We would like to thank the Reviewers and the editor for their diligent and careful review of our work. We will be addressing the points made by the referees below. The bold quoted indented sections show the comment made by the Reviewer, if modifications are made regarding the comment they are presented in quotes and in light blue. Line numbers are given for the new additions in the text, the line numbers refer to the modified pdf file.

Reviewer #1

> **"This is overall a well-designed and comprehensive modelling study about the secondary organic aerosol formation over the Landes pine forest in south-western France. I think the manuscript can be well worth to be published in atmospheric chemistry and physics after some relatively minor but essential revision."**

We thank the Reviewer for their encouraging words. We will do our best to provide the minor changes proposed by the Reviewer below.

General comments:

> **1. "Since this is a model study about biogenic secondary organic aerosol (SOA) formation I think it is essential to describe the terpene gas-phase chemistry and SOA formation scheme used in the CHIMERE model in some detail. You state that you use the SAPRC-07A chemical scheme and provide several references to the used SOA scheme but I also think that it is appropriate to at least briefly describe the SOA formation scheme in the presented manuscript. Since the modelled BSOA formation is overestimated while the modelled terpene concentrations seem to be underestimated, at least in the surface layer at the measurement station in the Landes forest they most straight forward explanation that come to my mind would be too high BSOA yields in the model."**

This is a suggestion made by both Reviewers. We have added the following paragraph to explain more about the SOA formation scheme (lines 195—206):

> "The scheme is based on Odum et al. (1997), a simple two-product scheme with the advantage of being numerically light. In this scheme it is considered that the oxidation of BVOCs results in the formation of semi-volatile products with a yield specific to each BVOC family. Some BVOCs consist of specific species (isoprene, α-pinene, β-pinene), others are surrogate groups consisting of similar species (ocimene, limonene, sesqui-terpenes, etc.). As mentioned above, it considers only one step of oxidation for the BVOCs. Assuming homogeneous mixture, Raoult's law is applied combined with the Pankow theorem (Pankow 1987) resulting in the calculation of the SOA production yield. The scheme is explained in more detail in Pun et al. (2007). The oxidation reactions that are applied for the production of semi-volatile VOCs have been updated in order to take into account more recent (and more detailed) reactions and reaction rates. The values given on the SAPRC site (following Carter, 2010) were used in order to update the reaction rates of the BVOCs mentioned above with OH, O_3 and NO_3. The SOA yields have been kept the same as what is provided in Pun et al (2007) and Bessagnet et al (2008) as well as provided in the CHIMERE documentation. We have also added the SOA-related reactions of BVOCs with different oxidants in the SI (refer to SI-8)."

> **2. "In figure 7 you present the modelled and observed monoterpene concentrations over the Landes forest and in Fig. 4 you show maps with total monoterpenes and**

isoprene emissions. However, I miss information about the actual "base case" and "final" BVOC species emission factors (e.g. the individual monoterpene species emission factors implemented into CHIMERE) for the maritime pine forest and how the BVOC emissions were handled over the non-forested patches (e.g. meadows an various fields with crops). Could you provide some statistics on this?"

In order to respond to this question, we have to explain the procedure of simulation of biogenic emissions in CHIMERE in more detail. The CHIMERE model does not depend on individual tree types, it takes in pre-calculated emission factors for the BVOC species it represents (which have been listed in the article as well as in the response to point 1 of this Reviewer's remark. These pre-calculated emission factors are provided in global netcdf files with a 300mx300m horizontal resolution giving the total emission factor (of all tree types/plants combined) of each BVOC lumped species. These files are created by the MEGAN model.

The way the MEGANv3 model calculates these factors is it uses a global file (which will be called the ecosystem file from here on), in which global land surfaces are divided into smaller areas that have similar vegetation types. This should not be confused with the landcover. The file indicates the vegetation type regardless of what percentage of the land they cover, the latter fact being decided by the landcover files (taking into account LAI). Let's consider a non-factual example of a city near the Landes forest. The landcover files indicate that this example city has a low tree cover of around 10% (the tree cover density is determined by the LAI in the landcover files). The ecosystem file will, in this case, indicate that this 10% is going to be 90% pine trees. The basic emission factor of these pine trees is given as inputs to the MEGAN model along with the ecosystem file, the landcover and leaf area index files; the MEGAN model calculates total emission factors for each requested BVOC species. This is then fed to the CHIMERE model as input. Therefore, having given updated landcover data to both MEGANv3 and CHIMERE models, it becomes possible to produce the placement of forested areas as well as crop fields and meadows more accurately.

Regarding statistics, initially the ecosystem file used in MEGANv3 told us that the Landes region in France contains 28% pinus pinaster (maritime pines), 24% Quercus robur, 11% Picea abies and 30 other species all having a density of under 5%. This distribution is not correct. Other databases (BDforet) show a 95% of maritime pine for the Landes forest. This value is used in the final case. The emission factor of this tree species was also modified since it was measured on the canopy and the litter levels during the same campaign this study simulates. The measurements resulted in an average of 3.8 $\mu g.g^{-1}.h^{-1}$ at the canopy level and an average of 1.6 $mg.g^{-1}.h^{-1}$ at the litter level of total monoterpene emissions. The values provided here for the canopy and the litter are respectively almost 3 times and 2 times higher than the previously measured value. These values apply to the "forest" portion of the landcover, which is around 65% of the Landes area. Upon the recommendation of the reviewer, we have added the averaged emission factors in the manuscript (lines 336 – 340).

3. "Several of the figures have very small legends and units (e.g. Fig. 4, 13) or are smaller than they need to be (e.g. Fig. 5 and 11). Please consider if the font size could be increased. I also think that all subpanels should be named a, b, c, d … and all panel should be described briefly in the figure text. E.g. Panel a shows …"

We have added panel names to all figures that needed them and added references to these panels in the text. We have made figures 5, 11 and 13 bigger, we have also changed the labels and units in figures 4. In relation to another comment made by the Reviewer (below) we have also added a second scale to figure 3 regarding panel a.

