Zeppelin flights (approximately 1300 data points) the OA normalized mean bias was -4% and the normalized mean error equal to 40%.

20. Line 404-408. Although it is only one site S4 show a quite pronounced difference between the cases. May be interesting to include in the paper. Is the deviation between measurements and model for the smallest particles a question of detection limit or actual difference?
The difference in the predictions of the two simulations (with and without ELVOCs) in this site are modest. The discrepancy between model predictions and measurements is due to both the weakness of the measurements (particles smaller than 3 nm were not measured) and a tendency of the model to overpredict nucleation event intensity in this area. This information has been added to the paper.

21. Line 453. From 35 to 35 % I think can be called constant even if the is a "numerical change"
We corrected the typo. The change is from 38% (not 35%) to 35%.

22. Table 1 and 2. Given that the individual stations are not discussed in the text, I think that the tables can be moved into supplementary material and replaced by totals or regional values.
We would prefer to keep the two tables with the model performance in each station in the main text given that we refer to them and the corresponding performance several times during the discussion.

23. Table 4,5 7,8 : Only need fractional or absolute change. Move the other into Supplementary
Please note that Table 4 includes the evaluation of PM_1 OA predictions against available AMS measurements during the PEGASOS campaign while Table 5 shows the corresponding evaluation results against $PM_{2.5}$ OC measurements converted to OA mass in other sites in Europe. In both only the normalized mean error and normalized mean bias are shown. There are only 5 tables in the main manuscript.

Responses to the Comments of Referee #2

1. The manuscript "Simulation of the effects of low volatility organic compounds of aerosol number concentrations in Europe" by Patoulias and Pandis presents a model investigation on how including extremely low volatile organic compounds and intermediate volatility organic compounds affect simulated aerosol number (and mass) concentrations. The manuscript is very well written and the topic of the paper addresses relevant scientific questions within the scope of Atmospheric Chemistry and Physics. There are few models around that can simulate the formation and growth of aerosol by gas-to-particle partitioning of semivolatile organic compounds as detailed as PMCAMx-UF. Although the results indicate that these compounds have a minor effect on aerosol number concentrations over the studied region, it is an interesting result. I recommend publishing this manuscript once the following minor points have been addressed.

We thank the reviewer for the positive assessment of our study. We do agree that the main result of our paper is rather surprising. Our responses to the comments of the reviewer and the corresponding changes to the paper (in regular font) follow each comment of the reviewer (in italics).

2. In the model description, it is laborious to piece together the methods that the model uses for aerosol physics since the description relies on referenced articles. For example, solving condensation of inorganic and organic compounds simultaneously remains unclear to me. It seems that organics are always assumed to be in a separate phase from water and inorganics. Is this correct? In addition, it seems that water uptake uses a parameterization for bisulfate. Is the amount of sulfate equal to the amount of bisulfate in particles? Are organic compounds assumed to be hydrophobic?

We have added information to the model description section addressing the main points raised by the reviewer. Indeed, the model assumes that organics and inorganics are in different phases, but in the same particles. Therefore, the condensation of one affects the size distribution of the particles and therefore the condensation rate of the other. The inorganic aerosol thermodynamics including the sulfate/bisulfate split and the water uptake by all inorganic aerosol components are simulated with a detailed aerosol thermodynamics model, ISORROPIA. The water content of the organic aerosol is neglected in this version of PMCAMx-UF and the aerosol water is dominated by the inorganic aerosol components. Additional information can be found in previous publications describing the evolution of PMCAMx-UF (Jung et al. 2010; Fountoukis et al. 2012; Patoulias et al. 2018).

3. It would also be helpful for the reader to summarize the ELVOC yields and IVOC emissions in a table.

We have followed the suggestion of the reviewer and moved Table S1 from the supplementary material to the main text. This was also recommended by reviewer 1 (comment 2). We have made changes in the main text, clarifying the emissions/yields used in each simulation.

4. Line 174: Murphy at al. => Murphy et al.
We have corrected the typo.

5. Line 183: Are IVOCs additional to POA?
IVOCs were not included in the original emission inventory and therefore have been added to the emissions. Their emission rate is scaled based on the non-volatile POA emissions included in the inventory. Their total emission rate is assumed to be 1.5 times the non-volatile POA emissions. This is now explained in the revised paper.

6. Is modelled OA in $PM_{2.5}$ and filter measured OA in $PM_{2.5}$ fully comparable as part of semivolatile compounds in filter samples can be evaporated, while modelled OA will include all semivolatile material?
The measurement of OA using filters is characterized by two artifacts: a positive one involving adsorption of organic vapors on the quartz filters used for the sampling and a negative one related to the evaporation of some of the semi-volatile material. There is a rich literature on the magnitude of these artifacts and on ways to minimize them or correct for them (involving denuders for removal of organic vapors and after-filters). In this work, we use the reported measurements for the model evaluation keeping in mind their uncertainty. A brief discussion of this point has been added to the paper.

7. In Conclusions Lines 485-489 it is said that the growth of the newly formed particles is suppressed because changes in size distribution decrease nucleation rates, sulfuric acid concentrations, and increase the coagulation sink. However, these changes are not backed up with numbers. This conclusion is probably true, but needs to be diagnosed from the model.
We have followed the suggestion of the reviewer and prepared a new figure (included in the supplementary information) showing the fractional change in the number concentration of $N_{1\text{-}10}$ (reflecting nucleation rates), sulfuric acid, condensational sink and coagulation sink due to the ELVOCs. This figure supports quantitatively our argument of decreasing nucleation rates, decreasing sulfuric acid levels and increasing coagulation/condensational sinks when ELVOCs are added to the model. These changes are especially pronounced in the Scandinavian Peninsula.