4. “At several places in the manuscript you include links to datasets directly in the text but no references which describes the datasets. I think the webpage links should be complemented with references to publications where these datasets are described, if existing. E.g. I am sure that you can find some publication(report) that describes the EMEP anthropogenic emission inventories (L222) and Copernicus database on tree cover density (L276).”

We agree with the Reviewer that this is the proper way of handling URLs in an article. We have added references for all the URLs that were in the article instead.

5. “In table 2 you provide some statistics about how well the different model tests results agree with the observations in the Landes forest. I don’t think you specify what you mean with R. Is it the correlation coefficient?, What do you mean with “bias”, is it just the absolute difference between the arithmetic mean concentrations from the model and observations? I think you need to perform a bit more rigorous statistical analysis of how the different model versions performs. At least I think it is also important to report the Normalized Mean Bias (NMB) and the fraction of predictions within a factor of two of observations (FAC2). Then I also think you should refer back to table 2 in the text when you describe the results from the different model results in Sect. 4 and 5.”

Yes, R is the correlation coefficient. Bias is the absolute difference between the means as the Reviewer correctly points out. We can of course add the two factors the Reviewer has requested. We have modified table 2 to include this information, we have also modified the caption to the table.

6. “It is no problem for me to understand the general method or results of the present study, but I think the present work would benefit from a through English grammar check prior to the final publication. I have specified a few sentences which I think need to be revised below.”

This suggestion has been made by both Reviewers. We have made a thorough rereading of the article in order to improve English grammar.

Specific comments

1. “L29, “Henrys constant”. I think it should be Henry’s law constants.”

The Reviewer is correct, this has been corrected.

2. “L80 “improve (or not) agreement”. Consider to rephrase. E.g. influence the agreement”

This has been corrected.

3. “L103-105 “Regarding biogenic emissions, the site is quite homogeneous since the majority of the trees in the surrounding area are maritime pines. However, as

mentioned above, since the forest is parcellated the density of the forest and the geographical distribution of these emissions have a certain degree of heterogeneity." Looking at satellite images over the area the landscape seem to be very patchy and not at all an homogeneous forest. Do you mean that almost all patches have planted maritime pines but of variable age? Please explain what you mean with that the site is "quite homogeneous" but at the same time "parcellated"."

The forest contains privately owned parcels which can be agricultural fields or pine trees of different ages, this is what was meant as "parcellated". While the type of trees that are present in the forest (in both publicly or privately owned parts of it) are in vast majority of cases maritime pine trees (therefore homogeneous in regards to tree types and therefore emission factors resulting from these trees) the parcels can become agricultural fields or be replanted with pine trees from one year to the next resulting in the patchy appearance that the Reviewer has correctly mentioned in their comment. We have modified the text to explain better (lines 102—105):

> "Regarding biogenic emissions, the site is quite homogeneous regarding tree types since the large majority of the trees in the surrounding area are maritime pines. However, as mentioned above, since the forest is parcellated, the density of the forest and the geographical distribution of these emissions have a certain degree of heterogeneity; parcels can become agricultural fields or replanted with pine trees, resulting in pine trees with different ages."

4. "L125-L126 "Since the purpose of the article is to focus on pollutant build-up from biogenic compounds" Consider if you should replace "pollutant build-up" with secondary organic aerosol formation. Too me it sounds a bit strange to call BSOA formation "pollutant build-up" even if I agree that it in potentially could be a health concern."

We have made the following modification (lines 126 – 127):

> "Since the purpose of the article is to focus on organic aerosol formation from biogenic compounds and the effect of anthropogenic VOCs in the atmospheric chemistry of region is limited, anthropogenic VOC measurements are not explored."

5. "L160-163 "The parameters common to all sensitivity tests are explained in section 3.1. After the description of the base simulation (section 3.2), sections 3.3 through 3.6 describe sensitivity tests changing inputs/variable calculations. Keep in mind that each modification is added on top of the previous ones. The implemented improvements are indicated in Table 1."

I think the manuscript is well structured, but can you always state that the implemented changes are "improvements"? From the statistics presented in table 2, this is not always clear. I would replace "improvements" with changes or possibly "increasing complexity/resolution" "

This is a good remark, it is indeed not always an improvement, nor it was meant to as such all the time. The modifications were made as sensitivity tests which might not always result in actual improvements. We have modified the word "improvements" to "changes" (line 165):

> "The implemented changes are indicated in Table 1."

6. “L279-281 “These changes are, on the average, about +30% forest growth in cells where the changes were positive and about -40% in cells where the changes were negative. Given these important changes, only the most recent (2015) data was considered.”

I don’t understand what you mean with these sentences. What do you mean with “cells”?”

We are using the word “cell” to designate the model grid cells that constitute the Eulerian model grid. The domain discussed here contains 220x200 grid cells with a resolution of 1x1km. The values given there are the average for all the grid cells in the domain where the change in the forest density is positive for the first value or negative for the second one. We will replace “cell” by “grid cell”.

7. “Figure 3. I miss a legend about the absolute tree density “shades of green” in the upper left figure.”

We have added this legend, we have also added panel lettering in regards to the general comment made by the Reviewer.

8. “L306 “only slightly lower densities”. Replace with some numbers about how much lower the densities actually are.”

The phrase has been modified to the following (lines 316 – 319):

> “It can be seen that there are practically no differences between Theia-2018, BDTopo and Theia-2016 (average density over the forest is respectively 68.32%, 68.95% and 68.20%), and only slightly lower densities in the BDForêt and MEGAN databases (63.2% and 58.29% respectively), while strong differences appear with the MODIS, Globcover, Copernicus and USGS data.”

9. “L317-318 “It was considered necessary to modify the tree type distribution in the MEGAN model since MEGAN 2.1 assumes 28% of maritime pine coverage for the Landes forest, while the BDForet database shows around 95% of the same species.”

Is it actually the MEGAN 2.1 model which assumes 28% of maritime pine coverage that is the problem in this case or just how the MEGAN 2.1 model has been setup? I guess the 28% of maritime pine coverage is not hard coded into the MEGAN 2.1 source code but provided as some input variables to the model or? In this case it should be enough to change how MEGAN v2.1 is setup and not the version of MEGAN, v2.1 or v3 or?”

Up to MEGAN v3 global emission factors were presented as netcdf input files for BVOC species (having been pre-calculated by the development team of MEGAN). Starting from MEGAN v3, a pre-processor was added to the model which gives the user the possibility of recalculating the emission factors using specific tree types for different regions. This pre-processor has input files with ecosystem types (each containing specific tree/plant types); the ecosystem having been defined in a global netcdf file. We used this pre-processor to recalculate the emission factors for BVOCs by correcting the tree types used for the Landes region in the model’s input. Since this pre-processor comes with MEGAN v3 we have mentioned that we have used MEGAN v3.

10. "L326-328 "Figure 4 justifies the necessity of this modification, since as mentioned above the emissions of isoprene have dropped by about half, while the emissions of terpenoids have increased significantly, by about a factor of 2"

I don't think you can state that this by just looking at the results in Figure 4. Possibly you can draw this conclusion when comparing the modelled monoterpene and isoprene concentrations with the observations from the station."

In this sensitivity case the input data especially of the tree species distribution have been refined for regional conditions and thus been made more realistic. In **L326-328**, we state that the effect of the modification is large, so this justifies a posteriori having made such a sensitivity test. In addition, as mentioned before, the sensitivity tests were not meant to be seen as definite improvements, but as changes that might result in changes in the model results. However, we do agree with the Reviewer that the sentence is ambiguous and also that it should have been mentioned that model/observation comparisons have been performed for both isoprene and terpenoids, which is done in a later section of the paper. We have modified the sentence to the following (lines 344 – 346):

"Figure 4 justifies a posteriori the importance of this sensitivity test, since as mentioned above the emissions of isoprene have dropped by about half, while the emissions of terpenoids have increased significantly, by about a factor of 2; in Section 4.1, we will explore the effect of these emission changes on BVOC concentrations and compare the results to observations."

11. "L360-362 "While the effect of the canopy on BVOC emissions is already taken into account in the MEGAN scheme, the canopy effects on wind speed and vertical diffusion inside the forest are not simulated in the CHIMERE model. This normally causes no issues, since most measurements are performed above canopy level"

Just because you normally do not compare the modelled concentrations inside the forest canopy with observations from inside the canopy this can still be a general model issue. E.g. when it comes to the modelled deposition of BVOC oxidation products in the forest canopy. Please consider reformulating this statement."

We agree with the reviewer. We have tried to explain this can be a genuine issue on high resolution simulations and it is probably necessary to couple the model to a forest canopy model or at the least put into place a subgrid scheme that takes into account sub variations of a forest canopy when looking at high resolution simulations. This probably could have allowed a more coherent treatment of all canopy induced processes, including deposition of oxidized BVOC products but which is included, but only for the first layer (at ground level). We have modified the sentence to the following to make this more apparent (lines 377 – 385):

"While the effect of the canopy on BVOC emissions is already taken into account in the MEGAN model, the effects of the canopy on wind speed and vertical diffusion inside the forest are not simulated in the CHIMERE model. On a large-scale simulation and not comparing to inside-canopy measurements this usually does not cause major issues. But since the goal of this work was to understand the atmospheric situation inside the canopy, almost all measurements are performed inside it. Furthermore, simulations performed here are high resolution simulations, therefore, we have added corrections for the aforementioned physical parameters inside the canopy in order to simulate the physical conditions of the forest more realistically. These modifications will be presented as the 6th sensitivity test,

theoretically thought to be the most realistic one. In this test, a small inconsistency subsists because deposition is only treated in the lowest layer in the model, while it also could affect the second layer which contains part of the canopy."

12. "L399. In eq. 7 the altitude is denoted by λ but in eq. 1,2 and 6 you use z to denote altitude."

The Reviewer is correct. The two equations coming from different references their nomenclature is a bit different. We have modified the λ in equation 7 to z.

13. "L401-402. "k is an extinction factor, which was measured to be 0.33 for the Landes forest (calculated by Ogee et al. (2003) using experimental data)."

Considering how patchy the Landes forest is, can you assume that the same empirical k parameter is valid over the whole Landes forest?"

No. But the equation is used only for cells where the forest density is higher than 70% (as mentioned a few lines later) and it is multiplied by the LAI to take into account the heterogeneous density. It should also be mentioned that the extinction factor was calculated to a patch with the same canopy height as the year 2017 in the reference paper. Therefore, we think it can be applied to similar conditions as the reference that calculated the extinction factor.

14. "L437-440 "The sum of monoterpenes is the first group of species that we consider here from PTR-Tof-MS measurements (m/z 137 peak as their main fragment and m/z 81 values as their second most important one) and sum of simulated α-pinene, β-pinene, limonene, ocimene concentrations, see section 3.4) since these are main drivers of atmospheric chemistry for the given terpene emitting maritime pine forest."

This paragraph needs to be clarified. I am not an expert on PTR-Tof-MS measurements so I cannot follow how the measured total monoterpene concentrations actually were derived from the mass spectrum. C10H16 should be closer to mass 136, but I guess you add a proton H+ for the detection (thus m/z 137) or? But then what about m/z 81? Why is this mass peak also counted as monoterpenes?"

This is a remark made by both reviewers. The reviewer is correct on the m/z 137. When looking at mass to charge ratios (m/z) for monoterpenes two main values are taken into account: 137 for α/β-pinene and 81, a terpenoid fragment. As the reviewer correctly points out 137 is the mass for monoterpenes (taking into account the proton), while 81 is the mass for one of the major fragments seen in mass spectrum of monoterpenes. This has been seen in multiple studies; Li et al (2020) explores the PTR-Tof-MS measurements for the Landes forest and explains that previous studies have shown that oxygenated terpenoids can fragment inside the PTR instrument and produce a dominant ion at m/z 81, Li et al (2021) working on the Landes forest and the boreal forest in southern Finland makes a similar observation. We have added these two references that explain the fragmentation of terpenoids inside a chamber.

15 "L441 "BVOCs has", BVOCs have …"

It has been modified, thank you.

16. “L469. Consider replacing “heavily” with substantially.”

Thank you, it has been modified.

17. “L473-476 “It is also important to keep in mind that in the canopy test case, the changes seen in the concentrations arise almost entirely from the Kz modifications and not from the swrd ones (as they do not affect the emission of BVOCs since their effect is already taken into account in the emission factor calculations).“

Consider to reformulate this sentence. What do you mean with “swrd ones” ?”

It was meant to point towards the modifications made to the model variable swrd which stands for short wave radiation at ground, as explained in section 3.6. We have modified the phrase and removed a part of it (since it did not convey was we wanted to convey) (lines 500 – 502).

> “It is also important to keep in mind that in the canopy test case, the changes seen in the concentrations come almost entirely from the K_z modifications and not from the modification made to the shortwave radiation.”

18. “L486. What do you mean with “mean bias” and “0.43 for correlation”. Is it the normalized mean bias (NMB) and correlation coefficient?”

With mean bias, we indicate the average difference between simulations and observations, in absolute values. We have modified this in the text and call this variable “bias (simulation – observation)”, which should be clear enough’. Correlation indeed is the correlation coefficient; we have modified every mention of correlation in the text to correlation coefficient.

19. “L500-502 “While simulations with the standard model version correctly reproduce the day-time O3 maxima, the significantly overestimate the O3 minima, which never decrease below to 20 ppb.” Correct this sentence”

The phrase has been corrected to the following (lines 528 – 530):

> “While simulations with the standard model version correctly reproduce the day-time O_3 maxima, they significantly overestimate the O_3 minima; simulations never show below 20 ppb, while measurement minima can reach 0 during nighttime.”

20. “L503-505 “Daily O3 maxima are only slightly changed in sensitivity tests, the largest impact being noted for the test with refined emissions, leading to enhanced NOx emissions and an increase in the O3 peak on July 7 from about 60 to about 70 ppb (Figure 6).”

Too me it looks like the major change in the maximum O3 concentrations occur already between simulation 1 (base case) and simulation 2 (changed meteorology).”

The overall changes for O_3 concentrations occur already in the meteorology test case, however, the sentence highlighted by the Reviewer is discussing O_3 maxima, which, as seen in table 2, change the most in the sensitivity test concerning anthropogenic emissions. The confusion we admit comes from the fact that the text does not mention that the changes are caused by both

meteorological input changes and emission changes, to address this we have modified the text to the following (lines 531 – 536):

> "Daily O_3 maxima are only slightly changed in sensitivity tests, the largest impact being noted for the test with refined emissions, leading to enhanced NO_x emissions and an increase in the O_3 peak on July 7 from about 60 to about 70 ppb (Figure 6 and Table 2). It should be mentioned that while the maximum change in maxima of O_3 is seen in the emissions test case, the maximum for the aforementioned day already changes when the meteorological inputs are modified in test case 2."

21. "L521 "seem well-simulated" replace with some statistical measure of the model performance."

We have added where the statistics and figures can be accessed in the parenthesis that comes after this phrase.

22. "L524 "dry deposition speed". Maybe this is OK but I think you normally call it dry deposition velocity and not dry deposition speed."

It has been modified.

23. "L524 "Another plausible candidate for it might be an underestimation of deposition of O3 over forested areas." Maybe reformulate this sentence."

It has been modified to the following (lines 554 – 555):

> "Another plausible candidate for this issue could be an underestimation of deposition of O_3 over forested zones."

24. "L540 "maxima (up to 0.1 ppt) in contrast due to our simulated night-time maxima." I don't understand this sentence."

The Reviewer is correct in pointing out that this phrase makes no sense. The phrase mentioned by the Reviewer as well as the sentence after have been modified to the following (lines 569 – 574):

> "The night-time overestimation of O_3 minima probably leads to an overestimation in NO_3 concentrations. Interestingly, Mermet et al. (2021) calculated the NO_3 concentrations through steady state equations inside the canopy using measured NO, NO_2, O_3, monoterpene and isoprene concentrations as well as radiation parameters; they found day-time maxima of up to 0.1 ppt in contrast to our simulated night-time maxima (the reason for calculating a daytime maximum in the aforementioned study for a species that should not have significant day-time concentrations is unclear)."

25. "L548 "diminution of OH" Do you mean decreasing OH concentration?"

Yes, it has been modified to "a decrease in OH concentration".

26. "L553 and figure 8. I suggest that you also report the modelled OH concentration in unit molecules/cm^3 and not in ppb."

Yes, the figure has been modified to show the concentrations in molecules.cm^{-3}.

27. "L568-571 "It is concluded from the model results and known OA sources, that most of it is biogenic SOA (84% at Salles-Bilos over the campaign period). For instance, the concentration of anthropogenic OA (primary and secondary is quite low at this site (average of 0.11 µg.m−3 overall test cases)."

How can you know the OA sources and that 84 % is biogenic SOA? Are these numbers coming from model results or observations? Please explain in more detail and provide some reference. I would suggest that you actually perform a model sensitivity run without BVOC emissions over the Landes forest in order to quantify the modelled BSOA originating from the local BVOC emissions."

The given value comes from the simulations, as it is stated in the text. The model is able to separate OA on the basis of its origins and its age; BSOA and ASOA are presented as separate species with separate chemical reactions and chemical outputs, therefore it is possible to calculate the contribution of BSOA to total OA quite easily using the model outputs.

28. "L576-580 "Changing the Kz parameterization and in canopy radiation decrease both OA and BSOA, as the concentrations of the three oxidants (OH, NO3 and O3) participate in the biogenic OA formation process, while increasing the concentrations of terpenes (its major precursors). This test reveals an interesting feature: for BSOA formation around Salles-Bilos, changes in oxidation rates have a larger effect than changes in precursors (i.e. changes in terpenoids and isoprene)."

Do you mean changes in precursor emissions or changes in precursor concentrations in the canopy? It is obviously the case that decreasing mixing and decreasing in canopy radiation will result in higher surface (in canopy) terpene concentrations and decreasing biogenic OA formation since the integrated total boundary layer oxidation rates of the terpenes will decrease and thus also the biogenic SOA formation in the whole boundary layer. If less terpenes are oxidized in the boundary layer you get less biogenic SOA. "

We agree with the reviewer's argumentation. Our reasoning should have been based on BVOC emission and not concentration changes, however BVOC emissions are kept constant in this sensitivity case that is being discussed. We reformulate the sentence as follows (lines 610 – 612):

"Changing the K_z parametrization and in canopy radiation decreases both OA and BSOA, as well as the concentrations of the three oxidants (OH, NO_3 and O_3) which participate in the biogenic OA formation process, while increasing the concentrations of terpenes (its major precursors). So, a lesser portion of the emitted terpene is oxidized and less BSOA is therefore formed."

29. "Figure 10. Can you give some explanation to the higher O3 concentrations over the ocean compared to the ozone concentrations over the Landes forest?"

It should be due to the slower deposition of ozone over the ocean. Looking at the deposition in the model, ozone has a lower deposition rate over the ocean and therefore the deposition rate is lower over the ocean. We have added the following sentence on this regard:

"The higher concentration of O_3 observed over the ocean in Figure 10 (panels a4 to d4) is due to the lower deposition of O_3 over bodies of water, since its dry deposition rate is lower."

30. “L596 “outstanding in this group” Consider reformulating this sentence.”

We have changed outstanding to noteworthy.

31. “L608 “Differences with respect to our study can be explained by many factors which we do not attempt to quantify here:”

What other study do you compare with? What is the difference between?”

The comparison is made to the results seen in the experimental study for the same campaign cited in the previous paragraph. The reviewer makes a good point that we did not mention this, so we add this to the revised text.

32. “Section 5.2. I think the case studies focusing on the see breeze fronts are very interesting but the onset of the OA peak and the rapid drop in OA seem to be a bit shifted between the model and observations, at least with a few hours. In the observations the OA mass seem to drop before noon while in the model it occurs later in the afternoon. Can you provide some potential explanation to this? Could it also have to do with the planetary boundary layer height (PBLH)? Maybe the model underestimates the PBLH?”

We cannot exclude the PBLH rise contributed to the earlier drop in observed OA concentrations with respect to simulated ones on July 7 (while in July 5 the afternoon drop is more coincident). In the absence of any boundary layer height measurements, we are unable to control its time evolution. However, the good agreement in the simulated and observed daily temperature evolution gives some credibility to that the simulated sea breeze is real. In both cases, maximum temperature appears quite early in the afternoon, which in our interpretation is caused by the sea breeze. We added in the manuscript (lines 732 – 734):

> “Differences in the exact timing of the frontal passages are visible especially for July 7th, when observed organic aerosol concentration at the Salles-Bilos site drops earlier than the simulated one. This could be due to other factors as for example an increase in boundary layer height not accounted for in the simulations.”

33. “L721-723 “The scenario representing physical changes to the simulations of vertical diffusivity, wind speed and radiation penetration inside the canopy seems to have a more realistic view of what the measurements show about the atmospheric chemistry inside the forest, especially for terpenes, radicals and BSOA formation.” I don’t understand this sentence completely. Please consider to reformulate it. What do you mean with “realistic view”“

The sentence wants to express that this test case (the one taking into account the physical changes related to the canopy) corresponds to measurements in more aspects than just OA concentrations, i.e. the inputs and the parametrization used for it are closer to what a realistic view of the domain should be. We have modified the phrase to the following (lines 756 – 760):

> “What has been noted as the physical test case (taking into account physical changes to the simulations of vertical diffusivity, wind speed and radiation penetration inside the

canopy) presents a closer representation of the measurements observed in the Salles-Bilos site as well as of several air quality measurement stations around the forest."

34. "L724-727 "Using the final test case, we also simulated the formation of BSOA from different precursors and oxidants. This showed us that the chemical pathways behind the BSOA formation for the base case scenario is inaccurate in the base case since the majority of BSOA is formed through the oxidation of isoprene, while this is changed to terpenes (specifically sesquiterpenes and β-pinene) in the final simulations."

Is it the chemical pathways behind the BSOA or the BVOC emissions which are inaccurate in the base case simulations? Can you make any definite statement about the accuracy of the chemical pathways of the BVOCs and the SOA formation in this work?"

We agree with the reviewer that this sentence is badly formulated. The chemical pathways were updated for all the test cases starting from the base case. Since the pathways do not change between the tests it is hard to believe the chemical pathway is the reason for the changes seen in the test cases. What was meant by the phrase "chemical pathways behind the BSOA formation for the base case scenario is inaccurate" is that more BSOA is formed through isoprene than through monoterpenes in the first test case, which is demonstrably wrong for this region. The reason for this is the bad representation of BVOC emissions (high isoprene emissions and low monoterpene emissions) resulting in the issue mentioned above. We have reformulated the sentence to the following (lines 760 – 764):

"Using the final and the base test cases, we also simulated the formation of BSOA from different precursors and oxidants. This showed us that the formation of BSOA for the base case scenario comes from the wrong precursors, as BSOA is mostly formed from the oxidation of isoprene which is not accurate for this region. The formation of BSOA changes to terpenes (specifically sesquiterpenes and β-pinene) in the final simulations, which is more in line with the observations in the Landes forest."

Reviewer #2

General comments:

> **1. "The authors in this study attempted to understand the formation of BSOA, in a maritime Pine Forest located in Southwestern France. They aim to accomplish this by making relevant changes, for e.g., to the land cover model, including measured emission rates for different biogenic precursor species, and incorporating a canopy model. This is a well-thought-out comprehensive study and I would recommend publication, subject to modifications and suggestions put forward below."**

We would like to thank the Reviewer #2 for his positive appreciation of this work and we will be responding to the comments made by the Reviewer point by point below.

> **2. "Since the manuscript heavily focuses on the modeling of BSOA, in the maritime pine forest in France, the authors do not explain in detail the SOA model used in CHIMERE. The authors do recognize the fact that it would be worth testing out different SOA schemes (L746), but, in my opinion, it would benefit the reader to know more details about the SOA scheme used in this study."**

This is a suggestion made by both Reviewers. We have added the following paragraph to explain more about the SOA formation scheme (lines 195 – 206):

> "The scheme is based on Odum et al. (1997), a simple two-product scheme with the advantage of being numerically light. In this scheme it is considered that the oxidation of BVOCs results in formation of semi-volatile products with a yield specific to each BVOC family. Some BVOCs consist of specific species (isoprene, α-pinene, β-pinene), others are lumped species consisting of similar species (ocimene, limonene, sesqui-terpenes, etc.). As mentioned above, it considers only one step of oxidation for the BVOCs. Assuming homogeneous mixture, Raoult's law is applied combined with the Pankow theorem (Pankow 1987) resulting in the calculation of the SOA production yield. The scheme is explained in more detail in Pun et al. (2007). The oxidation reactions that are applied for the production of semi-volatile VOCs have been updated in order to take into account more recent (and more detailed) reactions and reaction rates. The values given on the SAPRC site (following Carter, 2010) were used in order to update the reaction rates of the BVOCs mentioned above with OH, O_3 and NO_3. We have also added the SOA-related reactions of BVOCs with different oxidants in the SI (refer to SI-8)."

> **3. "Figures: The captions should be described in detail, and the figures in general could be enlarged, with bigger fonts and legends."**

This suggestion was also made by Reviewer #1. We have enlarged most figures, changed the size of the legends/axis labels in some places. We have also verified the captions and added some explanation to the captions for Figures/Tables when needed.

> **4. "Parts of the manuscript can benefit from rephrasing and grammatical corrections."**

This suggestion has been made by both Reviewers. We have made a thorough rereading of the article to improve its editorial quality.

Specific comments:

1. “L26: Although it is true that the react with OH, NO_3 and O_3, I would not recommend using the phrase “consume” since, for e.g., OH is also formed during the ozonolysis of certain terpenes.”

We have changed consume to “react with”.

2. “L30-32: these two sentences can be combined.”

We have changed the two phrases to the following (lines 30—32):

“BVOC oxidation processes are strongly affected by anthropogenic emissions, especially NO_x availability (e.g. Sartelet et al. (2012); Shrivastava et al. (2019)), and depending to this availability, the associated oxidation pathways may lead to O_3 formation.”

3. “L38: What do the authors mean by “interplay of biogenic with anthropogenic emissions”?”

We meant the interactions between biogenic and anthropogenic emissions. The fact that having both types of emissions in one region complicates our understanding of each type of chemical processes separately. We have changed it to “interaction between biogenic and anthropogenic emissions”.

4. “L49-59: What kind of information do the authors mean when the use the phrase “dispose”? Do the authors mean that the use of simplified schemes hinders the accurate investigation of BVOC and their oxidation products and SOA?” “

The reviewer is correct to point out that “dispose” is the wrong word here. We meant possess. While we have all the information necessary on a global level to simulate OAs, in small and local scales this might not be true since the information for local regions need to be more detailed.

5. “L62: “and to characterization of organic aerosol (to be published)”. I am not sure if this can be stated here since its not published, but I will leave it up to te editor to decide on this.”

We agree with the reviewer that the introduction is not the place to mention unpublished papers. We have removed the phrase in question.

6. “L65: “but also in prior to it and has been linked to BVOC oxidation processes” I don’t understand this sentence.”

We have modified the phrase to the following (lines 64—65):

“Frequent episodes of night-time new particle formation (NPF) have been observed during this campaign as well as during earlier campaigns, and have been linked to BVOC oxidation processes (Kammer et al. (2018); Kammer et al. (2020)).”

7. “L71: “to which extent SOAs in the Landes Forest may impact regional climate and precipitation”. Can be rephrased as “the impact of BSOA from Landes Forest on regional climate and precipitation”.”

Thank you, we have modified the phrase as the reviewer suggests.

8. “L90: “Additionally, 10 air quality monitoring sites”. Aren’t there only 4 air quality monitoring stations in Figure 1?”

It is shown on Figure one that there are 7 stations in Bordeaux itself (with the dashed line pointing to the left side of the figure saying ‘7 stations’). So there are 3 on the south shown with separate dots and then there are 7 on the north of the forest shown with one dot. Since this causes confusion we have modified the image caption as follows (caption of Figure 1) :

> “Measurements and monitoring stations in the Landes region and the high resolution simulation domain (horizontal resolution of 1 km). Black dots represent where the used air quality stations are located, the red circle shows the main measurement site in Salles-Bilos. Note that the northern point located on the city of Bordeaux regroups 7 stations (not shown separately because of their proximity).”

9. “L118: “on a relative scale,”. Relative to what?”

Relative to what the background concentrations are depending on NO_y concentrations. The phrase causing confusion we have removed “on a relative scale”.

10. “L188: “10 families”. Do you mean surrogate species or functional groups?”

Yes, lumped species or surrogate groups. We have modified families to surrogate species.

11. “Figure 2 captions: Which WRF simulations are presented here (referring to SI2 table). It’s not entirely clear from the text as to which WRF simulation (amongst the 8) was finalized for the Base runs?”

This is a fair comment. We have added in the supplementary information good amount of information about the WRF runs that were performed and the one that was chosen in the end, but we have not mentioned that in the caption. The description for the chosen parametrization has been explained in detail in the text of the article as well. We have added the following sentence to the caption of figure 2:

> “More information about all the WRF simulations and the one that was chosen (noted as WRF3) to be presented here has been shown in SI-2.”

12. “L287: DBTopo and DBForet to BDTopo and BDForet.”

We have changed all DBTopo and DBForet to BDTopo and BDForet.

13. “L295: “remote observations”. Do you mean remote sensing observations?”

Yes, it has been modified.

14. “L306: Doesn’t BDForet show quite a drastic decrease in forest cover in a few grids? Is it because BDFOet is using 2016 data set. Also, it should be made clear in the figure or text the why BDTopo and BDForet is using 2019 and 2018 data? Weren’t they updated for the year 2017 and 2016 respectively?”

The reviewer is correct to point this out. We have found an issue in plotting the sub-variables for the BDForet database in the script that was used to produce this figure. While the text in the paper is based on the correct information, the figure ignores to take into account all species of trees in the region when calculating the differences. We have modified the figure to correct the problem with the Bdforet representation. For the second point mentioned by the reviewer we also agree that this is an issue. We had updated the versions of the datasets recently, but we did not modify the text accordingly. In the revised version we have harmonized the text with the Figure.

15. “L323-324: emission factors- are the emission factors that were integrated in CHIMERE are same as the measured emission rates update in MEGAN or? it seems that since MEGAN is integrated in CHIMERE, it should be one and the same.”

The CHIMERE model does not depend on individual tree types, it takes in pre-calculated emission factors for the BVOC species it represents (which have been listed in the article as well as in the response to point 1 of this Reviewer’s remark. These pre-calculated emission factors are provided in global netcdf files with a 300mx300m horizontal resolution giving the total emission factor (of all tree types/plants combined) of each BVOC species. These files are created by the MEGAN model. The way the MEGANv3 model calculates these factors is it uses a global file (which will be called the ecosystem file from here on), in which global land surfaces are divided into smaller areas that have similar vegetation types. This should not be confused with the landcover. The file indicates the vegetation type regardless of what percentage of the land they cover, the latter fact being decided by the landcover files. Let’s consider a non-factual example of a city near the Landes forest. The landcover files indicate that this example city has a low tree cover of around 10%. The ecosystem file will, in this case, indicate that this 10% is going to be 90% pine trees. The basic emission factor of these pine trees is given as inputs to the MEGAN model along with the ecosystem file, the landcover and leaf area index files; the MEGAN model calculates total emission factors for each requested BVOC species. This is then fed to the CHIMERE model as input.

16. “Figure 4: middle panel: shouldn’t it be MEGAN2.04-Theia 2018?”

No, since we updated the landcover in MEGAN as well as in CHIMERE to keep consistency between the models. The middle panel shows the effects of only changing the landcover in MEGAN-v3 but not the tree types, the right-hand side panel changes both.

17. “Figure 5: wasn’t the intermediate grid cell 5x5 km? or why was 11X11 km grid selected?”

The intermediate grid horizontal resolution is indeed 5kmx5km. What is shown here is the results from simulations on a 1kmx1km grid. The solid curve presents the values simulated in the grid cell corresponding to the measurement site. The shaded area depicts a confidence interval to assess the spatial model representativity for the measurement site. It is constructed from simulated values from all neighboring grid cells within a square of 11kmx11km centered at the cell where the measurements were done. This has been explained in lines 428-431.

18. “L363: “This normally causes no issues, since most measurements are performed above canopy level, but since the goal of this work was to understand the atmospheric situation inside the canopy, almost all measurements are performed inside it”. Isn’t this a self-contradictory statement? Do you mean that the meteorological measurement was performed above the canopy and gas/aerosol measurements were performed within the canopy?”

The measurement site houses a tower going above canopy. Meteorological measurements were performed both inside and above the canopy, while the majority of gas and the entirety of aerosol measurements were performed on the ground inside the canopy, at about 6 m height. The other gas measurements having been performed above canopy included PTR-Tof-MS measurements for VOCs that were supposed to make VOC measurements available above and below canopy, but the above canopy measurements were not available to this author. We have added the following sentence in the measurements part of the paper (section 2.1) to be clear about this fact (lines 146 – 147):

> “It should be mentioned that all the measurements used in this work were performed inside canopy, except meteorological parameters, which were measured both inside and above the canopy. For meteorological parameters the above canopy measurements have been used.”

19. “Eq 1: Is “*k*” the von Karman constant?”

Yes, we have added the explanation for k in the text.

20. “L381: it would be useful to see the updated Kz equation after the wind correction factors have been factored in.”

We have added the final Kz equation to section 3.6.

21. “L400: It’s a good idea to be consistent with the variable names. In the manuscript both “z” and ” indicate altitude, which can cause confusion.”

This is a common comment between the two Reviewers. We have homogenized the altitude variable to “z” in all equations.

22. “L414: “The use of correction factors that are specifically calculated for the Landes Forest make their use unrealistic for other forested areas in Europe”.

Is it really the case? For the K_z correction factors in equations 4-6, do not contain any site-specific variables except for maybe canopy height. One would assume that these corrections can be applied to other areas as well. For swrd as well only the LAI and altitude are site specific, which can easily be treated as variables when applied to other European sites or?”

The reviewer is correct, however with a caveat. The extinction factor, the correction factors for wind speed and vertical diffusivity were calculated on a case study specifically for this site according to the reference used (Ogée et al, 2003). This author has not tested this parametrization elsewhere, and it would need to be adjusted for conditions in other forests. It is indeed a good idea to test it in

other forests. We have modified the phrase mentioned by the reviewer to the following to emphasize this fact (lines 438 – 440):

> "The correction factors for diffusivity and wind speed have been calculated for this measurement site according to the work from Ogée et al (2003). While they should be usable in other forests, the specific values should be adjusted to the specific conditions there (canopy height and LAI, especially)."

23. "L432: "an overestimation of friction velocity during these nights". Is this really the case? The modified kz is lower, which would mean that friction velocity is low as well. So, isn't this statement is contradicting the results from figure 5?"

We think that this is not contradictory. We have modified the simulation of the vertical diffusion, while keeping the friction velocity, because it is calculated and measured outside of the canopy. An overestimated friction velocity as shown in Figure 2, 4th panel for the nights between July 4–5 and 16–17 will induce overestimated turbulence which will in fact outweigh the effect decreased turbulence due to the canopy parameterization.

24. "L437: why is m/z 81 important?"

This is a remark made by both reviewers. When looking at mass to charge ratios (m/z) for monoterpenes two main values are taken into account: 137 for α/β-pinene and 81, a terpenoid fragment. As the reviewer correctly points out, 137 is the mass for monoterpenes (taking into account the added proton), while 81 is the mass for one of the major fragments seen in mass spectrum of monoterpenes. This has been seen in multiple studies: Li et al (2020) explores the PTR-Tof-MS measurements for the Landes forest and explains that previous studies have shown that oxygenated terpenoids can fragment inside the PTR instrument and produce a dominant ion at m/z 81. Li et al (2021) working on the Landes forest and the boreal forest in southern Finland makes a similar observation. We have added these two references that explain the fragmentation of terpenoids by PTR-MS.

25. "Figure 7: It will be useful to indicate the changes (%) in relative fractions in the organics."

The figure shows BVOC comparisons, which should be explored on its own before getting into BSOA comparisons. This is because some of the modifications we have made modify directly the BVOCs, therefore it is pertinent to look into the distribution of BVOCs before and after the modifications. We have afterwards explored the changes these modifications make on the OA concentrations.

26. "Figure 10: What is the reason for an increase in isoprene emissions at 03 hr over the ocean in the final case? Is it down to the meteorological changes i.e ECMWF data or?"

Yes, it is because of the meteorological conditions changing, which induces locally enhanced transport of isoprene emitted over land to the ocean. It is nighttime however, therefore the concentrations are quite low.

27. "L577: can be reformulated as "Changing the Kz parameterization and in canopy radiation decrease both OA and BSOA, *aswell* as the concentrations of the three oxidants (OH, NO3 and O3) *participating*in the biogenic OA formation process""

Thank you, that indeed sounds better. We have modified the phrase as the Reviewer has suggested.

28. "L589-590 and L608-610 can be reformulated to make it clear."

The second sentence highlighted here has already been modified following a comment from Reviewer #1. We have modified the phrase to (lines 622 – 624):

> "To do so, two sets of additional simulations have been performed for the base case and the final case, in which the formation of BSOA from each precursor and via each oxidant was followed individually during the model run."

29. "L601-604: Are these results compared anywhere in the manuscript?"

The entire paragraph talks about the comparison of this study to the experimental study cited in the paragraph. We have not explicitly mentioned the results obtained by the reference here, but we have explained that there are differences and we have provided reasons for these differences.

30. "L622: Although the authors mention SOA yields, there is no discussion or values provided here to visualize this result. I would like the authors to show some SOA yields changes, since this work heavily features on simulation of BSOA."

The SOA yields have not been modified in the reactions compared to what is given in Pun et al (2007), Bessagnet et al (2008) and what is presented in the model documentation (https://www.lmd.polytechnique.fr/chimere/docs/CHIMEREdoc_v2020r3.pdf, page 317), we have however updated the reaction rates used specifically for each BVOC and each oxidant from Qin et al (2018). Both of these points have been raised in the modeling chain part. The following section has been added to make sure the SOA scheme is clear (lines 195 – 206):

> "The scheme is based on Odum et al. (1997), a simple two-product scheme with the advantage of being numerically light. In this scheme, it is considered that the oxidation of BVOCs results in formation of semi-volatile products with a yield specific to each BVOC family. Some BVOCs consist of specific species (isoprene, α-pinene, β-pinene), others are surrogate groups consisting of similar species (ocimene, limonene, sesqui-terpenes, etc.). As mentioned above, it considers only one step of oxidation for the BVOCs. Assuming homogeneous mixture, Raoult's law is applied combined with the Pankow theorem (Pankow 1987) resulting in the calculation of the SOA production yield. The scheme is explained in more detail in Pun et al, (2007). The oxidation reactions that are applied for the production of semi-volatile VOCs have been updated in order to take into account more recent (and more detailed) reactions and reaction rates. The values given on the \ cite{saprc} site (following the article Carter, 2010) were used in order to update the reaction rate constants of the BVOCs mentioned above with OH, O_3 and NO_3. The SOA yields have been kept the same as what is provided in Pun et al (2007) and Bessagnet et al. (2008) as well as provided in the CHIMERE documentation. We have also added the reactions of BVOCs with different oxidants in the SI (refer to SI-8)."

31. "L639: what is "local configuration"? Do the authors mean base simulation?"

We meant to say the configuration which is modified with parametrization pertaining to the Landes forest. We understand how that would cause confusion, so we have modified it ‘the final case simulation’.

32. “Figure 14: both the panels could show same time snapshots for better understanding.”

We chose the timesteps where the point we are trying to get across is seen more clearly in them since they are 2 distinct episodes. We would like to keep the figure as it is.

33. “SI-2: What do the numbers in the table indicate?”

They indicate the number allocated to a specific parametrization in the WRF configuration file. They can be found in the WRF documentation. We have added the following phrase in the caption to make this clear:

> “The numbers in the table indicate the parametrization number that is used for a specific parametrization the WRF configuration file; these numbers can be checked by looking the parameter up in the WRF documentation (Wang et al., 2015).”

34. “SI-4: what is PPM2 and PPM3?”

They represent the fine and coarse parts of the aerosols emitted in the anthropogenic emissions inventory. We have added the following phrase to make this clear:

> “For clarity, PPM2 and PPM3 from the EMEP emissions inventory indicate the fine and coarse portions of the emitted aerosols respectively.”

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